

Neptunium - calcite coprecipitation

308
Scientific Notebook # 361

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CNWRA
CONTROLLED
COPY 361

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D.R. Turner

(DR)

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Radionuclide Transport

Initial Entry 8/2/99 by Melissa Nugent

This laboratory notebook chronicles the investigations of radionuclide transport.

This notebook is dedicated to investigating the potential role of Np coprecipitation with calcite as an Np retardation mechanism during radionuclide transport in the alluvium at Yucca Mountain, where approximately 5% calcite is present. The purpose of the experiments documented in this notebook is twofold. First, the amount of Np taken up by calcite as calcite precipitates will be determined. Second, the effect of the variability of important geochemical parameters found in the Yucca Mountain alluvium on Np uptake by calcite will be examined.

MN

8/2/99 M. Nugent
NBS
6/12/00

237 Np Spike #46 (pg. 295, Notebook #031)
NIST Certificate; Catalog # 7237; Source No 678-32-2
Ref. Date 09/01/99

Calcite-neptunium Coprecipitation Experiments
Melissa Nugent, CNWRA

Objective:

To investigate the potential role of Np coprecipitation with calcite as a retarding mechanism for Np during radionuclide transport in the UZ and SZ in the alluvium at Yucca Mountain, where approximately 5% calcite is present. These experiments will determine the amount of neptunium coprecipitated with calcite as overgrowth onto calcite seeds under steady state conditions for growth rate, Ca_{aq} , Np_{aq} , HCO_{3aq} , Pco_2 , and temperature.

Equipment:

1 L reaction vessel and lid
Harvard Apparatus PHD2000 duel syringe pump
Plastic Benton Dickson syringes (30 ml)
Teflon tubing
Plastic ferrules, stopcocks & fittings
pH electrode (Ross combination)
pH meter (Orion 920 A)
ATC probe
pH buffers
Floating stir-bar
Stir-plate
Plastic bottles of various sizes (4 L to 60 ml)
Glass-fritted bubblers
Gas washers

Fish pumps
Weigh boats
Deionized ultra-pure water ("nanopure")
Vacuum filtration set-up
47 mm, 0.4 micron filters
13mm 0.4 micron filters
gelman-type filters
~250ml 10% HCl acid bath
~250 ml nanopure water rinsing bath
3 ml plastic luer-lock syringes
LSA scintillation vials (22 ml glass LSA vial)
Ultima-Gold liquid scintillation cocktail
Pipettors and pipette tips

Chemicals:

Calcite seeds:	Manufacturer: Fisher ACS grade	Lot # 986396	$CaCO_3$
Calcium chloride:	Manufacturer: Fisher ACS	Lot # 913025	$CaCl_2 \cdot 2H_2O$
Sodium Chloride:	Manufacturer: Fisher ACS	Lot # 914913	NaCl
Sodium bicarbonate:	Manufacturer: Fisher ACS	Lot # 963883	
Neptunium: MN 81199	Spike #46	MN 1120199	
1M HCl Conc HCl	Manufacturer: Fisher	Lot # 418110	

These chemicals will be used unless otherwise noted.

Spike #46
diluted to 118 ppm

Experimental Procedure:

1. Several solutions should be prepared in advance of an experiment.

Preparation of 'Equilibrium Solutions':

This solution will be used in the reaction vessel. A solution of 0.1M NaCl in equilibrium with calcite is prepared by measuring out 5.85 grams of NaCl for each liter of nanopure water used. Preparation of a two solutions (A and B) of relatively large volume (4L) is suggested. Excess calcite is then added to solution, the solution is capped and thoroughly shaken, the cap is loosened, the bubblers inserted, and air is bubbled through the solution (using the fish pumps, gas washers, and glass-fritted bubblers) until the pH remains constant. The bottle should be loosely capped to minimize evaporation, and occasionally shaken. This solution will expire after two years.

Preparation of 'Syringe Stock Solutions':

Stock solutions will be fed into the reaction vessel via syringes. Separate calcium chloride and sodium bicarbonate solutions should be prepared. A set of solutions at 0.1 and 0.01M/L for both Ca and HCO_3 , should be prepared. Each solution should be prepared with a 0.1M NaCl background (to eliminate dilution effects on the ionic strength due to syringe addition). Np-Ca syringe solutions are prepared at the start of each experiment by adding Np to aliquots of this solution. These solutions will expire after two years.

2. Preparation

The pH electrode should be conditioned (soak in ~10% HCl for 20 min., change electrode filling solution, and soak in storage solution for 1.5 hours) as necessary (typically ~ every two weeks). The pH meter should be internally calibrated. The electrode should then be attached to the meter and calibrated using the appropriate buffers (7 & 10, in most cases) and the ATC probe.

The 30 ml syringes should be filled, one with Ca solution (without Np), and one with HCO_3 solution. The syringe pump rate is determined by assuming that the moles of Ca added by syringe equal the moles of calcite precipitating onto the calcite seeds. The syringe pump should be set to the desired rate and turned on,

allowing the syringes to drain into a beaker, until the syringes are making good contact with the syringe pump. This minimizes potential erroneous syringe delivery rates associated with start-up.

One liter of Equilibrium Solution is then vacuum-filtered into a clean beaker and transferred to the reaction vessel. The pH electrode and ATC probe will be inserted into the reaction vessel, and stirring and bubbling should begin. The gas washers, fish pumps, and glass-fritted bubblers should be used for bubbling. The pH will rise as the solution Pco_2 adjusts to atmospheric Pco_2 (vacuum filtration causes the solution to de-gas). Aqueous aliquots of Ca and HCO_3 should be added to help bring the solution supersaturation up to a value close to that during precipitation. This also prevents a large pH decrease upon seed addition. The amount of Ca and HCO_3 added must be determined by trial and error, but ~5 ml of both the 0.1 M Ca and the 0.1 M HCO_3 solutions are a good starting point).

3. **Precipitation:** Once the pH is greater than 8.4, the calcite seeds are added, the syringes are attached, and the syringe pump is started. pH and temperature should be monitored and recorded. Once the pH is constant, the desired amount of neptunium is added to the reaction vessel, with a simultaneous, mole-equivalent amount of NaOH. The NaOH is added to prevent a large drop in pH due to the acidity of the Np solution. The pH may decrease or increase, but should rebound to nearly the same value. An aqueous sample should be taken as soon as possible.

4. **Sampling:** Aqueous samples should be withdrawn periodically. All aqueous samples should be at least 4 ml and should be filtered using the gelman-type filter. After each filtration with the gelman-type filter, the gelman filter set-up should be placed in a small acid bath kept nearby. After soaking in acid, the filter should be transferred to a small nanopure water bath. Both of these baths should be changed frequently. The mass of the aqueous sample should be recorded, then the sample pH should be adjusted to 1 with 1M HCl. The mass before and after acid addition should be recorded.

For the remainder of the experiment, the pH, temperature, sampling, and time should be monitored and recorded. Aqueous samples should be taken periodically (such that approximately 7 sample are taken altogether). The experiment should continue until the moles of Ca added equal approximately 10% of the moles of calcite seed added (e.g., for 1 gram of seed (=0.01 moles calcite), 0.001 moles Ca should be added). [NOTE: This limit on the amount of calcite precipitated is placed because, after >~15% is added, the pH begins to decrease. This pH decrease is attributed to an increase in calcite surface area due to crystal growth.]

4. **Calcite Seed Recovery:** The reaction should be stopped (stop syringe pump and detach syringes, stop and remove bubblers, stop stirring), the reaction vessel solution should be vacuum-filtered, and the seed material should be dried in air. Once the calcite seed is dry, it can be scraped from the filter and weighed.

5. Analytical Methods:

Aqueous Calcium analyses will be performed by Atomic Adsorption (AA). Samples will be diluted to an estimated 2.5 ppm. Standards will be made by matrix-matching.

Aqueous Neptunium analyses will be performed using the LSA. A blank that has been matrix-matched to the solution will be run each time.

Calcite seed analyses will be performed by dissolving the samples in an acidic solution and analyzing them by LSA. The amount of seed, strength and volume of acid should be determined based on the minimum detection limits for the LSA.

4

Solution A cont

4/3/2000

3L Nanopure + 8.82g NaCl. Shaken bubbled over night M Nugent
(should be 8.725g) \rightarrow 4/4/2000: Added 8.80g NaCl Br. 0.1m NaCl

5/2/2000

3L nanopure + 8.76g NaCl

5/2/2000

Added an additional 0.0 g to solution. solution
is fine without any additional NaCl.

5

Equilibrium Solution - Solution A

8/2/99 M. Nugent

Prepare Calcite equilibrated water as described on pages 2-3 of
this notebook, controlled copy # 361, CNWRA.

8/2/99:

Set up ~4L deionized water + excess calcite
to equilibrate.

M. Nugent

- bubbling air through solution

- shaking sltn approximately 1x per day for

first 5 days

8/6/99 MN

pH ~ 8.15 - 8.2

8/13/99

M. Nugent

Predicted pH (phreeqc file ccdi.out) : 8.287

@ $p_{CO_2} = 10^{-3.5}$ (note, $\downarrow p_{CO_2}$, \uparrow pH)

8/16/99

M. Nugent

Adjusted solution volume to 4 liters and adjusted MN
solution composition to 0.05m NaCl ~~Fusing Fisher ACS grade~~

$$\frac{0.05m}{L} \times 4L \times \frac{58.5g}{m} = 11.7g$$

~~lot # 984321~~
MN 8/16/99

8/16/99 Actually added: 11.7g

predicted pH (Phreeqc file
Ccnach.out): 8.339

Actual concentration 0.05m

NaCl: Fisher certified ACS
Lot # 914913

9/27/99

M. Nugent

Because solution was open to atmosphere, some may have
evaporated - resulting in higher NaCl concentration.
Solution discarded.

M. Nugent

9/27/99

5pm

MA 9/27/99
~~4L~~ Four (4) liters deionized water + excess
calcite + 0.05m NaCl sltn prepared again
Calcite: lot # 986396 Fisher Scientific
NaCl: lot # 984321, Fisher Scientific
both are Certified ACS.

continued on page 6 then

Cont on
page 4

NaCl: should Add 11.7g Actually added: 11.7g

M Nugent
8/2/99

11/22/99 M. Nugent

Calculate the mass of NaCl required for 3L of 0.1M/L NaCl:

$$\begin{array}{r} 5.85 \\ \times 3 \\ \hline 17.55 \text{ grams} \end{array}$$

~~2/2/99~~
~~9/27/99~~
 M. Nugent

Solution A, con't

M. Nugent
8/2/99

9/27/99 5:25 pm

pH = 9.85 Covered slth and then
Began bubbling air

9/28/99 9:10 pm

pH = ~8.29 (some drift in pH measurement)

This drift is probably because the solution is not stirred. After letting electrode stay in solution longer, pH was steady at ~8.38. This measurement was taken by moving the electrode in solution in quick circles — therefore 'semi-stirred' or slowly stirred. This is how all future measurements of solution will be taken, unless otherwise noted.

9/29/99 11 AM

pH 8.27

10/4/99 4:20 pm

pH = 8.31 pH measurement was slow-drifting

Stopped bubbling Set slth aside

11/22/99

Added 3L H₂O (nanopure)

Added 17.56 grams of NaCl (for 0.1M NaCl.)

See page 4

↓
note: this is 0.1M NaCl

Pages 8 and 9 Are Intentionally
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M Nugent 10/4/99

calculate the mass of NaCl required for a 0.1 M/L NaCl solution -
for 4 liters:

$$0.1 \frac{\text{m}}{\text{L}} \times 58.5 \frac{\text{g}}{\text{m}} \quad 5.85 \text{ g/L} \times 4 \text{ L} = 23.4 \text{ g NaCl for 4 L}$$

Equilibrium Solution Solution B

M Nugent 10/4/99

Follow the same procedure as on page 5 of this notebook, Controlled Copy 361, CNWRA

C - Deionized water - NaCl Stn @ atm. Pw.

~~Made~~ MW 10/4/99 prepared according to procedure, pages 2, 3 of
this notebook, CNWRA controlled copy #361.

10/4/99

Try 0.1 M NaCl

Added - 4 L H₂O

- 23.52 g NaCl actually added

- excess calcite

MW 10/4/99

NaCl: Fisher certified ACS Lot # ~~911913~~ 984321

Calcite: Fisher Scientific ACS Lot # 986396

10/4/99

pH = 9.92 → immediately after adding calcite.

Started bubbling air into bottle.

10/5/99

10³⁰ AM pH = 7.89

1/13/99

Dump out all old stuff (< 1 L) and make new ^{MW 1/13/99} solution

• 4 L nanopure water

• XS calcite

• 23.40 g NaCl

} added to solution B.

4/20/2000

Add 3 L nanopure + ~~FE~~ MW 4/20/2000 17.54 g NaCl.

MW 5/11/2000

Added 3.5 L nanopure, more calcite, and
Added 18.03 g NaCl

Pages 12 and 13 Are Intentionally
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M. Nugent
10/4/99

5 mM Ca Syringe Solution

M. Nugent

Follow procedure described on pages 3-4 of CNWRA controlled copy #361 = this notebook

NaCl:

$$0.05 \frac{\text{m}}{\text{L}} \times 0.5 \text{ L} \times 58.5 \frac{\text{g}}{\text{m}} = 1.4625 \text{ g NaCl per } \frac{1}{2} \text{ L H}_2\text{O}$$

$$\text{CaCl}_2 \cdot 2\text{H}_2\text{O} \quad 0.005 \frac{\text{m}}{\text{L}} \times 0.5 \text{ L} \times 147.02 \frac{\text{g}}{\text{m}} = 0.36755 \text{ g } \frac{1}{2} \text{ L H}_2\text{O}$$

(dihydrate)

CaCl₂: Fisher Scientific certified ACS

Lot # 913025

NaCl: Fisher certified ACS Lot # 984321

5 mmol Ca syringe stock sln

M. Nugent

Follow procedure described on page 3-4 of this notebook, CNWRA 361

10/4/99

10/4/99

 $\frac{1}{2}$ L deionized H₂O:+ 0.3693 g CaCl₂ · 2H₂O

+ 1.4625 g NaCl.

0.005 M Ca

0.05 M NaCl

therefore, solution is 0.05 M Na, ~~0.05 M Ca~~,
and 0.00502 M Ca

~~10/4/99~~ ^{MN} 10/9/99

10/9/99 Added 1.4620 g NaCl to solution.

Therefore solution is 0.09998 M Na

10/4/99
M Nugent

10mmol Ca Syringe Stock solution

12/6/99

M Nugent

Follow procedure described on p34 of this notebook, correct controlled copy #361

12/6/99

0.01M Ca, 500ml, 0.1M NaCl.

CaCl₂·2H₂O:
 $147.9 \text{ g/mol} \times 0.01 \text{ M/L} \times 0.5 \text{ L} = 0.735 \text{ g}$ Fisher lot 913025 ACS grade

NaCl:

 $58.5 \text{ g/mol} \times 0.1 \text{ M/L} \times 0.5 \text{ L} = 2.925 \text{ g}$ Fisher ACS grade lot: 981321

Actually added:

500.52 g nanopure water.

2.94 g NaCl

0.7356 g CaCl₂·2H₂O

Therefore, solution is 0.1004M Na and 0.009996M Ca.

12/6/99 M. Nugent

10 mmol HCO_3^- syringe

12/6/99

Stock solution

M. Nugent

Follow procedure described on page ~~36~~ ^{MN 12/6/99} 3, 4 of this notebook, CNWKA controlled copy 761.

12/6/99

 NaHCO_3 NaCl 0.01 M NaHCO_3 , 0.1 M NaCl , 0.5 L: $84.9 \text{ g/mol} \times 0.01 \text{ mol/L} \times 0.5 \text{ L} = 0.42 \text{ g}$ Fisher ACS grade Lot # 963883 $58.5 \text{ g/mol} \times 0.1 \text{ mol/L} \times 0.5 \text{ L} = 2.925 \text{ g}$ Fisher ACS grade Lot # 984321

Actually added:

503.19 g nanopure water

0.4212 g NaHCO_3 2.9253 g NaCl Therefore, solution is 0.009965 M HCO_3^- and 0.0994 M Na^+

11/19/99 M. Nugent

0.1m Ca solution

11/19/99

M. Nugent

Follow procedure described on pages 3, 4 of this notebook, controlled copy #361, CNWRA.

11/19/99

100ml of 0.1m Ca solution:

$$147 \text{ g/mole} \times 0.1 \text{ m/L} \times 0.1 \text{ L} = 1.47 \text{ g/100ml}$$
 using Fisher grade ACS $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$
 Lot # 913025

100ml nanopure water

1.4703g $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ added**DISCARD THIS SOLUTION!**

because an unknown amount has been used
 and the solution needs to be adjusted to 0.1m NaCl
 (to account for dilution effects from syringe addition).

12/6/99 M. Nugent

500ml of 0.1m Ca solution with 0.1m NaCl to be prepared:

Ca: $147 \text{ g/m} \times 0.1 \text{ m/L} \times 0.5 \text{ L} = 7.35 \text{ g } \text{CaCl}_2 \cdot 2\text{H}_2\text{O}$
 using Fisher Lot # 913025

NaCl: $58.5 \text{ g/m} \times 0.1 \text{ m/L} \times 0.5 \text{ L} = 2.925 \text{ g}$

Actually added: (using Fisher ACS Lot # 913025 for CaCl_2
 503.00g nanopure water and Fisher ACS grade NaCl, Lot
 7.36g $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ # 984321)
 2.9262g NaCl

Therefore, solution is 0.0995m Ca and 0.0994m Na

0.1m HClO_3 Solution

M. Nugent

Follow procedure on pages 3-4 of this notebook, CMWRA controlled copy # 361

11/19/99

0.1m NaHClO_3 , 100ml of solution:

$$849\text{m} \times 0.1\text{M/L} \times 0.1\text{L} = 0.849\text{g}/100\text{ml}$$

using Fisher ACS grade NaHClO_3 , Lot # 963883

Actually Added:

100ml by volumetric flask

0.849g

0.8409g NaHClO_3

DISCARD SOLUTION

because an unknown amount has been used and
The solution needs to be adjusted to 0.1m NaCl .

12/6/99

0.1m NaHClO_3 , 500ml, 0.1m NaCl : NaHClO_3

$$849\text{m} \times 0.5\text{L} \times 0.1\text{M/L} = 4.2\text{g } \text{HClO}_3^- \text{ using Fisher ACS lot 963883}$$

 NaCl

$$58.59\text{m} \times 0.1\text{M/L} \times 0.5\text{L} = 2.925\text{g } \text{NaCl} \text{ using Fisher ACS Lot \#}$$

Actually Added:

500.14g nanopure water

4.22g NaHClO_3 2.9319g NaCl Therefore, Solution is 0.1004m HClO_3^- and 0.1002m Na .

Preparation of Blanks for LSA

M. Nugent

Several blanks will be prepared for LSA analysis.

1. "Np 0.12g Blank": 8/27/2000

for seed analysis [for Np]: 0.1458g calcite seed (unreacted
calcite seed which presumably contains no Np) + 3.75 ml
of 0.1 N HCl. Total weight, after dissolution: 3.8662g.

Also described on page 78 of this notebook, CMWRA
controlled copy # 361.

LSA blanks will be described chronologically in this
notebook, instead of listing them on this page. Therefore, no
further entries will be made on this page. M. Nugent.

Pages 24 Through 30 Are Intentionally
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Test Run.

Date 11/18/99

Melissa Nugent

Objective:

Precipitate Calcite without adding Neptunium. Try to get steady state. Refer to procedure in this notebook CNWRA Controlled copy # 361, page 2. No samples will be collected.

Using:

- 1 Liter filtered, solution (0.1M NaCl) in equilibrium with calcite (solution A).
- ~1 gram of seed calcite
- add 1 bubbler (if 1 bubbler is not sufficient, add another)
- choose precipitation rate of $\sim \log r = -3.5$ moles/m²/hr.
- use 0.01M Ca syringe and 0.01M H₂O₃ syringe.

Rate Calculation: (note, numbers are approximate).

for 0.01M/L Ca syringe, 0.2 ml/min, assuming 0.6 m²/g surface area, assume 1 mole Ca added = 1 mole CaCO₃ precipitated.

$$r = \frac{0.01 \text{ M}}{\text{L}} \times \frac{0.2 \text{ ml}}{\text{min}} \times \frac{1 \text{ L}}{1000 \text{ ml}} \times \frac{60 \text{ min}}{\text{hr}} \times \frac{1 \text{ g}}{0.6 \text{ m}^2} \times \frac{1}{1 \text{ g}} = 2 \times 10^{-4} \frac{\text{mol}}{\text{m}^2 \cdot \text{hr}}$$

$$\# \log r = -3.699$$

estimated surface area of calcite - surface

area analyses are presently being done.

M Nugent

11/18/2000

Time:

Measure pH upon addition of calcite seed:

Calibrate electrode with 7, 10 buffers

10:25 Am pH = 8.28 electrode response is poor. Electrode slowly drifts to value.

$T_{\text{solution}} = 22.8^\circ\text{C}$

10:28 8.30

10:40 am 8.31 Then added seed material

pH \rightarrow 8.32 immediately

Start syringes at 0.2 mL/min, 0.1 M concentrations

10:42 8.33

10:45 8.31

10:50 8.31 / 8.30

10:59 8.31

11:08 8.32 Attached a second air bubbler

11:09 8.32 $T_{\text{solution}} = 23.1^\circ\text{C}$ ^{M.N. 11/19/99}

11:19 8.31

11:29 8.31

11:49 8.31 / 8.30

12:20 8.30

12:43 8.28 / 8.29 but very 'drifty'!

12:45 8.30 $T_{\text{solution}} = 23.4^\circ\text{C}$

1:30 8.30

2:19 8.30 / 8.31

Stop reaction

It seems the pH electrode is not responding well. Will re-condition electrode immediately before the next run to see if electrode will respond better.

Test Run - No NP.

Date: 11/19/99

M Nugent

Repeat Test run of 11/18/99, except:

- condition electrode and use

fresh buffers.

2.0061 grams Calcite Seed.

- use ~2g of seed calcite (to make pH response upon addition of calcite more obvious)

- use Syringe addition rate = 2x that of 11/18 b/c using 2x the seed material.

- add Ca and HCO_3 to solution immediately before seed calcite, to supersaturate the solution and decrease any transient to steady state.

- use 2 bubblers the whole time.

- don't collect/analyze samples - just observe pH. ^{M.N. 11/19/99}

Rate calculation:

$$r = 0.01 \text{ M} \times 0.4 \text{ mL} \times \frac{60 \text{ min}}{1000 \text{ mL}} \times \frac{1}{0.6 \text{ M/g}} = 2 \times 10^{-4} \frac{\text{mol}}{\text{m}^3 \cdot \text{hr}}$$

$$\log r = -3.699$$

Objective:

to repeat the test ^{M.N.} 11/19/99 test run performed on 11/18/99 and recorded on pages 31 and 32 of this notebook (CNWRA controlled copy #361) - however, the electrode will be conditioned for better pH readings, and 2 air bubblers will be used to control pCO_2 . Also, the amount of calcite seed used will be doubled (and so will all relevant parameters) to increase the observer's sensitivity to controls or influences on pH measurement.

M Nugent
11/19/99

Condition Electrode:

- Acid soak (10% HCl) 20 minutes.
- Change filling solution
- Change storage solution
- Allow electrode to soak 1.5 hrs in storage solution.

- Filter 1 L of CC-DI solution (using solution A).
- Transfer 1L to reaction vessel, begin stirring and bubbling.
- Use fresh 7, 10 buffer solution to calibrate pH electrode

Time: Notes:

3:38pm pH = 8.31/8.30

3:50 8.30

3:54 pH = 8.30 Then, add 2 ml of 0.1M Ca
pH → 8.294:00 pH = 8.30 Then, add 2 ml 0.1M HCl
pH → 8.28

4:03 8.31

4:06 8.31

4:06:30 8.31 Then, added seed material
pH → 8.28

4:07 8.28

4:09 8.28

4:13 8.28

4:16 8.28 T solution = 24.0 °C

4:24 8.27

4:24:30 Add 2ml 0.1M Ca → 8.26 } should have
4:25 Add 2ml 0.1M HCl → 8.25 } added this
before seed

4:26 8.24

4:28 8.25

4:31 8.21

4:33 8.21

4:39 8.20/8.19

11/19/99

M Nugent

4:50 8.19

4:51 8.19

5:10 8.19 ⇒ 24.463ml added from each syringe
STOP

Note: syringe solutions are not adjusted to 0.1M NaCl
(to account for dilution) + any other dilution effects. Syringe
solutions will be adjusted.

M.N. 11/19/99

The use of 2 bubblers helped stabilize pH - presumably
by controlling pCO₂ (@ atmospheric pCO₂) more effectively,
than one bubbler. Will continue to use 2 bubblers
in future experiments.

I will use 1 gram of calcite seed in experiments
in the future - 2 grams were used to 'exaggerate' the effect
of pH change upon seed addition. However, doubling the
amount of seed material doubles the # moles Np that would have
to be added, and that is not necessary. To conserve Np,
1 gram of calcite seeds will be used. The purpose of 2 grams - to
see how much the pH dropped upon seed addition - has been met.

36

M Nugent

Test Run - No Np.

Date: 11/22/99

Repeat 11/19/99 but use only ~1 gram of seed
and syringe infuse rate of 0.2 ml/min.

Objective: to see if calcite precipitation is steady-state
under the following conditions:

- No Np
- 1g calcite seeds
- 2 bubblers
- Syringe infuse rate of 0.2 ml/min
- Syringe composition: 0.1M Ca in one syringe, 0.1M HCO_3 in other
- 1 Liter of filtered, 0.1M Na 0.1M Cl water in
equilibrium with calcite
- monitoring temperature

11/22/99 37

Calcite Precipitation - Test Run

M Nugent

- Reconditioned electrode \pm calibrated with fresh 7, 10 buffers. 11/22/99
- filtered 1L of solution A

set solution to stir and bubble (air).

Objective: to precipitate calcite at steady state according to the
procedure described on pages 2 and 3 of this notebook, CNWRA controlled copy 36L
and the conditions described above.

Time: Added 3ml 0.1M Ca and 3ml 0.1M HCO_3 .

2:04 pm pH = 8.42 T solution = 25.2°C

2:07 pm pH = 8.42 Then, added seed calcite (1.0077g)
and attached syringes (0.1M concentrations) at
0.2 ml/min.

2:08 pm 8.39

2:09 8.35

2:10 8.33

2:15 8.28

2:17 8.26

2:22 8.25

2:26 8.26

2:38 8.27

2:46 8.27

2:57 8.27

3:19 8.27 T solution = 25.5°C

3:30 8.27

4:25 8.26/7 Stop syringe addition \pm refill syringes.
T solution = 25.6°C

4:31 8.27 Started syringes again.

5:07 pH = 8.27

5:55 pH = 8.27

6:38 pH = 8.27

STOP.

pH seems steady!
electrode performing well.

Ready 11/22/99

Ready to add Np.

12/9/99

Melissa
NugentTest Run ^{with MW 12/9/99} ~~NP~~ Np.

Try:

1g seed, 2 bubblers

logr ~ -3.5

try to balance ^{moles} acid and base (simulate Np addition to solution)

try sampling

use syringe solutions which have been

NaCl adjusted.

recondition electrode: use fresh buffers.

use solution B

Objective: precipitate calcite at steady state in the presence of Neptunium, at an unchanged Np concentration. Refer to the procedures and equipment described on Pages 2 and 3 of this notebook, CNWRA Controlled copy #361. Aqueous samples will be collected.

Set syringe rate: approximate!

$$10^{-3.5} \frac{\text{moles Ca}}{\text{m}^2 \cdot \text{hr}} \times \frac{0.6 \text{ m}^2}{\text{g}} \times 1 \text{ g} \times \frac{1 \text{ hr}}{60 \text{ min}} \times \frac{L}{0.01 \text{ m}} \times \frac{1000 \text{ ml}}{L} = \frac{0.316 \text{ ml}}{\text{min}}$$

with 0.1 mL syringes

with 0.1 mL Syringe 0.0316 mL/min

need to add ~ 0.1g CaCO₃ = 0.001 moles Ca

$$0.001 \text{ m Ca} = \left(\frac{0.01 \text{ m}}{L} \right) \times V \quad V = 0.1 \text{ L} \sim 100 \text{ mL}$$

with 0.1 mL syringe, ~10 mL.

∴ choose 0.1 mL syringe. No, see p. 39 of this notebook. Use 0.01 mL syringes.

MW 12/9/99

12/9/99

Note: Calcite surface area determination (see page 169, this Notebook)

1 - 0.246 m²/g

OUT gassed 720 min

2 - 0.257 m²/g

3 - 0.257

average 0.253 m²/g

for logr = -3.5:

$$10^{-3.5} \times 0.253 \frac{\text{m}^2}{\text{g}} \times 1 \text{ g} \times \frac{1 \text{ hr}}{60 \text{ min}} \times \frac{L}{0.01 \text{ m}} \times \frac{1000 \text{ ml}}{L} = 0.1333 \frac{\text{ml}}{\text{min}}$$

with 0.1 mL syringe, 0.0133 mL/min.

Mass of calcite added to reaction vessel:

1.0011 g Calcite

calibrate electrode

T probe 66.8 °C cal to 6.99 and 9.78

Filter 1 L of solution B.

200 pm

Stirring, bubbling (2 bubblers) (air)

T 66.9 °C

204

T sth = 66.2 °C (in reaction vessel)

pH 8.02 and climbing

calibrate electrode with temperature probe in reaction vessel.

cal as 6.99 and 9.78 at 67.8 °C

212

pH = 8.16

215

8.18/8.19

224

8.19

230

8.19/8.20 Added 3 mL 0.1 M Ca → 8.18

233

2 mL NaHCO₃ 0.1 M added 8.23

238

8.24 4 mL 0.1 M H₂O₃ added → 8.28

240

8.27 MW 12/9/99 4 mL 0.1 M Ca added → 8.27

Hopefully these Ca and H₂O₃ additions will prevent a large dip in pH upon calcite seed addition

note: T probe should be saying 68 °F?? Will do this run and figure out y later.

12/9/99

- 242_{pm} 8.27 Sample 12999-1 collected.
 246 pH = 8.28
 T solution = 67.8°C
- 248 8.28 Added seed → 8.27 ^{mmagent 12/9/99}
- 249 8.26 Started Syringes ~~at 0.1333 ml/min~~
 at 0.1333 ml/min, 0.01 m sltns (NaCl adjusted)
- 256 8.25
- 251 8.24 Sample 12999-2 collected: FILTERED
 used 0.2 µm dynagard syringe tip filter.
- 254 8.24
- 258 8.22 T solution = 67.9°C
- 317 pH = 8.28 ^{mmagent 12/9/99}
 T sltn = 68.6°C
- 322 pH = 8.22
 Added 250 µL of 118 ppm Np sltn and 250 µL of 0.32 M
 NaOH simultaneously
 pH = 8.37 and then dropping
- 322 1/2 8.29
- 323 8.22
- 325 8.19 Sample 12999-3 filtered
- 327 8.21
- 333 8.21/2
- 334 8.23
- 350 8.24 T sltn = 69.2
- 356 8.25
- 408 8.24 Sample 12999-4
 filtered
- 410 8.25 T sltn = 69.3°C 24.5°C
- 447 8.24 69.6°
 Re-calibrate Buffers at 68.6°C
- 450 8.25/8.26
 Will keep buffers next to rxn vessel so
 buffer T = reaction vessel T.
- 507 8.25 sample 12999-5 filtered → note: H₂O₂ syringe is
 dripping again.

12/9/99

Sample Vial Weights.

Name:	Mass:	note: mass is bottle + cap (NO SAMPLE)	
12999-1	16.9509		
12999-2	17.1345		
12999-3	16.5687		
12999-4	16.8459		
12999-5	16.8985		
12999-6	16.8715		
12999-7	16.9653	not used. mmagent 12/9/99	

Sample Weights

Mass = mass of bottle + cap + sample

Name: → Mass:

12999-1	20.7466
12999-2	23.1225
12999-3	22.9846
12999-4	23.1795 ^{mmagent 12/9/99} 23.1795
12999-5	23.2418
12999-6	23.2758

Mass of Acid Added

Name:	Mass Acid:
12999-1	0.2507g
12999-2	0.2514
12999-3	0.2518
12999-4	0.2520
12999-5	0.2523
12999-6	0.2526

Type of acid: 1M HNO₃

Sample Name	vial+cap mass	vial+cap+ sample mass	sample mass	acid 1M HNO ₃ mass	Total Mass	Total sample mass
12999-1	16.9509	20.7466	3.7957	0.2507	20.9973	4.0464
12999-2	17.1345	23.1225	5.9880	0.2514	23.3739	6.2394
12999-3	16.5687	22.9846	6.4159	0.2518	23.2364	6.6677
12999-4	16.8459	23.1795	6.3336	0.252	23.4315	6.5856
12999-5	16.8985	23.2418	6.3433	0.2523	23.4941	6.5956
12999-6	16.8715	23.2758	6.4043	0.2526	23.5284	6.6569

12/9/99

528 pm pH = 8.24/8.25 T solution = 69.4°C 25.5°F

529 Stop syringes, detach them, and
refill them. 12/9/99 ↑ calibrated electrode21.201 ml have been added (21.201)
from each syringe thus far.

531 START new syringes

533 pH = 8.24

546 ~~534~~ pH = 8.23/8.24
M. Nugent 12/9/99← could lack of H₂O₃delivery done pH ↓ (same
but not all H₂O₃ gets in).

624 pH = 8.25

62.106? M. Nugent 12/9/99

Sample 129996 filtered

628 STOP EXP. 8-25

Additional 7.5188 ml added from each
Syringe.

Filter solution and save calcite.

Mass Recovered: ~~0.8689~~ 0.8699g
Recovered on 12/10/99 by M. NugentNote, not a quantitative recovery. Quantitative recoveries are
difficult to achieve in this case because the filter
slipped during filtration.Note: Temperature readings are not accurate. An ATC
probe will be used in all experiments after this one.

Analysis of 12/9/99 Experiment.

12/9/99
M. Nugent

Dilutions - for Ca analysis by A.A.

12/13/99

AA Samples
Sample
NameSmpl

Dilns-Ca
~2.5ppm Ca
vol for 7ml

Date Diluted

Mass
Bottle+cap
grams

Actually added:

Ca (reaction
(solution)
gramsH₂O (7)
H₂O+sample
Total mass:

AA Samples	Smpl #	Dilns-Ca
12999-1	Ca1	0.4366
12999-2	Ca2	0.4366
12999-3	Ca3	0.4366
12999-4	Ca4	0.4366
12999-5	Ca5	0.4366
12999-6	Ca6	0.4366

Date Diluted	Mass	Ca (reaction)	H ₂ O (7)	Total mass:
12/13/99	4.0590	0.4962	7.0592	
"	4.0424	0.4985	6.9920	
"	4.0609	0.4994	7.0039	
"	4.0651	0.4979	7.1128	
"	4.0800	0.4976	7.0447	
"	4.0679	0.4990	7.0045	

M. Nugent

AA analysis of aqueous samples collected during the experiment:

AA measurements (ppm)

Date performed: 12/14/1999

Performed by M. Nugent

12/14	AA measurements (ppm)
Ca1	3.914
Ca2	4.230
Ca3	4.189
Ca4	4.215
Ca5	4.208
Ca6	4.065

Using standards described on
pages 158-159 of this notebook,
CNWRA Controlled Copy # 361.

Calcium, ppm, of diluted solution

millimole/L

	12/14 (ppm)	12/14 (mmol/L)
Ca1	55.68	1.39
2	59.38	1.48
3	58.75	1.47
4	60.21	1.50
5	59.57	1.49
6	57.06	1.42

ppm
mmol/L
Ca concentrations for reaction vessel solution

12/9/99

Np analyses by LST

Try a 2ml sample and a 3ml sample (both taken from the original sample) to ^{MN 12/14/99} ~~see~~ check for the least volume of sample which will yield good results.

Add 10ml of AB cocktail to the 2ml sample and 75ml of the AB cocktail to the 3ml sample.

made 12/14/99 →

Sample #	New Sample #	Sample Mass (g)	New Sample #	Sample Mass (g)
12999-1	xx	xx	xx	xx
12999-2	xx	xx	xx	xx
12999-3	Np3a	2.0505	Np3b	3.0002
12999-4	Np4a	1.9995	Np4b	3.0009
12999-5	Np5a	2.0005	Np5b	3.0044
12999-6	Np6a	2.0021	Np6b	3.0105

Blank

15 ml cocktail + ^{MN 12/14/99} ~~3.0284g~~ of solution B +
10 mL of 1M HNO₃

SYSTEM NORMALIZED

C14 IPA DATA PROCESSED - 16-Dec-1999 14:26

C14 Eff (0-156 keV) = 96.49 %

C14 CHI SQUARE IPA DATA PROCESSED - 16-Dec-1999 14:37

C14 Chi Square = 25.69

H3 IPA DATA PROCESSED - 16-Dec-1999 14:38

H3 Eff (0-18.6 keV) = 64.97 %

WARNING: Questionable H3 Efficiency value - Please rerun quench curves & view historic data

H3 CHI SQUARE IPA DATA PROCESSED - 16-Dec-1999 14:49

H3 Chi Square = 30.61

BKG IPA DATA PROCESSED - 16-Dec-1999 15:49

Bkg (0-18.6 keV) = 17.57 cpm

Bkg (0-156 keV) = 25.07 cpm

C14 E²/B (1-156 keV) = 488.32H3 E²/B (1-18.6 keV) = 240.0117 Dec 1999 08:38
Protocol #28ALPHA/BETA - 1.09
NP-237 10(-7)Page 81
User: MIKE ALMENDAREZ

Time: 999.00

Data Mode: Alpha/Beta

Nuclide: NP/PA

Discriminator: 126

Background Subtract: 1st Vial

	LL	UL	LCR	2SX	BKG
Beta A:	0.0 - 400	0	0.1	8.02	
Beta B:	100 - 400	0	0.5	0.13	
Alpha:	100 - 400	0	5.0	7.65	

Quench Indicator: SIS

2x 2x ALPHA COUNT REGION

Luminescence Correction On

Low Level Count Mode On

Coincidence Time(ns): 18

Delay Before Burst(ns): Normal

Protocol Data Filename: PROT.DAT

Count Data Filename: c:\prot28\data28.001

Spectrum Data Drive & Path: C:\PROT28

S#	TIME	CPMA	CPMB	CPMa	a:2SX	SIS FLAG
1	999.00	8.02	0.13	7.649	2.288 50.469	*B Blank
2	16.31	2.47	0.02	90.573	5.422 49.764	*Np3a
3	27.07	0.00	0.00	51.457	5.753 18.506	*Np4a
4	65.39	0.00	0.00	16.820	7.348 0.000	*Np5a
5	MISSING TUBE(S)					[Np6a]
6	10.90	5.87	0.00	139.140	5.276 52.836	*Np3b
7	18.73	2.12	0.00	77.776	5.496 68.071	*Np4b
8	52.26	2.41	0.59	23.006	6.701 190.04	*Np5b
9	999.00	4.02	2.19	0.000	0.000 445.20	*Np6b
10	999.00	0.00	0.00	0.000	0.000 0.000	

SYSTEM NORMALIZED

C14 IPA DATA PROCESSED - 18-Dec-1999 23:31

C14 Eff (0-156 keV) = 96.59 %

C14 CHI SQUARE IPA DATA PROCESSED - 18-Dec-1999 23:42

C14 Chi Square = 11.55

H3 IPA DATA PROCESSED - 18-Dec-1999 23:43

H3 Eff (0-18.6 keV) = 64.88 %

WARNING: Questionable H3 Efficiency value - Please rerun quench curves & view historic data

WARNING: Questionable H3 Chi square value - Please view historic data

Note: 12/16 LST results are suspect due to problems w/ LST - 3/17/2000 LST results are collected after LST problems resolved. Use 4/17/2000 LST results - see page 72, This notebook.
3/17/2000 (MN 4/1/2000)

12/9/99

M. Nugent

Converting LSA Np data into ppb Np
 $^{237}\text{Np} = 237.041678 \text{ g/m}$
 half life: 2.14×10^6 years

Name	Mass g	counts	efficiency corr	# atoms	moles	m/l	ppb
np3a	2.0505	90.573	90.573	$1.47\text{E}+14$	$2.441\text{E}-10$	$1.191\text{E}-07$	28.22
np4a	1.9995	51.457	51.457	$8.35\text{E}+13$	$1.387\text{E}-10$	$6.936\text{E}-08$	16.44
np5a	2.0005	16.82	16.820	$2.73\text{E}+13$	$4.533\text{E}-11$	$2.266\text{E}-08$	5.37
np3b	3.0002	139.14	139.140	$2.26\text{E}+14$	$3.75\text{E}-10$	$1.250\text{E}-07$	29.63
np4b	3.0009	77.776	77.776	$1.26\text{E}+14$	$2.096\text{E}-10$	$6.985\text{E}-08$	16.56
np5b	3.0044	23.006	23.006	$3.73\text{E}+13$	$6.201\text{E}-11$	$2.064\text{E}-08$	4.89
np6b	3.0105	0	0.000	0	0	0	0

ppb Np, average:
 np3 28.93
 np4 16.50
 np5 5.13
 np6 not detected.

Comments:

Np concentration is clearly decreasing - need to test if this is all due to coprecipitation or if it is in part due to adsorption onto reaction vessel. Therefore, the experiment will be repeated with no calcite added - sampling through time to determine Np adsorption.

Can seeds be analyzed by LSA?

Assume that Np lost ($28.93 - 5.13 = 23.8$ ppb) is captured by seed mat'l.

- Total g calcite precipitated = $\frac{\text{grams}_{\text{CC, int}}}{\text{MW}_{12/16/99}} + \frac{\text{grams}_{\text{CC, added}}}{\text{MW}_{12/16/99}}$, assuming all Ca added precipitates as CC

$$(7.5188 + 21.201) \text{ ml added} = 28.7198 \text{ ml of Ca syringe added}$$

$$28.7198 \text{ ml} \times 0.01 \text{ M} = 2.87198 \times 10^{-4} \text{ moles Ca} = \text{moles calcite} = 2.87198 \times 10^{-4} \text{ g calcite}$$

$$\frac{1000 \text{ mg/L}}{1000 \text{ mg/L}} \times \text{Total moles Calcite, final} = \frac{1.0011 \text{ g}}{\text{MW}_{12/16/99}} + 0.0287198 = 1.0298 \text{ g}$$

- Convert Np lost to $\frac{\text{moles}_{\text{Np}}}{\text{MW}_{12/16/99}}$: $23 \text{ ppb} \times \frac{1}{1000 \text{ ppb}} \times \frac{1}{237.041678 \text{ g/m}} = 1.00405 \times 10^{-7} \text{ moles in 1 L sltn.}$

assume all this neptunium is in 1.0298 g. calcite

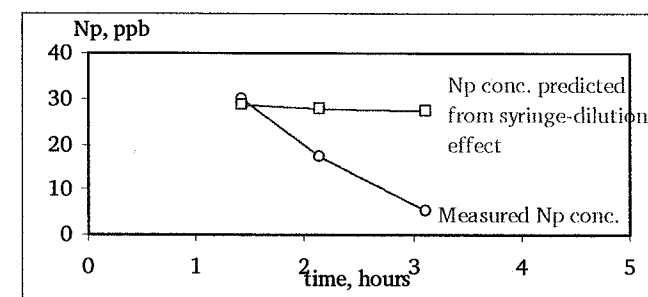
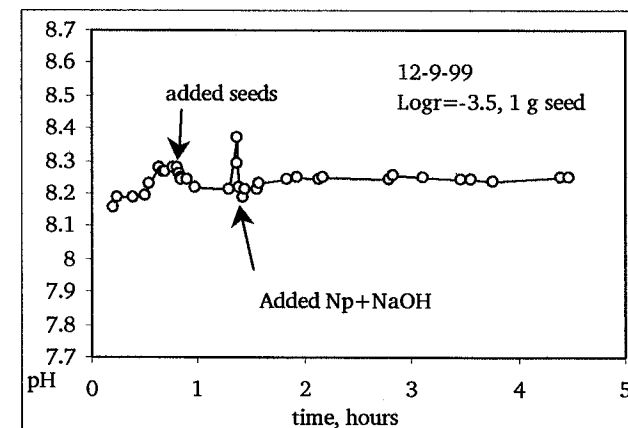
- Dissolve 0.4g Seed: $0.4 \text{ g} = 30.82\% \text{ of final calcite } (0.4 / 1.0298)$. Therefore 30.82% of moles of Np in 0.4g calcite seed: $0.3082 \times 1.00405 \times 10^{-7} = 3.09399 \times 10^{-8} \text{ moles Np.}$

- Dissolve 0.4g seed in 10 ml of acid: $3.094 \times 10^{-8} \text{ M} = 3.094 \times 10^{-6} \text{ m/L} = 0.733 \text{ ppm Np}$

this should be sufficient for LSA analysis

12/9/99

M. Nugent



Dissolution

of calcite seed for LSA analysis for Np.:

Bottle + Cap: 16.6123g

0.1399g seed added to bottle, 3.5 ml 1.0N HCl added -

Total mass after dissolution = 3.6112g

This sample was analyzed by LSA.

LSA sample prepared on 12/10/99 by M. Nugent

Sample Name: S1A

Comments:

- This is a trial run - temperature readings were inaccurate. Will use ATC probe
- need to check how much Np is adsorbed to surface of experimental equipment before concluding that the observed loss of Np is due to uptake.

12/9/99
MNugent

UK
MNA

3/31/00 9:24:58 AM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 1
User: Nugent

Assay Definition-
Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

LST results - seed analysis
for 12/9/99

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00
Quench Set:
Count Time (min): 999.99
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off
Regions Half Life Units Reference Date Reference Time
Beta A
Beta B
Alpha

Cycle 1 Results	S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMA	alpha2S%	SIS	MESSAGES
1	999.99	37.21	1.04	45.64	0.94	1.58	5.03	897.87	Blank B	
2	0.69	5964.24	3.13	5965.95	3.13	6517.26	2.98	230.37	S1A	
3	0.55	9768.24	2.73	9774.36	2.73	8174.60	2.98	232.68	S2	

MNA

MNA

12/30/99

M Nugent

12/30/99
MNugent

Neptunium-Calcite Coprecipitation

Objective: To see how much Np is adsorbed onto experimental set-up (reaction vessel, bubblers, etc)

Try to duplicate experiment of 12/9/99 except:

- no seed, no syringe addition, no filtering samples
- measure pH, bubble air, use filtered solution b.
- sample at ~20 minute intervals.
- add 250µL of 118ppm Np seth + equivalent base.
- analyse samples for Ca and Np.

Note: because the presence of calcite seed mat'l may influence

Np-adsorption onto reaction vessel, an adsorption experiment may have to be performed in the presence of calcite seeds

(@equilibrium w.r.t. calcite, not during growth!) - if the adsorption without seeds is significant. However, first adsorption w/o calcite will be performed.

- add acid/base to keep pH ~ 8.24

-reconditioned electrode, made fresh 7, 10 buffers.

-filtered 1L of seth.

-using acid washed glassware (this is typical).

Time:

145 calibrated electrode

ATC probe 85.8°C (temp probe)

Calibrated to 7.04 and 9.86 w/ 7, 10 buffers.

Not sure why temp probe is reading 85.8°C when it is 24.5°C. Replaced temp probe, recalibrated to 7.00 and 10.01. Reads 25.2°C

149 stirring pH = 7.60 (drifting due to pCO2 adjustment)

Begin bubbling

152 7.39 & climbing

153 7.52 sample (not filtered) 123099-1

for Ca analysis only.

155 7.73 Add 3mL 0.1M Ca 7.76

155
MN
12/30/99

/ / / / /

12/30/99 M. Nugent

MN 12/30/99

Time: 1:56 pm 7.83 Add 4ml 0.1m ~~NaHCO₃~~ Ca → 7.84
 157 7.89 Add 7ml 0.1m NaHCO₃ → 8.24
 200 pH = 8.24
 * 202 8.26 Sample 123099-2 for Ca only.
 204 8.27 Tetc probe = 25.1 °C
 206 8.28
 ↓ 20700 8.28 → 8.41 upon addition of 250 µL 118 ppm NP
 20710→25 8.26 removed ~9ml 123099-3 and 250 µL 0.32m NaOH
 209 pH = 8.27, 25.2 °C
 * 220:20→2:20 35 8.33 Sample 123099-4
 221 8.34 add 2x 0.25ml 0.01m HNO₃ → 8.35
 224 8.35 5x 0.25ml 0.01m HCl → 8.30/8.29
 Tetc probe = 25.4 °C
 239 8.35 Add HCl 7x 0.25ml 0.01m HCl → 8.28
 * 241 8.28 Sample 123099-5
 246 pH = 8.31 5x 0.25ml 0.01m HCl → 8.27/8.26
 304 8.34/5
 * 305 8.35 Sample 123099-6
 306 8x 0.25ml 0.01m HCl → 8.27
 327 pH = 8.35
 329 8.35 1ml x 0.01m HCl → 8.31
 329 1ml x 0.01m HCl → 8.26
 * 333 pH = 8.28 Sample ~~123099-7~~ MN 12/30/99 Sample 123099-7
 335 8.29
 344 8.32 1ml 0.01m HCl → 8.28
 355 8.31
 403 8.34 2ml 0.01m HCl → 8.25
 * 408 8.26 Sample # 123099-8.
 417 8.30/8.31 1ml 0.01m HCl → 8.27
 425 8.30
 431 8.31 Tslm = 27.1 MN 12/30/99
 * 454 8.35 Sample 123099-9
 455 8.36 2ml 0.01m HCl → 8.26
 501 8.28
 522 8.34 2ml 0.01m HCl → 8.25

12/30/99 cont.

M. Nugent

525 pH = 8.24 Sample 123099-10
 528 8.28
 534 8.32
 538 8.33
 542 8.32 1ml 0.01m HCl → 8.28
 604 8.33 Sample 123099-11
 STOP.
 Filtered solution and

MN 12/30/99

Sample Name	Mass (all in grams) bottle+cap b+c+s	Sample b+c+s st+acid	TOTAL St+A	%sample	Subdivide the samples:								
					Name	b+c	b+c+s	Sample	Name	b+c	b+c+s	s+di	
123099-1	16.7333	21.8915	5.1582	22.1437	5.4104	95.3386	Np7a	x	x	Ca7a	4.0542	4.5485	10.8460
123099-2	16.8108	21.8576	5.0468	22.1106	5.2998	95.2262	Np8a	x	x	Ca8a	4.0602	4.5540	11.1287
123099-3	16.7274	26.6313	9.9039	26.8808	10.1534	97.5427	Np9a	16.6570	19.6600	Ca9a	4.0235	4.5171	11.0519
123099-4	16.666	24.5985	7.9325	24.8516	8.1856	96.9080	Np10a	16.8700	19.8575	Ca10a	4.0605	4.5539	11.1209
123099-5	16.9428	24.6258	7.6830	24.8799	7.9371	96.7986	Np11a	16.7604	20.7419	Ca11a	4.0348	4.5282	11.1591
123099-6	16.5833	24.4586	7.8753	24.7091	8.1258	96.9172	Np12a	16.8714	19.8786	Ca12a	4.0440	4.5365	11.1406
123099-7	16.7939	25.0639	8.2700	25.3169	8.5230	97.0316	Np13a	17.0764	20.3393	Ca13a	4.0385	4.5336	11.1812
123099-8	16.6844	24.5381	7.8537	24.7906	8.1062	96.8851	Np14a	16.5858	20.5958	Ca14a	4.0695	4.5641	11.0203
123099-9	16.8774	24.9904	8.1130	25.2441	8.3667	96.9677	Np15a	16.8647	20.8621	Ca15a			
123099-10	16.8548	25.2342	8.3794	25.4873	8.6325	97.0681	Np16a	16.9094	20.9020	Ca16a			
123099-11	17.1471	26.9853	9.8382	27.2392	10.0921	97.4842	Np17a	16.9295	20.9188	Ca17a			

12/30/99 cont
MNugent

SYSTEM NORMALIZED

C14 IPA DATA PROCESSED - 04-Jan-2000 12:53
C14 Eff (0-156 keV) = 96.59 %
C14 CHI SQUARE IPA DATA PROCESSED - 04-Jan-2000 13:03

NOTE! These LSA
results are suspect
due to problems with
the LSA -

The samples were
reanalyzed by LSA
after the problems
were resolved -

The correct results
are on pages 73
and 74 of this
notebook!

DO NOT USE
THESE →
LSA
RESULTS
!!!

04 Jan 2000 13:03 ALPHA/BETA - 1.09 Page #2
Protocol #:28 NP-237 10(-7) User: MIKE ALMENDAREZ

C14 Chi Square = 27.12
H3 IPA DATA PROCESSED - 04-Jan-2000 13:05
H3 Eff (0-18.6 keV) = 65.28 %
H3 CHI SQUARE IPA DATA PROCESSED - 04-Jan-2000 13:15
H3 Chi Square = 22.67
BKG IPA DATA PROCESSED - 04-Jan-2000 14:1c
Bkg (0-18.6 keV) = 17.38 cpm
Bkg (0-156 keV) = 24.58 cpm
C14 E²/B (1-156 keV) = 492.68
H3 E²/B (1-18.6 keV) = 244.44

03 Jan 2000 07:04 ALPHA/BETA - 1.09 Page #1
Protocol #:28 NP-237 10(-7) User: MIKE ALMENDAREZ

Time: 999.00
Data Mode: Alpha/Beta Nuclide: NP/PA Discriminator: 126
Background Subtract: 1st Vial

	LL	UL	LCR	25%	BKG
Beta A:	0.0 - 400	0	0.1	8.50	
Beta B:	100 - 400	0	0.5	0.60	
Alpha:	100 - 400	0	5.0	4.98	

Quench Indicator: SIS
5% 2s ALPHA COUNT REGION
Luminescence Correction On
Low Level Count Mode On
Coincidence Time(ns): 18
Delay Before Burst(ns): Normal
Protocol Data Filename: PROT.DAT
Count Data Filename: c:\prot28\data28.002
Spectrum Data Drive & Path: C:\PROT28

S#	TIME	CPMA	CPMB	CPMa	a:25%	SIS FLAG
1	999.00	8.50	0.60	4.978	2.836	89.5050 Black B
2	8.99	26.98	0.00	173.442	5.138	79.528 Np9a
3	9.04	27.05	0.00	172.345	5.140	83.989 Np10a
4	7.06	27.71	0.05	222.076	5.108	76.931 Np11a
5	9.61	19.01	0.00	161.619	5.153	79.266 Np12a
6	8.76	18.24	0.00	177.899	5.137	80.613 Np13a
7	7.53	21.31	0.00	207.505	5.120	70.956 Np14a
8	8.03	17.15	0.00	194.399	5.127	60.196 Np15a
9	7.86	13.80	0.00	198.712	5.124	68.888 Np16a
10	7.92	12.74	0.00	197.042	5.127	66.099 Np17a

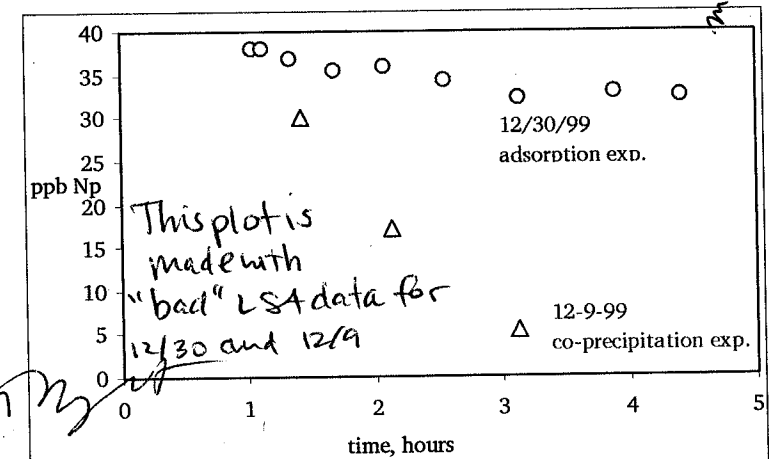
Name	Mass sample+ acid	%ofmass that is sample	Mass Sample g	counts	efficiency corr	# atoms	moles	m/l	ppb
123099-3 Np9a	3.003	97.542695	2.9292	173.442	173.442	2.82E+14	4.67E-10	1.60E-7	37.83
123099-4 Np10a	2.9875	96.907985	2.8951	172.345	172.345	2.80E+14	4.65E-10	1.60E-7	38.03
123099-5 Np11a	3.9815	96.798579	3.8540	222.076	222.076	3.60E+14	5.99E-10	1.55E-7	36.81
123099-6 Np12a	3.0072	96.917227	2.9145	161.619	161.619	2.62E+14	4.36E-10	1.49E-7	35.43
123099-7 Np13a	3.2629	97.031562	3.1660	177.899	177.899	2.89E+14	4.79E-10	1.51E-7	35.90
123099-8 Np14a	4.01	96.8851	3.8851	207.505	207.505	3.37E+14	5.59E-10	1.44E-7	34.12
123099-9 Np15a	3.9974	96.967741	3.8762	194.399	194.399	3.16E+14	5.24E-10	1.35E-7	32.04
123099-10 Np16a	3.9926	97.068057	3.8755	198.712	198.712	3.23E+14	5.36E-10	1.38E-7	32.76
123099-11 Np17a	3.9893	97.484171	3.8889	197.042	197.042	3.20E+14	5.31E-10	1.37E-7	32.37

adjusting the mass of sample for the amount of acid
added.

Much of uptake
in 12/9/99 experiment
is NOT due to adsorp-
tion onto container
walls - is presumably
uptake by calcite.

Note: This is still
true using the LSA
data on p. 73 and 74
of this notebook.

Note: Calcium analyses were not performed, because
experiment was not at steady state.



1-12-2000 M. Nugent

Calculate how much Np to add to Ca syringe solution: account for

- 1) dilution effect of syringe addition
- 2) adsorption onto container walls, and
- 3) coprecipitation effect.

1. Dilution Effect: if I add 60 ml from each syringe (120 ml total) $\frac{1}{2}$ the initial conc. Np = 30 ppb: $C_1 V_1 = C_2 V_2$ $30 \text{ ppb} \times 1000 \text{ ml} = C_2 \times 1120 \text{ ml}$
 The sltn is diluted to 26.785 ppb. \therefore set syringe sltn at 30 ppb:

make 30 ml of syringe sltn: $C_1 V_1 = C_2 V_2$

$$60 \text{ ml} \times 0.03 \text{ ppm} = 118 \text{ ppm} \times V_2 \Rightarrow V_2 = 0.015 \text{ ml}$$

 \therefore Add 15 μL of 118 ppm Np stock sltn to syringe sltn.

2. Adsorption: from 12/30/99 experiment, $\Delta \text{Np} / \Delta t = (\sim 38 - \sim 33) / 4.42 - 1.03 \text{ hrs}$
 $= 5 \text{ ppb} / 3.39 \text{ hrs} \sim 1.48 \text{ ppb Np lost per hr due to adsorption. } [6.22 \times 10^{-9} \text{ M/L per hr}]$

3. Coprecip: from 12/9/99 exp., $\Delta \text{Np} / \Delta t = (30.061 - 5.336) / 3.1 - 1.4 = 24.725 \text{ ppb} / 1.7 \text{ hrs}$
 $\sim 14.544 \text{ ppb lost per hr}$ [This probably accounts for adsorption + coprecip!]

So, Assuming a linear loss of Np with time, replace $\sim 15 \text{ ppb}$ of Np/hr.Prep of syringe solution:

Bottle (no cap): 19.2005 tare

0.01 M Ca sltn: 60.0350 g

118 Np sltn: 1.5518 g

calculate concentration: $C_1 V_1 = C_2 V_2$

$$C_2 = \frac{118 \text{ ppm} \times 1.5518 \text{ ml}}{(60.0350 + 1.5518) \text{ ml}} = 2.9732 \text{ ppm}$$

1.0050 g cc added

This is the mass of calcite added
 to the reaction vessel as described
 on page 55 of this notebook, CNWRA
 Controlled copy # 361.

Neptunium - Calcite Coprecipitation

1/12/2000

M. Nugent

Objective: Repeat exp of 12/7/99
 but with syringe sltn adjusted with Np
 so that the aqueous Np stays steady.

- add 250 μL Np stock solution
- syringe rate 0.133 ml/min, 0.01 M syringes
- $\sim 1 \text{ g}$ calcite seed
- 2 bubblers
- 1 L of filtered Equilibrium Solution

Summary: Experimental Conditions

Exp Date: 1/12/2000

Pre-Np spike:

Volume (L): 1 L

Solution A/B: B

ml Ca added: conc:

ml HCO₃ added: conc:

Mass calcite added (g): 1.0050

Post-Np spike:

ml Np added: 0.25 conc: 118 ppm

ml 0.32 M NaOH added: 0.25 ml

Syringe Np conc (m/l): 2.97 ppm

Syringe Ca conc (m/l): 0.01 M

Pump Infusion Rate: 0.133 ml/min

Steady State pH: 8.31/8.32

Temp, init. (Celsius): 23.5°C

Temp, final (Celsius): $\sim 27^\circ\text{C}$

Post-experiment:

Mass calcite recovered: (g)

AA - Ca: units:

LSA - Np: units:

Sample Numbers: 18-24

~~Calculation necessary~~Ptn Rate: ~~to calculate~~Np/Ca aq: ~~(in spread sheets)~~Np/Ca solid: ~~(in spread sheets)~~Kd: ~~(in spread sheets)~~

for each syringe:

4.562 ml added

1/13/2000

MN 1/12/2000

Reset pH meter, calibrated pH meter, conditioned electrode
 let electrode soak in storage sltn 1.5 hrs. Made fresh buffers.

Using working ATC probe!

Calibrate Electrode w/ 7, 10 buffers (23.5°C Room temp)

Time: (p.m.) Filter 1 L of sltn B

2:17 8.45

2:21 8.45

Added Seeds

2:22 8.42 Attached syringes @ 0.133 ml/min.

2:25 8.34

2:27 8.30 sample # 1 11200-1 filtered

2:32 8.28 Np + NaOH 0.25 ml of each added simultaneously

8.42 Added

2:33 8.30

56

1/12/2000 M. Nugent

236 8.27

238 8.28 Sample #2 1200-2

243 8.28 Detached HCO_3 syringe and replaced ferrule because of a leak.246 8.30 Reattached HCO_3 syringe. Not leaking254 8.32/31 23.5°C in solution.

302 8.32

310 8.32 23.8°C

316 8.32

340 8.32 ~~T = 24.1~~ MN 1/12/2000 Tsltn = 24.1°C

↳ sample 11200-3

351 8.32 ~~TS~~ MN 1/12/2000

415 8.32

446 8.31 Tsltn 25.7°C

449 8.32 Sample 11200-4

511 8.31

612 Stop syringes (refill) 30.66 ml added from each 8.31 pH

614 Start syringes 8.32 Tsltn = 27.2°C

619 pH = 8.32 Sample # 5 11200-5

622 Recalibrate electrode, pH = 8.34

725 8.31

728 Sample 11200-6 pH = 8.32

804 pH = 8.32

805 8.32 Sample 11200-7

807 STOP

45.562 ml added from each syringe

Solution filtered and solid saved.

Mass of Solids recovered: 0.9218 grams

Date Recovered: 1/13/2000

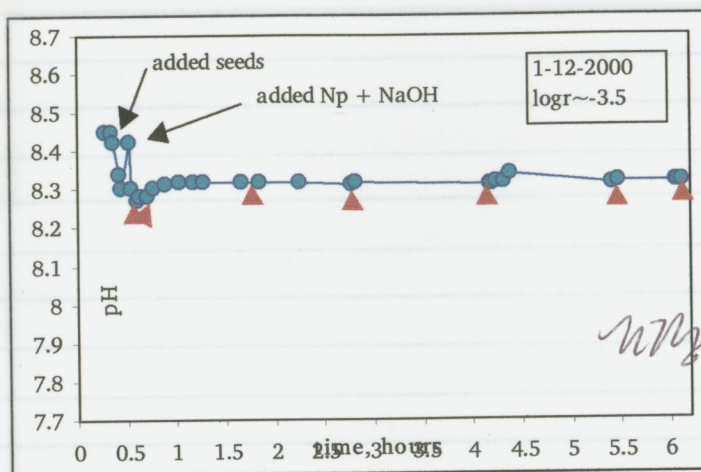
Recovered by: M. Nugent

M. Nugent 1/12/2000
con't

Sample Name	Mass (all in grams) bottle+ca b+c+s	Sample s+acid	TOTAL S+A	%sample	Subdivide the sample:											
					Name	b+c	Neptunium	b+c+s	Sample	Name	b+c	Calcium	b+c+s	b+c+	s+di	
11200-1	16.8377	25.8399	9.0022	94.3696	Np18a	16.9928	19.9931	3.0003	Ca18a	4.0532	4.5469	11.0988	Ca18b	4.0401	4.5333	11.1480
11200-2	16.6611	23.5153	6.8542	96.4579	Np19a	16.6067	19.6030	2.9963	Ca19a	4.0527	0.4935	7.0278	Ca19b	4.0539	0.4951	7.0636
11200-3	16.9059	23.4476	6.5417	96.2708	Np20a	16.5847	19.5720	2.9873	Ca20a	4.0619	0.4916	7.0470	Ca20b	4.1468	0.4977	7.0493
11200-4	16.6157	23.5055	6.8898	96.4539	Np21a	16.6867	19.7838	3.0971	Ca21a	4.0658	0.4934	7.2394	Ca21b	4.0867	0.4988	7.0198
11200-5	16.9079	24.7681	7.8602	96.8685	Np22a	16.8644	19.8544	2.9900	Ca22a	4.0346	0.4954	7.0908	Ca22b	4.1507	4.6483	11.0782
11200-6	16.8410	24.7048	7.8638	96.8556	Np23a	16.7914	19.7796	2.9882	Ca23a	4.0540	0.4963	7.0294	Ca23b	4.0349	0.4983	7.0455
11200-7	16.8551	24.7643	7.9092	96.8636	Np24a	16.8111	19.8083	2.9972	Ca24a	4.0788	4.5733	11.1372	Ca24b	4.0446	0.4953	7.2237

(Note: some are b+c+s and some are s only)

(Note: some are b+c+s and some are s only)



1/12/2000

M. Nugent

Seed analysis by LST: (Make estimate for amt. of seed to dissolve)

Assuming, from 12/9/99 exp, that ~ 15 ppb Np lost per hour.

$$45.562 \text{ mL} \times \frac{\text{min}}{0.1333 \text{ mL}} \times \frac{\text{hr}}{60 \text{ min}} \times \frac{15 \text{ ppb}}{\text{hr}} = 85.45 \text{ ppb Np lost}$$

$$\text{Convert Np} \rightarrow \text{moles: } 85.45 \text{ ppb} \times \frac{\text{ppm}}{1000} \times \frac{1}{237041 \text{ g/mol}} \times 1 \text{ L} = 3.6049 \times 10^{-7} \text{ moles Np}$$

Calculate total Calcite at end of reaction:

Assume all Ca added by syringe is precipitated as calcite:

$$45.562 \text{ mL} \times \frac{1 \text{ L}}{1000 \text{ mL}} \times 0.01 \text{ M Ca} = 4.556 \times 10^{-4} \text{ moles Ca} = \text{moles Calcite}$$

(0.04556 g Calcite precipitated)

$$\text{Total calcite} = \text{Seed} + \text{precipitated} = 1.0050 \text{ g} + 0.04556 \text{ g} = 1.05056 \text{ g}$$

Assume 3.6049×10^{-7} moles Np in 1.05056 g calcite

dissolve 0.1 g calcite in 10 mL acid:

$$0.1 \text{ g} = 9.5\% \text{ of calcite} \quad 0.1 / 1.05056 = 0.095787$$

$$\therefore 0.095787 \times 3.6049 \times 10^{-7} \text{ moles Np} = 3.4314 \times 10^{-8} \text{ moles Np}$$

$$\frac{3.4314 \times 10^{-8} \text{ moles Np}}{0.01 \text{ L}} = 3.4314 \times 10^{-6} \frac{\text{mol}}{\text{L}} \text{ Np} = \boxed{0.8133 \text{ ppm Np}}$$

This should be sufficient for LST analysis.

Seed prep for LST:

Mass of Bottle + Cap: 16.8827 g

0.1282 g of seed added, 3.5 mL 1.0 N HCl added.

Mass after dissolution = 3.6086 g

Date of sample prep: 1/13/2000

Prepared by: M. Nugent.

Sample Name: S2

Pages 59 Through 61 Are Intentionally
Left Blank

3/7/2000 M. Nugent

12:09 pm

2.71 5ml $\text{CaCl}_2 \rightarrow 7.70$

12:11 pm

5ml NaHCO_3 0.1M $\rightarrow 8.54$

12:15

Bubblers are not overwhelming the system w/ bubbles.
8.26

Reaction Vessel: Calculate the effect of addition of Tc spike on rxn vessel:

 $(30 \text{ ppb spike}) \times V_1 = (30 \text{ ppb})(100 \text{ mL}) \Rightarrow \text{Add } 100 \text{ mL of spike}$

NaCl correction:

 $0.1 \text{ L} \times 0.1 \text{ M NaCl} = 0.01 \text{ moles}$ $0.9 \text{ L} \times 0.1 \text{ M NaCl} = 0.09 \text{ moles}$ no correction 0.1 mLSyringe set

Add 10 mL of Tc spike to make 100 mL soln.

 $0.1 \text{ L} \times 0.01 \text{ M} \times 147 = 0.147 \text{ g Ca}$ $0.09 \text{ L} \times 0.1 \text{ M} \times 58.5 \text{ g} = 0.5265 \text{ g NaCl}$ 10.05 g Tc spike diluted to 100.82 g (nanopure)
Added Ca and NaCl.

Calculate the rate of syringe addition:

$$\frac{0.0316 \text{ mL} \times 60}{\text{min} \cdot \text{hr}} = \frac{33 \text{ mL}}{1896 \text{ mL/hr}} = 1.73\% \text{ ok}$$

Actual Rate:

$$0.0316 \text{ mL/min} \times \frac{1}{1000 \text{ mL}} \times \frac{60 \text{ min}}{\text{hr}} \approx 0.01 \text{ M} = 1.896 \times 10^{-5} \frac{\text{mol}}{\text{min}} \times 0.253 \frac{\text{m}^3}{\text{g}} \times 1 \text{ g}$$
$$7.49 \times 10^{-5} \text{ mol/m}^2 \cdot \text{hr}$$
$$\log r = -4.1253$$

3/7/2000

M. Nugent

Tc - Calcite Coprecipitation

Objective:

Try to Add Tc and coprecipitate w/ calcite to see how much Tc is taken up by Cc. Perform an experiment identical to the one described by the procedure on p. 3, 4 of this notebook (Controlled copy 361)

But Choose:

Log rate ≈ -4.5

1 g Calcite seed

Syringes @ 0.01 mL

Ionic strength 0.1 M w/ NaCl

Tc $\approx 30 \text{ ppb}$

Tc used in experiment

Tc 99 SPIKE 43A

Summary: Experimental Conditions

Tc!

Exp Date: 3/7/2000

Pre-Np spike:

Volume (L):

Solution A/B:

ml Ca added: 5 mL conc: 0.1 M

ml HCO_3 added: 5 mL conc: 0.1 M

Mass calcite added (g): 1.0046 g

Post-Np spike:

ml Na_2CO_3 added: 100 mL conc: $\approx 30 \text{ ppb}$

ml 0.32 M NaOH added: —

Syringe Np conc (m/L): 10 mL Tc spike diluted to 100.82 g

Syringe Ca conc (m/L): 0.01

Pump Infusion Rate: 0.0316 mL/min

Steady State pH: 8.3 (diffing down a little?)

Temp, init. (Celsius): 21.4°C

Temp, final (Celsius): 25.1°C Tc spike 43A

Post-experiment:

Mass calcite recovered: 0.8123 (g)

AA - Ca: — units: —

LSA - Np: — units: —

Sample Numbers: 25 - 32

Calculated:

Ptn Rate: —

Np/Ca aq: —

Np/Ca solid: —

Kd: —

85.026 mL added from each Syringe

Tc Spike #43 + 43A (Notebook 031, pg 288-289)

NIST Cert:

Source No. 630-2-2 + 630-2-3

06/12/00

Catalog No. 95335; Date 08/21/98 (Spike 43);

Vendor: Isotope Products Laboratories

3/7/2000
M. Nugent

1.0051g seed

0.0005g

1.0046g seed Actually Added

Mass of calcite added to the
reaction vessel recorded 3/7/2000.

3/7/2000
cont

M. Nugent

Added stirbar, bubblers, filtered 0.9 L sltnA

12⁰⁰ pm pH = 7.71 Added 5ml 0.1M CaCl₂ → 7.70

12¹¹ pm 5ml 0.1M Na₂HPO₄ pH → 8.54

12¹⁵ pm 8.26

12¹⁹ 8.06 = pH

12²³ 8.05

Bubblers are producing low volume of
bubbles.

STOPPED!

Discarded solution, cleaned bubblers, changed 1 bubbler
to a more efficient bubbler. Use 1 L of sltnA

400 = MW 3/7/2000

901.0g sltnA filtered bubbling, stirring

12⁴⁴ pm 8.10 21.4°C

45 5ml 0.1M Ca → 8.01

46 " 1HCO₃ → 8.42

12⁴⁸ 8.33 = pH

12⁴⁹ Added seeds 8.30 → 8.28

Attached syringes (Ca, no Tc) 0.0316 ml/min

12⁵² pH = 8.26 21.6°C

235 pH = 8.39 T = 24.0°C

240 8.40

327 8.40/8.41

415 pH 8.40 Sample #1 372000-1 (filtered)

417 STOP Synges Add Tc 100.80g

pH → 8.41

Added 1ml of 0.1M Ca, 1ml 0.1M H₂O 6.5358 ml Added

422 Resume Synges - using Tc-Ca-NaCl syringe.
pH = 8.43

424 Sample #2 372000-2 pH = 8.42 T = 24.9°C

446 pm 8.37 24.9°C

448 Calibrate pH electrode pH = 8.40

517 pH = 8.41

628 pm 8.40/1 T = 25.3°C

3/8/2000

453 AM "Pump stalled" 30.93x ml delivered

Hit 'set' and cleared screen - pump was ⇒

66

3/7/2000
M. Nugent

delivering

954 8.38 25.9°C

956 Calibrate electrode \rightarrow 8.35

The pump was stuck - removed block that was preventing pump from delivering @ 31.00 gml. Hit run on pump - pH has \downarrow it is possible that not enough sltn was delivered (ie not 31.00 gml) - weigh total mass @ end to see.

10:01 am 8.34/5 Sample #3 372000-3

10:04 am 8.33/8.34

11:47 am 8.34

12:15 pm 8.34

12:17 pm 8.31 26.1°C

calibrate

12:29 8.33 37.763 ml Added - STOP Syringes @ 1:31

Add sltn to both syringes.

1:35 pm ~~Re-attach~~ attached syringes

pH = 8.34

2:27 8.34

2:30 8.35 Sample #4

2:30 Opened stopcocks for syringes! 39.550 ml

Some sltn leaked \therefore < (39.550 - 37.763) ml added from each syringe!

3:35 pH = 8.32 T = 26.6°C

4:14 pH = 8.32

6:41 pH = 8.30 27.6°C

calibrate

6:44 8.33

6:47 Sample #5 372000-5 8.33

Continued on next page

67

3/7/2000
Cont.
M. Nugent

3/9/2000

9:21 am pump stalled 72.134 ml

9:25 pH = 8.32 T = 25.0°C

refilled syringes + started pump

Calibrated electrode

9:28 pH = 8.30

9:33 8.30 Sample #6 372000-6

10:04 8.29

11:39 8.29 24.8°C

12:15 8.28

2:56 8.26 25.1°C

2:58 Calibrate 8.30 25.1°C

3:09 Sample #7 372000-7 8.30

DISCARDED - CONTAMINATED w/ CALCITE

Filter did not work - Calcite in aqueous sltn.

3:23 8.30 Sample #8 372000-8

4:11 8.31

4:14 8.31 Stop

1573.9 g = run vessel + stir bar + sltn + seeds

85.026 ml of each sltn added.

0.8123 g of sample recovered.

Date recovered: 3/10/2000

Recovered by: M. Nugent

3/7/2000

Tc analyses using 25a as blank!
 AT Mn 3/11/2000. Also use a blank prepared properly. Run true blank first, 25a second, to determine if 25a can be a blank.

Calculation of K_D : See spreadsheet.

M. Nugent 3/7/2000 69

Calcite seed analysis: (by LST)

3/27/2000

1. Dissolve most seed in HCl:

Sample Name on Bottle: Tc Seed 372000

Bottle + Cap: 16.9072 (tare)

Calcite seed: 0.6868g

In 1ml increments, 7ml 1.0M HCl added, then 1ml (in 0.5ml increments) conc. HCl (12M). Seed dissolves.

Mass of Seed + acid after dissolution:

2. Transfer ~3ml

Sample name on bottle: Tc Seed

Bottle + cap: 16.5499 (tare)

Avg. Sample: 3.2276 grams

3. Prepare blank in same way, but w/ unreacted calcite seed. Name: Tc Blank 1

B+C: 16.6937g

S: 0.6958g

7ml 1.0n + 1ml 12.0n HCl

Total mass ~~after~~ after dissoln of calcite + acid = 8.5473g DR 8/14/00

4. Transfer ~3ml of Tc Blank 1

Name: Tc Blank 2

B+C 16.8672 tare

S 3.2186 grams.

Run in LST 3/27/2000

See p. 72

Sample Name	Mass (all in grams) bottle+cap	b+c+s	Sample	b+c+ s+acid	TOTAL S+A	%sample	Subdivide the sample:			
							Name	b+c	b+c+s	Name
372000-1	16.5735	21.2366	4.6631	21.4925	4.9190	94.7977	Tc25a	16.8927	19.9015	Ca25a
372000-2	16.7426	25.7285	8.9859	26.2412	9.4986	94.6024	Tc26a	16.8273	19.8355	Ca26a
372000-3	16.5612	24.9719	8.4107	25.4855	8.9243	94.2449	Tc27a	16.8854	19.8849	Ca27a
372000-4	16.8786	25.5554	8.6768	26.0676	9.1890	94.4259	Tc28a	16.6305	19.632	Ca28a
372000-5	16.9019	24.8981	7.9962	25.4122	8.5103	93.9591	Tc29a	16.911	19.8247	Ca29a
372000-6	16.9130	25.2225	8.3095	25.7361	8.8231	94.1789	Tc30a	17.1262	20.1231	Ca30a
372000-7	16.8008	25.2868	8.4860	26.0553	9.2545	91.6959	Tc31a	16.8408	19.8255	Ca31a
372000-8	16.8368	25.2191	8.3823	25.7323	8.8955	94.2308	Tc32a	16.7241	19.7197	Ca32a

13/00 5:35:14 PM QuantaSmart (TM) - 1.10
Protocol# 41 - 99Tc 3% 2sigma (1 hSerial# 405314)

Page # 1
User: Nugent

Assay Definition-

Assay Description:

Protocol Name: TC-99 CPM

Add'l Heading: BETA CPM TC-99

Original Protocol Settings (actual values may have changed):

Delay Before Burst: 75

Coincidence Time: 18

Count Mode: Normal

Half Life A: 0.00 Hours

Half Life B: 0.00 Hours

Assay imported during upgrade 1/25/00.

Assay Type: CPM

Report Name: 99Tc 3%2sigma

Output Data Path: C:\Packard\Tricarb\Results\Nugent\99Tc 3% 2sigma (1 hour)

Raw Results Path: C:\Packard\Tricarb\Results\Nugent\99Tc 3% 2sigma (1 hour)

Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\99Tc 3% 2sigma (1 hour)\99Tc 2% 2sigma.txt

Count Conditions-

Nuclide: 99Tc

Quench Indicator: SIS

External Std Terminator (sec): n/a

Pre-Count Delay (min): 0.00

Quench Set: n/a

Count Time (min): 60.00

Count Mode: Normal

Assay Count Cycles: 1

Repeat Sample Count: 1

#Vials/Sample: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
A	0.0	300.0	1st Vial	3.00
B	0.4	300.0	1st Vial	0.00
C	0.0	2000.0	1st Vial	0.00

Count Corrections-

Static Controller: On

Luminescence Correction: Off

Colored Samples: n/a

Heterogeneity Monitor: n/a

Coincidence Time (nsec): 18

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
A				
B				
C				

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMC	C:2S%	SIS MESSAGES
1	60.00	39	4.13	39	4.13	52	3.57	198.48 Blank B
2	1.54	2860	3.03	2860	3.03	2863	3.04	137.68 Tc27
3	1.66	2762	2.98	2762	2.98	2762	2.98	137.71 Tc28
4	1.60	2747	3.04	2747	3.04	2745	3.05	135.41 Tc29
5	1.70	2593	3.04	2593	3.04	2591	3.04	136.50 Tc20
6	1.68	2623	3.04	2623	3.04	2622	3.04	136.76 Tc31
7	1.69	2608	3.04	2608	3.04	2608	3.04	136.88 Tc22

98.907 g

3/13/00 5:35:14 PM

QuantaSmart (TM) - 1.10

Protocol# 41 - 99Tc 3% 2sigma (1 hSerial# 405314)

Page # 2
User: Nugent

8 1.66 2645 3.04 2645 3.04 2645 3.05 137.91

Calculations for LSA data conversions 3/10/2000 M. Nugent

Mass ⁹⁹Tc 98.907g $\frac{1}{2} \text{ life } 213,000 \text{ years} = 6.717168 \times 10^{+12} \text{ seconds}$

0.693

 $= 1.1195 \times 10^{+11} \text{ minutes}$ $\frac{0.693}{6.717168 \times 10^{+12}} = 1.195 \times 10^{-11}$ $\lambda = 6.19 \times 10^{-12}$

Converting LSA Tc data into ppb Tc

237Np = 237.041678 g/m

half life: 2.14×10^6 years

Name	Mass S+A	%sample	Mass S g	CPMA counts	efficiency corr	# atoms	moles	m/l	ppb	
372000-1	Tc25	4.919	94.798	4.6631	39	39.000	6.30E+12	1.05E-11	2.24E-9	0.22
372000-2	Tc26	9.4986	94.602	8.9859	2860	2860.000	4.62E+14	7.67E-10	8.54E-8	8.44
372000-3	Tc27	8.9243	94.245	8.4107	2762	2762.000	4.46E+14	7.41E-10	8.81E-8	8.71
372000-4	Tc28	9.189	94.426	8.6768	2747	2747.000	4.44E+14	7.37E-10	8.49E-8	8.40
372000-5	Tc29	8.5103	93.959	7.9962	2593	2593.000	4.19E+14	6.96E-10	8.70E-8	8.60
372000-6	Tc30	8.8231	94.179	8.3095	2623	2623.000	4.24E+14	7.04E-10	8.47E-8	8.38
372000-7	Tc31	9.2545	91.696	8.4860	2608	2608.000	4.21E+14	7.00E-10	8.24E-8	8.15
372000-8	Tc32	8.8955	94.231	8.3823	2645	2645.000	4.27E+14	7.10E-10	8.46E-8	8.37

72 3/17/2000

3/30/00 10:50:36 AM QuantaSmart (TM) - 1.10
Protocol# 41 - 99Tc 3% 2sigma.lsa Serial# 405314Page # 1
User: Nugent

Assay Definition-

Assay Description:

Protocol Name: TC-99 CPM
Add'l Heading: BETA CPM TC-99
Original Protocol Settings (actual values may have changed):
Delay Before Burst: 75
Coincidence Time: 18
Count Mode: Normal
Half Life A: 0.00 Hours
Half Life B: 0.00 Hours
Assay imported during upgrade 1/25/00.

Assay Type: CPM

Report Name: 99Tc 3%2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\99Tc 3% 2sigma
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\99Tc 3% 2sigma
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\99Tc 3% 2sigma\99Tc 2% 2sigma.txt

Count Conditions-

Nuclide: 99Tc

Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00
Quench Set: n/a

Count Time (min): 999.99

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
A	0.0	300.0	1st Vial	3.00
B	0.4	300.0	1st Vial	0.00
C	0.0	2000.0	1st Vial	0.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
A				
B				
C				

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMC	C:2S%	SIS	MESSAGES
1	999.99	37	1.03	37	1.04	51	0.89	173.30	
2	5.73	739	3.15	739	3.15	738	3.18	418.50	

Blank
Tc SeedLSA analysis:
Tc-Ce seed
analysis

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Re-counting LSA samples 73
for [NP] 3.17.2000
M. Nugent3/14/00 6:55:21 PM QuantaSmart (TM) - 1.10
Protocol# 28 - Manual Np_Pa 3%2S.lSerial# 405314Page # 1
User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta

Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa

Quench Indicator: SIS

External Std Terminator (sec): n/a

Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 999.99

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
Blank	1	813.66	34.09	1.20	40.88	1.10	2.00	4.95	845.91
Np 3b	2	35.07	112.93	3.64	113.55	3.72	124.71	3.05	275.41
Np 4b	3	57.48	64.24	4.12	64.96	4.23	75.31	3.08	298.66
Np 5b	4	163.94	23.83	5.28	24.84	5.41	25.11	3.26	378.33
Np 6b	5	156.19	2.92	36.13	3.97	29.25	0.74	38.09	1198.02
Np 10a	6	31.62	124.10	3.62	124.99	3.68	138.57	3.04	288.60
Np 11a	7	24.36	157.74	3.57	159.36	3.61	180.59	3.03	285.53
Np 12a	8	32.15	119.22	3.68	120.08	3.75	136.22	3.05	299.30
Np 13a	9	28.67	124.61	3.79	125.21	3.86	153.04	3.04	264.62
Np 14a	10	25.03	155.56	3.55	157.28	3.59	175.54	3.03	318.15
Np 15a	11	25.02	150.40	3.62	152.00	3.67	175.65	3.03	289.83
Np 16a	12	25.07	146.29	3.68	146.11	3.75	175.26	3.03	251.37

72 3/7/2000

3/29/00 5:51:57 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

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Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 32.74 Date Processed: 3/29/00 5:51:57 PM
14C Chi Square: 20.17 Date Processed: 3/29/00 5:51:57 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 259.35 Date Processed: 3/29/00 5:51:57 PM
14C E²/B (0-156 keV and 1-156 keV): 472.13 Date Processed: 3/29/00 5:51:57 PM
3H Efficiency (0-18.6 keV): 65.70 Date Processed: 3/29/00 5:51:57 PM
14C Efficiency (0-156 keV): 96.63 Date Processed: 3/29/00 5:51:57 PM
IPA Background Date Processed: 3/29/00 5:51:57 PM
3H Background CPM (0-18.6 keV): 16.43 Date Processed: 3/29/00 5:51:57 PM
14C Background CPM (0-156 keV): 24.97 Date Processed: 3/29/00 5:51:57 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

Re-counting LSA samples 73
for ☒ Np
3.17.2000
M. Nugent

3/14/00 6:55:21 PM QuantaSmart (TM) - 1.10
Protocol# 28 - Manual Np_Pa 3%2S.1:Serial# 405314

Page # 1
User: Nugent

LSA analysis - re-counting
Samples for
experiments: 12/9/99 & 12/30/99

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Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 999.99

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
Blank	1	813.66	34.09	1.20	40.88	2.00	4.95	845.91	
Np 3b	2	35.07	112.93	3.64	113.55	3.72	124.71	3.05	275.41
Np 4b	3	57.48	64.24	4.12	64.96	4.23	75.31	3.08	298.66
Np 5b	4	163.94	23.83	5.28	24.84	5.41	25.11	3.26	378.33
Np 6b	5	156.19	2.92	36.13	3.97	29.25	0.74	38.09	1198.02
Np 10a	6	31.62	124.10	3.62	124.99	3.68	138.57	3.04	288.60
Np 11a	7	24.36	157.74	3.57	159.36	3.61	180.59	3.03	285.53
Np 12a	8	32.15	119.22	3.68	120.08	3.75	136.22	3.05	299.30
Np 13a	9	28.67	124.61	3.79	125.21	3.86	153.04	3.04	264.62
Np 14a	10	25.03	155.56	3.55	157.28	3.59	175.54	3.03	318.15
Np 15a	11	25.02	150.40	3.62	152.00	3.67	175.65	3.03	289.83
Np 16a	12	25.07	146.29	3.68	146.11	3.75	175.26	3.03	251.37

3/12/2000

MN3/17/2000

LSA, re-analyse samples.

3/21/00 2:42:48 PM

Serial# 405314

Page # 1

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00
Quench Set:
Count Time (min): 999.99
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results	S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	999.99	33.33	1.10	40.03	1.00	1.99	4.48	857.85	Blank B	
2	50.54	76.54	3.88	76.69	4.00	85.94	3.07	277.64	3aNP	
3	87.25	40.40	4.64	40.43	4.85	48.95	3.13	270.29	4aNP	
4	235.00	13.08	7.35	13.39	7.72	16.92	3.39	274.08	5	
5	999.99	0.00	0.00	0.00	0.00	0.00	0.00	0.00	6	
6	33.79	105.76	3.85	106.05	3.94	129.56	3.05	259.97	3bNP	
7	59.77	65.18	3.98	66.10	4.08	72.38	3.08	301.58	4	
8	168.14	23.37	5.21	24.24	5.36	24.45	3.26	358.44	5	
9	999.99	3.11	17.00	4.07	14.25	0.77	17.83	825.50	6	
10	31.47	125.61	3.59	126.61	3.65	139.26	3.04	278.03	Np9a	
11	32.31	123.90	3.57	125.03	3.63	135.68	3.04	274.65	10	

Lst re-analysis
for experiments
12/9/99, 12/30/99,
and 1/12/2000.

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M. Nugent 3/12/2000

4/5/00 9:58:51 AM

QuantaSmart (TM) - 1.10

Page # 1

Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00
Quench Set:
Count Time (min): 999.99
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results	S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	999.99	33.27	1.10	39.84	1.00	1.96	4.52	846.80	Blank B	
2	39.64	17.74	12.96	19.42	12.76	110.15	3.05	568.55	Np33a	
3	54.90	10.26	17.72	11.49	17.18	79.06	3.08	580.90	34	
4	72.78	6.63	23.01	8.91	18.90	59.10	3.10	1095.57	35	
5	103.86	6.65	19.45	8.24	17.21	40.84	3.15	845.79	36	
6	99.11	7.76	17.23	10.01	14.72	42.90	3.14	1049.80	37	
7	97.34	7.90	17.10	9.96	14.91	43.70	3.14	894.72	38	
8	25.40	142.51	3.70	143.08	3.76	173.00	3.03	295.06	Np20a	
9	18.41	202.41	3.54	203.13	3.58	239.70	3.02	291.62	21	
10	14.87	247.36	3.52	248.06	3.55	297.10	3.02	284.10	22	
11	11.25	324.68	3.48	325.59	3.50	393.42	3.01	306.06	23	
12	10.01	360.83	3.48	360.46	3.51	442.00	3.01	280.97	24	

4/5/00 9:21:05 AM: Warning: User Has Modified Count Conditions.

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3/17/2000

MN 3/17/2000

LSA, re-analyze samples.

3/21/00 2:42:48 PM

Serial# 405314

Page # 2

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12	24.17	160.21	3.54	161.88	3.58	181.88	3.03	298.15 11
13	32.57	122.12	3.59	123.71	3.64	134.46	3.05	297.23 12
14	29.89	131.64	3.58	132.74	3.63	146.76	3.04	286.77 13
15	24.36	156.16	3.58	158.29	3.61	180.48	3.03	290.46 14
16	24.65	149.67	3.65	150.24	3.71	178.29	3.03	263.94 15
17	24.71	154.41	3.58	155.16	3.63	177.86	3.03	265.29 16
18	25.29	150.18	3.60	151.79	3.64	174.05	3.03	300.64 17
19	999.99	4.76	11.24	6.51	9.04	0.00	0.00	1058.44 18
20	34.87	105.10	3.81	106.60	3.87	125.48	3.05	322.55 19
21	25.64	138.35	3.75	139.77	3.80	171.37	3.04	315.61 20
22	19.06	188.28	3.63	189.04	3.67	231.22	3.03	291.84 21
23	15.05	244.28	3.52	246.35	3.55	293.36	3.02	320.83 22
24	11.10	307.93	3.60	308.26	3.64	398.91	3.01	293.82 23
25	10.17	350.94	3.50	352.60	3.53	434.98	3.01	310.27 24

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M. Nugent 3/17/2000

75

4/5/00 9:58:51 AM

QuantaSmart (TM) - 1.10

Page # 2

User: Nugent

Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

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13 783.00 3.75 15.14 5.44 11.50 0.00 0.00 1172.67 Np18a
4/5/00 9:21:47 AM: Warning: User Has Modified Count Conditions.
14 36.06 108.02 3.68 108.89 3.75 121.34 3.05 293.71 Np19a

terminated counting at 873 minutes

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3/17/2000

MN 3/17/2000
LSA, re-analyse samples.3/21/00 1:36:47 AM
SNC ProtocolQuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 3/3

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 12.83 Date Processed: 3/21/00 1:36:47 AM
14C Chi Square: 8.51 Date Processed: 3/21/00 1:36:47 AM
3H E²/B (0-18.6 keV and 1-18.6 keV): 238.10 Date Processed: 3/21/00 1:36:47 AM
14C E²/B (0-156 keV and 1-156 keV): 442.55 Date Processed: 3/21/00 1:36:47 AM
3H Efficiency (0-18.6 keV): 65.72 Date Processed: 3/21/00 1:36:47 AM
14C Efficiency (0-156 keV): 96.17 Date Processed: 3/21/00 1:36:47 AM
IPA Background Date Processed: 3/21/00 1:36:47 AM
3H Background CPM (0-18.6 keV): 18.02 Date Processed: 3/21/00 1:36:47 AM
14C Background CPM (0-156 keV): 26.45 Date Processed: 3/21/00 1:36:47 AM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

M. Nugent

M. Nugent 3/17/2000

4/3/00 5:56:42 PM
SNC ProtocolQuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

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Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 11.58 Date Processed: 4/3/00 5:56:41 PM
14C Chi Square: 13.10 Date Processed: 4/3/00 5:56:41 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 261.48 Date Processed: 4/3/00 5:56:41 PM
14C E²/B (0-156 keV and 1-156 keV): 479.98 Date Processed: 4/3/00 5:56:41 PM
3H Efficiency (0-18.6 keV): 65.56 Date Processed: 4/3/00 5:56:41 PM
14C Efficiency (0-156 keV): 96.33 Date Processed: 4/3/00 5:56:41 PM
IPA Background Date Processed: 4/3/00 5:56:41 PM
3H Background CPM (0-18.6 keV): 16.18 Date Processed: 4/3/00 5:56:41 PM
14C Background CPM (0-156 keV): 24.65 Date Processed: 4/3/00 5:56:41 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

M. Nugent

M. Nugent

M. Nugent

M. Nugent

3/17/2000

LSA, re-analyse samples.

mn 3/17/2000

3/21/00 1:36:47 AM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 3/3

Calibration Information
Software Version IC: 2.09
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14C E²/B (0-156 keV and 1-156 keV): 442.55 Date Processed: 3/21/00 1:36:47 AM
3H Efficiency (0-18.6 keV): 65.72 Date Processed: 3/21/00 1:36:47 AM
14C Efficiency (0-156 keV): 96.17 Date Processed: 3/21/00 1:36:47 AM
IPA Background Date Processed: 3/21/00 1:36:47 AM
3H Background CPM (0-18.6 keV): 18.02 Date Processed: 3/21/00 1:36:47 AM
14C Background CPM (0-156 keV): 26.45 Date Processed: 3/21/00 1:36:47 AM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

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M Nugent

4/5/00 11:30:39 AM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

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Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 15.53 Date Processed: 4/5/00 11:30:39 AM
14C Chi Square: 16.13 Date Processed: 4/5/00 11:30:39 AM
3H E²/B (0-18.6 keV and 1-18.6 keV): 255.40 Date Processed: 4/5/00 11:30:39 AM
14C E²/B (0-156 keV and 1-156 keV): 482.18 Date Processed: 4/5/00 11:30:39 AM
3H Efficiency (0-18.6 keV): 65.72 Date Processed: 4/5/00 11:30:39 AM
14C Efficiency (0-156 keV): 96.26 Date Processed: 4/5/00 11:30:39 AM
IPA Background Date Processed: 4/5/00 11:30:39 AM
3H Background CPM (0-18.6 keV): 16.72 Date Processed: 4/5/00 11:30:39 AM
14C Background CPM (0-156 keV): 24.50 Date Processed: 4/5/00 11:30:39 AM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

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3/27/2000

Seed Analysis for

M. Nugent

Experiments 12/1/99 and 1/12/2000 = (11200)

Np 8/14/00 = 12999

1. Dissolve seed mat'l (which contains Np) in
1.0m HCl:

Experiment: 12/1/99 = 12999

Name on Bottle: S1A

Bottle+Cap: 16.6123g tare

0.1399 g of calcite seed.

~3.5ml 1.0m HCl added

Total mass = 22.2204g
after seed dissolution

Np 8/14/00

Experiment 1/12/2000 = 11200

Name on Bottle S2

Bottle+Cap 16.8827 tare

0.1282g calcite seed } mass = 3.6086 after seed
~3.5ml 1.0m HCl } dissolution

2. Prepare a Blank w/ unreacted calcite seed:

Name on Bottle: Np 0.12g Blank

Bottle+cap 16.7841

0.1458 g calcite solid } Total 3.8662g
~3.75ml 1.0m HCl }

Add cocktail, analyze by LSA.

3/1/00 9:24:58 AM

QuantaSmart (TM) - 1.10

Page # 1

Protocol# 41 - Manual Np_Pa 3%2S.1 Serial# 405314

User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta

Report Name: Manual Np_Pa 3% 2sigma

Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa

Quench Indicator: SIS

External Std Terminator (sec): n/a

Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 999.99

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions Half Life

Beta A

Beta B

Alpha

Units

Reference Date

Reference Time

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	999.99	37.21	1.04	45.64	0.94	1.58	5.03	897.87	Blank B
2	0.69	5964.24	3.13	5965.95	3.13	6517.26	2.98	230.37	S1A
3	0.55	9768.24	2.73	9774.36	2.73	8176.60	2.98	232.68	S2

3/30/2000 M. Nugent

Summary: Experimental Conditions

Pre-Np spike:

Volume (L): 1L
 Solution A/B: A
 ml Ca added: 512 conc: 0.1
 ml HCO₃ added: 97 conc: 0.1
 Mass calcite added (g): 1.0011g

Post-Np spike:

ml Np added: 0.25 conc: 118
 ml 0.32M NaOH added: 0.25
 Syringe Np conc (m/l): ~1.4 ppm
 Syringe Ca conc (m/l): 0.1
 Pump Infusion Rate: 0.2406 ml/min
 Steady State pH: pH dropped 8.32 → 8.01 not steady state
 Temp, init. (Celcius): 23.2
 Temp, final (Celcius): 24.5

Exp Date: 3/30/2000

Post-experiment:

Mass calcite recovered: 1.0112 (g)
 AA - Ca: _____ units:
 LSA - Np: _____ units:
 Sample Numbers: 33 - 38
 Seed # S3

Calculated:

Pttm Rate: not necessary to calculate 3/31/2000
 Np/Ca aq: not necessary to calculate 3/31/2000
 Np/Ca solid: _____
 Kd: _____

30.392 ml Ca added, 2.5612
 ml added to spike.

Neptunium Calcite Coprecipitation

3-30-2000

M. Nugent

Try Np - Calcite coprecipitation with ~1.4 ppm
 in syringe @ fast rate of precipitation

$\log r = -2.8 \text{ moles/m}^2/\text{g} \rightarrow$ Syringe 0.1m/L Ca
 @ 0.2406 ml/min

1 ppm Np in syringe

1g calcite seed / 1L

Np init. = 250 μL of ~~spike~~ ^{MN 3/30/2000} stock sltn into 1L.

Objective: to precipitate calcite at a steady state growth
 rate, with constant aqueous Np concentration, according
 to the procedures described on pages 3-4 of this notebook
 and the conditions above.

Np-Ca syringe solution:

Bottle + cap: 24.52g (tare)

0.1m Ca sltn: 60.042g ^{MN 3/30/2000}

0.71 ml Np stock sltn (118 ppm): 60.74g

Np-Ca syringe sltn is
 \Rightarrow ~1.399 ppm Np

Mass of calcite seed: 1.0011g

This is the mass of calcite added to the
 reaction vessel, as described on page 82 of this
 notebook CNWRA controlled copy # 361.

3-30-2000

M. Nugent

condition electrode

filter 1 L of slth A, transfer to rxn vessel

Bubbling, stirring

Calibrate electrode

3:09 pH = 8.19 ATC = 23.2°C

3:23 8.28

3:26 8.29 5 ml 0.1 M Ca → 8.26

3:27 2 ml 0.1 M Ca → 8.26

3:28 7 ml 0.1 M HClO₃ → 8.67

3:44 8.57 Added seeds → 8.53

Attached syringes 0.1 M Ca & 0.1 M HClO₃ @ 0.2406 ml/min

3:45 8.48 ATC = 23.4°C

3:47 8.38

3:50 pH = 8.31

3:52 8.29

3:54 pH = 8.27

3:55 Added 0.25 ml UR stock Np + 0.25 ml NaOH
pH = 8.32 ← removed ATC probe to add Np slth.Changed syringes to
0.1 M Ca + 0.1 M Np Syringe. (2.5612 ml delivered already)

3:57 pH = 8.23

3:59 8.25 But ATC = 23.4 b/c in air SAMPLE #1 33000-1

4:06 8.26 ATC = 23.8

4:12 pH = 8.24

4:19 8.20 ATC = 24.0 Sample #2 33000-2

4:26 8.17/8.18

4:37 8.13

4:42 8.12 ATC = Sample #3 ~~33000~~ 33000-3

5:09 8.06

5:06 8.06 Refilled syringes

5:11 8.06 24.9°C Sample #4 33000-4

5:30 8.03 25.1

5:44 8.01/8.02

5:46 8.01 Sample #5 3300-5

3-30-2000

M. Nugent

556 pH = 8.01 ATC = 24.5 Sample #6
STOP! Filter.30.392 ml of each syringe added.
(total ml, including Ca syringe and Ca-Np syringe).Mass seed recovered: 1.0112 grams Recovered on 3/31/2000 by M. Nugent
Length of exp (Np doped part, only): 556 - 355: 2 hr 1 min = 121 min
This experiment went too long.

Calculate seed prep for LST analysis:

Sample Name: S3

Mass of Bottle + Cap: 16.8726 g

0.1149 g of seed + ~3.5 ml 1.0 N HCl. Total mass after

Seed dissolution: ~~20.4945 g~~ MN ~~20.4945 g~~ 20.4945 grams. etc.

Date prepared: 3/31/2000

Prepared by: Melissa Nugent.

Note:

- Syringe rate was for wgr = -2.24, not -2.8, like planned.
- Exp. was probably too long (>10% of mass of seeds (initial mass) was ppted.)

3/30/2000

85

Lst analysis for
aqueous samples for
3/30/2000

page 1/4

Count Conditions-

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

```
Static Controller: On      Luminescence Correction: Off
Colored Samples: n/a      Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75
```

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results

S#	Count	Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	999.99	33.27	1.10	39.84	1.00	1.96	4.52	846.80	Blank	B
2	39.64	17.74	12.96	19.42	12.76	110.15	3.05	568.55	Np33a	
3	54.90	10.26	17.72	11.49	17.18	79.06	3.08	580.90	34	
4	72.78	6.63	23.01	8.91	18.90	59.10	3.10	1095.57	35	
5	103.86	6.65	19.45	8.24	17.21	40.84	3.15	845.79	36	
6	99.11	7.76	17.23	10.01	14.72	42.90	3.14	1049.80	37	
7	97.34	7.90	17.10	9.96	14.91	43.70	3.14	894.72	38	
8	25.40	142.51	3.70	143.08	3.76	173.00	3.03	295.06	Np20a	
9	18.41	202.41	3.54	203.13	3.58	239.70	3.02	291.62	21	
10	14.87	247.36	3.52	248.06	3.55	297.10	3.02	284.10	22	
11	11.25	324.68	3.48	325.59	3.50	393.42	3.01	306.06	23	
12	10.01	360.83	3.48	360.46	3.51	442.00	3.01	280.97	24	

4/5/00 9:21:05 AM: Warning: User Has Modified Count Conditions.

Sample Name	Mass (all in grams) bottle+cap b+c+s	Sample b+c+ s+acid	a	TOTAL S+A	%sample	Subdivide the sample:				Subdivide the sample:					
						Name	b+c	b+c+s	Sample	Name	b+c	b+c+s	Sample		
														Np	
33000-1	16.7542 24.3492	7.5950	24.8514	0.5022	93.7979	Npa33	16.8651	19.8673	3.0022	Caa33	4.0421	4.5408	11.111	0.4987	7.0693
33000-2	16.9326 23.8194	6.8858	24.3227	0.5033	93.1895	Npa34	16.7936	19.791	2.9974	Caa34	4.0737	4.5689	11.356	0.4952	7.2823
33000-3	16.9055 24.477	7.5715	24.9794	0.5024	93.7775	Npa35	16.9329	19.9329	3.0000	Caa35	4.1623	4.6658	11.082	0.5035	6.9197
33000-4	16.6249 23.6254	7.0005	24.1287	0.5033	93.2927	Npa36	16.777	19.7775	3.0005	Caa36	4.0496	4.5439	11.121	0.4943	7.0712
33000-5	16.6432 24.0139	7.3707	24.5156	0.5017	93.6271	Npa37	16.9124	19.9077	2.9953	Caa37	4.0724	4.7054	11.128	0.6330	7.0556
33000-6	16.859 24.7606	7.9016	25.2617	0.5011	94.0364	Npa38	16.9474	19.9442	2.9968	Caa38	4.0317	4.5218	11.027	0.4901	6.9956

2/30/2000

M. Nugent

3/30/2000	M. Nugent
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4/5/00 9:58:51 AM
Protocol# 41 - Manual Np_Pa 3%2S.1

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 2
User: Nugent

13 783.00 3.75 15.14 5.44 11.50 0.00 0.00 1172.67 Np18a
4/5/00 9:21:47 AM: Warning: User Has Modified Count Conditions.
14 36.06 108.02 3.68 108.89 3.75 121.34 3.05 293.71 Np19a

terminated counting at 873 minutes

Page 2/4

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M. Nugent
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Sample Name	Mass (all in grams) bottle+cap b+c+s		Sample b+c+s	b+c+ s+acid	a	TOTAL S+a	%sample	Subdivide the sample:			Subdivide the sample:		
								Name	b+c	b+c+s	Name	b+c	b+c+s
33000-1	16.7542	24.3492	7.950	24.8514	0.5022	8.0972	93.7979	Npa33	16.8651	19.8673	Caa33	4.0421	4.5408
33000-2	16.9326	23.8194	6.8868	24.3227	0.5033	7.3901	93.1895	Npa34	16.7936	19.791	Caa34	4.0737	4.5689
33000-3	16.9055	24.477	7.5715	24.9794	0.5024	8.0789	93.7775	Npa35	16.9329	19.9329	Caa35	4.1623	4.6658
33000-4	16.6249	23.6254	7.0005	24.1287	0.5033	7.5038	93.2927	Npa36	16.777	19.7775	Caa36	4.0496	4.5439
33000-5	16.6432	24.0139	7.5707	24.5156	0.5017	7.8724	93.6271	Npa37	16.9124	19.9077	Caa37	4.0724	4.7054
33000-6	16.859	24.7606	7.9016	25.2617	0.5011	8.4027	94.0364	Npa38	16.9474	19.9442	Caa38	4.0317	4.5218

Nugent

3/30/2000

M. Nugent

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3/30/2000

M. Nugent

3/30/2000

page 4/4

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 15.53 Date Processed: 4/5/00 11:30:39 AM
14C Chi Square: 16.13 Date Processed: 4/5/00 11:30:39 AM
3H E²/B (0-18.6 keV and 1-18.6 keV): 255.40 Date Processed: 4/5/00 11:30:39 AM
14C E²/B (0-156 keV and 1-156 keV): 482.18 Date Processed: 4/5/00 11:30:39 AM
3H Efficiency (0-18.6 keV): 65.72 Date Processed: 4/5/00 11:30:39 AM
14C Efficiency (0-156 keV): 96.26 Date Processed: 4/5/00 11:30:39 AM
IPA Background Date Processed: 4/5/00 11:30:39 AM
3H Background CPM (0-18.6 keV): 16.72 Date Processed: 4/5/00 11:30:39 AM
14C Background CPM (0-156 keV): 24.50 Date Processed: 4/5/00 11:30:39 AM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

M. Nugent
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Sample Name	Mass (all in grams) bottle+cap b+c+s		Sample	b+c+s s+acid	a	TOTAL S+A	%sample	Subdivide the sample:			Subdivide the sample:			s+di
								Name	b+c	Np	Name	b+c	Ca	
33000-1	16.7542	24.3492	7.5950	24.8514	0.5022	8.0922	93.7979	Npa33	16.8651	19.8673	Sample	4.0421	4.5408	7.0693
33000-2	16.9326	23.8194	6.8868	24.3227	0.5033	7.3901	93.1895	Npa34	16.7936	19.791	Sample	4.0737	4.5689	7.2823
33000-3	16.9055	24.477	7.5715	24.9794	0.5024	8.0739	93.7775	Npa35	16.9329	19.9329	Sample	4.1623	4.6658	6.9197
33000-4	16.6249	23.6254	7.0665	24.1287	0.5033	7.5688	93.2927	Npa36	16.777	19.7775	Sample	4.0496	4.5439	7.0712
33000-5	16.6432	24.0139	7.3707	24.5156	0.5017	7.8724	93.6271	Npa37	16.9124	19.9077	Sample	4.0724	4.7054	7.0556
33000-6	16.859	24.7606	7.9016	25.2617	0.5011	8.4022	94.0364	Npa38	16.9474	19.9442	Sample	4.0317	4.5218	6.9956

M. Nugent

3/30/2000

M. Nugent

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3/30/2000

M. Nugent

86

3/30/2000 M. Nugent

87

3/30/2000
M. Nugent4/17/00 10:59:30 PM QuantaSmart (TM) - 1.10
Protocol# 01 - Manual Np_Pa 3%2S.1 Serial# 405314Page # 1
User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta

Report Name: Manual Np_Pa 3% 2sigma

Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa

Quench Indicator: SIS

External Std Terminator (sec): n/a

Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 300.00

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions Half Life

Units

Reference Date

Reference Time

Beta A

Beta B

Alpha

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	300.00	38.33	1.87	46.74	1.69	1.98	8.21	881.73	Blank B
2	0.49	9129.01	3.00	9124.69	3.00	9138.84	2.99	247.63	53
3	0.37	11958.96	3.01	11966.77	3.01	12127.75	2.99	243.58	51
4	0.99	2140.45	4.38	2140.13	4.39	4499.03	3.00	243.73	12999 SI
5	1.68	1160.48	4.60	1163.97	4.61	2655.76	3.00	261.80	120051

86

3/30/2000 M. Nugent

87

3/30/2000

M. Nugent

4/17/00 5:52:21 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/2

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 24.85 Date Processed: 4/17/00 5:52:21 PM
14C Chi Square: 15.18 Date Processed: 4/17/00 5:52:21 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 257.14 Date Processed: 4/17/00 5:52:21 PM
14C E²/B (0-156 keV and 1-156 keV): 470.73 Date Processed: 4/17/00 5:52:21 PM
3H Efficiency (0-18.6 keV): 65.73 Date Processed: 4/17/00 5:52:21 PM
14C Efficiency (0-156 keV): 96.65 Date Processed: 4/17/00 5:52:21 PM
IPA Background Date Processed: 4/17/00 5:52:21 PM
3H Background CPM (0-18.6 keV): 16.60 Date Processed: 4/17/00 5:52:21 PM
14C Background CPM (0-156 keV): 24.58 Date Processed: 4/17/00 5:52:21 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

nt

M. Nugent

4/10/2000
M. Nugent

4/10/2000

M. Nugent

Neptunium Calcite Coprecipitation

Try Np-Cc coprecipitation with:

 $\log r = -4.5$

0.01M syringes @ 0.0133 ml/min

1g calcite seed

250 μ L Np stock solution added to rxn vessel.

Objective: achieve steady state calcite growth with steady state Np according to the procedure on page 3-4 of this notebook (CNA Controlled copy 361) and the conditions above.

Np-Ca Syringe solution:

Should be 60ml 0.01M Ca, 1g of Np stock soln \leftarrow 118 ppm soln.

Bottle: 19.208g Cap 5.6720

1.0171g Np stock solution

60.1939g = Np stock + nanopure

Mass of calcite seed:

The rxn vessel.

 $1.0038g - 0.0012g = 1.0026g$ This is the mass of calcite added to

Summary: Experimental Conditions

Exp Date: 4/10/2000

Pre-Np spike:

Volume (L): 1L
 Solution A/B: B
 ml Ca added: 4.0 conc: 0.1
 ml HCO₃ added: 4.0 conc: 0.1
 Mass calcite added (g): 1.0026

Post-Np spike:

ml Np added: 0.25 conc: 118
 ml 0.32M NaOH added: 0.25
 Syringe Np conc (m/l): 1.0171g / 8.6ml 118 ppm / 60.1939g
 Syringe Ca conc (m/l): 0.01
 Pump Infusion Rate: 0.0133 ml/min
 Steady State pH: 8.36 ? calibration
 Temp, init. (Celcius): 21.5^{°C}
 Temp, final (Celcius): 25.6^{°C}

Post-experiment:

Mass calcite recovered: _____ (g)
 AA - Ca: _____ units: _____
 LSA - Np: _____ units: _____
 Sample Numbers: 39-46
 Seed # 54
 Calculated: 1.0731 ml added b4
 Ptn Rate: _____
 Np/Ca aq: _____ Np spike
 Np/Ca solid: _____
 Kd: _____
 58.23 ml added total Ca.

4/10/2000
M. Nugent

Condition electrode, fresh buffer solution
Filter 1 L slfn. B₃ bubbling stirring.

306 pm 8.25 = pH T slfn = 21.5°C

309 pm 8.19

310 ~~4 ml 0.1 M HCl~~ 0.1 M HCl pH = 8.17

312 4 ml 0.1 M HCl → pH = 8.54

314 pm Added seeds pH → 8.52

Attached syringes @ 0.0133 ml/min

316 pH = 8.47

320 8.41

323 8.38 T = 21.6

330 8.35

343 pH = 8.34 T = 21.8°C

346 8.34

348 8.34 Added Np 0.25 ml of stock slfn
+ 0.25 ml 0.32 M NaOH

pH = 8.33

349 8.33 Sample #1 41000-1

401 8.32 T = 22.0°C

426 pm 8.33

425 8.33 Stopped syringes and changed to Np-Ca syringe
1.0731 ml added

429 8.30 T = 22.5°C

520 8.33 T = 23.3°C

522 8.33 Sample #2 41000-2

4/11/2000 928 am pH = 8.33 T = 25°C Sample #3 41000-3

10 am 8.34

11 am 8.33

156 pm T = 25.0°C 8.32

250 8.32

444 pH = 8.33 T = 25.0°C

Calibrate electrode

449 8.36 Sample #4 41000-4

4/10/2000

M. Nugent

530 8.36

659 8.37 25.0°C

4/12/2000

1244 pm 8.38

1247 pm 8.38 T = 24.4°C Sample #5 41000-5

250 8.37

337 8.37/8.38

454 pm 8.37 Sample 24.4°C 41000-6 Sample #6.

759 pm 8.37 24.5°C

4/13/2000

923 am 8.36 T = 24.5°C

Calibrate electrode

935 am 8.35

937 am 8.35 Sample #7 41000-7

1131 pH = 8.36 T = 24.5°C

231 8.36

320 8.38 25.9°C

411 8.38 25.8°C #8 41000-8

412 STOP. Filtered solution to separate solids.

58.23 ml added from each syringe.
Allowed solids to dry in air overnight.

92 4/10/2000 M. Nugent

M. Nugent 4/10/2000 93

Sample Name	Mass (all in grams)		Sample	b+c+s+acid	a	TOTAL S+A	%sample	Subdivide the sample:		
	bottle+cap	b+c+s						Name	b+c	b+c+s
41000-1	16.8306	25.4358	8.6052	25.9402	0.5044	9.1096	94.4630	Npa39	16.8839	19.8943
41000-2	16.5875	24.4732	7.8857	24.9784	0.5052	8.3909	93.9792	Npa40	16.7797	19.7829
41000-3	16.9442	25.0357	8.0915	25.5406	0.5049	8.5964	94.1266	Npa41	16.6431	19.6452
41000-4	16.6036	24.6549	8.0513	25.1565	0.5016	8.5529	94.1353	Npa42	16.5657	19.5695
41000-5	16.7794	25.1338	8.3544	25.6373	0.5035	8.8579	94.3158	Npa43	16.872	19.881
41000-6	16.7212	24.0846	7.3634	24.5864	0.5018	7.8652	93.6200	Npa44	16.981	19.9835
41000-7	16.7749	25.0635	8.2886	25.5677	0.5042	8.7928	94.2658	Npa45	16.8723	19.8753
41000-8	16.7228	24.8725	8.1503	25.3751	0.5026	8.6529	94.1915	Npa46	16.6472	19.6502

4/14/00 2:16:35 PM

QuantaSmart (TM) - 1.10

Page # 1

Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta

Report Name: Manual Np_Pa 3% 2sigma

Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa

Quench Indicator: SIS

External Std Terminator (sec): n/a

Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 999.99

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS MESSAGES
1	999.99	33.93	1.09	40.51	0.99	2.14	4.33	834.16 Blank B
2	33.62	25.98	10.37	28.23	10.23	130.19	3.05	597.40 Np39a ← Np39a
3	33.76	35.98	8.06	37.60	8.16	129.59	3.05	435.97 Np40a
4	27.64	11.15	23.14	14.37	19.81	158.64	3.04	935.09 Np41a
5	24.84	10.03	26.77	11.62	25.17	176.85	3.04	718.17 Np42a
6	19.38	15.87	20.34	17.54	19.87	227.22	3.03	484.32 Np43a
7	18.34	16.02	20.73	18.81	19.24	240.17	3.03	778.77 Np44a
8	14.94	14.40	25.11	16.18	24.20	295.39	3.02	607.03 Np45a
9	14.77	14.48	25.13	16.16	24.37	298.88	3.02	746.87 Np46a

Calcite Seed prep for LST:

Mass bottle + cap: 16.6648g

0.1594g calcite + ~4ml 1.0N HCl - Total mass after

Seed dissolution: 20.7911 grams.

Preparation Date: 4/14/2000

Prepared by: M. Nugent

Seed Sample Name: S4

92 4/10/2000 M. Nugent

M. Nugent 4/10/2000 93

Sample		Mass (all in grams)							Subdivide the sample:			
Name		bottle+cap	b+c+s	Sample	b+c+	a	TOTAL	%sample	Name	b+c	b+c+s	Sample
					s+acid		S+A			Np		
41000-1		16.8306	25.4358	8.6052	25.9402	0.5044	9.1096	94.4630	Npa39	16.8839	19.8943	3.0104
41000-2		16.5875	24.4732	7.8857	24.9784	0.5052	8.3909	93.9792	Npa40	16.7797	19.7829	3.0032
41000-3		16.9442	25.0357	8.0915	25.5406	0.5049	8.5964	94.1266	Npa41	16.6431	19.6452	3.0021
41000-4		16.6036	24.6549	8.0513	25.1565	0.5016	8.5529	94.1353	Npa42	16.5657	19.5695	3.0038
41000-5		16.7794	25.1338	8.3544	25.6373	0.5035	8.8579	94.3158	Npa43	16.872	19.881	3.0090
41000-6		16.7212	24.0846	7.3634	24.5864	0.5018	7.8652	93.6200	Npa44	16.981	19.9835	3.0025
41000-7		16.7749	25.0635	8.2886	25.5677	0.5042	8.7928	94.2658	Npa45	16.8723	19.8753	3.0030
41000-8		16.7228	24.8725	8.1503	25.3751	0.5026	8.6529	94.1915	Npa46	16.6472	19.6502	3.0030

4/13/00 6:17:36 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

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Calibration Information

Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 21.79 Date Processed: 4/13/00 6:17:36 PM
14C Chi Square: 13.18 Date Processed: 4/13/00 6:17:36 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 261.91 Date Processed: 4/13/00 6:17:36 PM
14C E²/B (0-156 keV and 1-156 keV): 470.61 Date Processed: 4/13/00 6:17:36 PM
3H Efficiency (0-18.6 keV): 65.67 Date Processed: 4/13/00 6:17:36 PM
14C Efficiency (0-156 keV): 96.89 Date Processed: 4/13/00 6:17:36 PM
IPA Background Date Processed: 4/13/00 6:17:36 PM
3H Background CPM (0-18.6 keV): 16.35 Date Processed: 4/13/00 6:17:36 PM
14C Background CPM (0-156 keV): 24.80 Date Processed: 4/13/00 6:17:36 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

Calcite Seed prep for Lst:

Seed Sample Name: S4

Mass bottle + cap: 16.6648g

0.1594g calcite + ~4ml 1.0N HCl - Total mass after

Seed dissolution: 20.7911 grams.

Preparation Date: 4/14/2000

Prepared by: M. Nugent

M. Nugent

92 4/10/2000 M. Nugent

M. Nugent 4/10/2000 93

Sample		Mass (all in grams)							Subdivide the sample:		
Name	bottle+cap	b+c+s	Sample	b+c+s+acid	a	TOTAL S+A	%sample	Name	b+c	b+c+s	Sample
41000-1	16.8306	25.4358	8.6052	25.9402	0.5044	9.1096	94.4630	Npa39	16.8839	19.8943	3.0104
41000-2	16.5875	24.4732	7.8857	24.9784	0.5052	8.3909	93.9792	Npa40	16.7797	19.7829	3.0032
41000-3	16.9442	25.0357	8.0915	25.5406	0.5049	8.5964	94.1266	Npa41	16.6431	19.6452	3.0021
41000-4	16.6036	24.6549	8.0513	25.1565	0.5016	8.5529	94.1353	Npa42	16.5657	19.5695	3.0038
41000-5	16.7794	25.1338	8.3544	25.6373	0.5035	8.8579	94.3158	Npa43	16.872	19.881	3.0090
41000-6	16.7212	24.0846	7.3634	24.5864	0.5018	7.8652	93.6200	Npa44	16.981	19.9835	3.0025
41000-7	16.7749	25.0635	8.2886	25.5677	0.5042	8.7928	94.2658	Npa45	16.8723	19.8753	3.0030
41000-8	16.7228	24.8725	8.1503	25.3751	0.5026	8.6529	94.1915	Npa46	16.6472	19.6502	3.0030

M. Nugent

Calcite Seed prep for LST: Seed Sample Name: S4
Mass bottle + cap: 16.6648g
0.1594g calcite + ~4ml 1.0N HCl - Total mass after
Seed dissolution: 20.7911 grams.
Preparation Date: 4/14/2000
Prepared by: M. Nugent

4/17/00 10:59:30 PM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1:Serial# 405314

Page # 1
User: Nugent

Disssolved
Seed

Page 3/4

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00
Quench Set:
Count Time (min): 300.00
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results									
S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	300.00	38.33	1.87	46.74	1.69	1.98	8.21	881.73	Blank B
2	0.49	9129.01	3.00	9124.69	3.00	9138.84	2.99	247.63	S3
3	0.37	11958.96	3.01	11966.77	3.01	12127.75	2.99	243.58	S1
4	0.99	2140.45	4.38	2140.13	4.39	4499.03	3.00	243.73	12999 S1
5	1.68	1160.48	4.60	1163.97	4.61	2655.76	3.00	261.80	12051

M. Nugent
M. Nugent
M. Nugent
M. Nugent

Sample Name	Mass (all in grams)		Sample	b+c+s+acid	a	TOTAL S+A	%sample	Subdivide the sample:			Sample
	bottle+cap	b+c+s						Name	b+c	b+c+s	
41000-1	16.8306	25.4358	8.6052	25.9402	0.5044	9.1096	94.4630	Npa39	16.8839	19.8943	3.0104
41000-2	16.5875	24.4732	7.8857	24.9784	0.5052	8.3909	93.9792	Npa40	16.7797	19.7829	3.0032
41000-3	16.9442	25.0357	8.0915	25.5406	0.5049	8.5964	94.1266	Npa41	16.6431	19.6452	3.0021
41000-4	16.6036	24.6549	8.0513	25.1565	0.5016	8.5529	94.1353	Npa42	16.5657	19.5695	3.0038
41000-5	16.7794	25.1338	8.3544	25.6373	0.5035	8.8579	94.3158	Npa43	16.872	19.881	3.0090
41000-6	16.7212	24.0846	7.3634	24.5864	0.5018	7.8652	93.6200	Npa44	16.981	19.9835	3.0025
41000-7	16.7749	25.0635	8.2886	25.5677	0.5042	8.7928	94.2658	Npa45	16.8723	19.8753	3.0030
41000-8	16.7228	24.8725	8.1503	25.3751	0.5026	8.6529	94.1915	Npa46	16.6472	19.6502	3.0030

4/17/00 5:52:21 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

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Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 24.85 Date Processed: 4/17/00 5:52:21 PM
14C Chi Square: 15.18 Date Processed: 4/17/00 5:52:21 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 257.14 Date Processed: 4/17/00 5:52:21 PM
14C E²/B (0-156 keV and 1-156 keV): 470.73 Date Processed: 4/17/00 5:52:21 PM
3H Efficiency (0-18.6 keV): 65.73 Date Processed: 4/17/00 5:52:21 PM
14C Efficiency (0-156 keV): 96.65 Date Processed: 4/17/00 5:52:21 PM
IPA Background Date Processed: 4/17/00 5:52:21 PM
3H Background CPM (0-18.6 keV): 16.60 Date Processed: 4/17/00 5:52:21 PM
14C Background CPM (0-156 keV): 24.58 Date Processed: 4/17/00 5:52:21 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

Calcite Seed prep for LST:

Seed Sample Name: S4

Mass bottle + cap: 16.6648g

0.1594g calcite + ~4ml 1.0N HCl - Total mass after

Seed dissolution: 20.7911 grams.

Preparation Date: 4/14/2000

Prepared by: M. Nugent

M. Nugent

M. Nugent

M. Nugent

M. Nugent

94

4/19/2000 M. Nugent

MN 4/19/2000 95

Neptunium Calcite Coprecipitation 4/19/2000

4/19/2000

Try $\log r = -3.0$

lg of calcite seeds / 1L rxn vessel

use 0.1m syringes ± 0.0422 ml/min

use 3ppm Np in syringe sltn. (2.5ml/15ml)

use 250 μ l of 118 Np stock sltn in rxn vessel

Objective: precipitate calcite at steady state with steady state Np aq, as described by the procedure on pages 3 and 4 of this notebook, CNWRA controlled Copy #361, and the conditions listed above.

Np - Ca Syringe: ppm

2.5095 g 118 Np sltn

Added syringe sltn (0.1m) to 15.2320 g

$$C_1 V_1 = C_2 V_2$$

$$2.5095 \times 118 = 15.2320 \times C_2 \quad C_2 = 19.44 \text{ ppm!}$$

Mass calcite added:

1.0023g This is the mass added to the rxn vessel.

Summary: Experimental Conditions

Exp Date: 4/19/2000

Pre-Np spike:

Volume (L): 1L

Solution A/B: A

ml Ca added: 7 conc: 0.1

ml HCO₃ added: 7 conc: 0.1

Mass calcite added (g): 1.0023g

Post-Np spike:

ml Np added: 0.25 conc: 118 ppm

ml 0.32m NaOH added: 0.25

Syringe Np conc (m/l): ~19.44! cop. This is too high

Syringe Ca conc (m/l): 0.1m

Pump Infusion Rate: 0.2406 ml/min

Steady State pH: ~8.09

Temp, init. (Celcius): 22.5°C

Temp, final (Celcius): 22.9°C

Post-experiment:

Mass calcite recovered: 0.8667(g)

AA - Ca: units:

LSA - Np: units:

Sample Numbers: 47-52

Seed SS

Calculated:

Pttm Rate:

Np/Ca aq:

Np/Ca solid:

Kd:

14.949 ml added

4/19/2000

M. Nugent

Conditioned electrode. calibrate electrode.

Filtered IL sltn (A) bubbling, stirring.

306

pH = 8.35

316

8.34 22.5°C

317

7 ml 0.1 mCa → 8.31

319

7 ml 0.1 m HCl → 8.59

321

8.64 Added seeds 8.52

323

8.49

325

Attached syringe 0.1 mCa 0.1 m HCl @ 0.2 + 0.6 ml/min

327

8.57

328

8.35 sample #1 41900-1

330

8.32

332

8.29 Added Np + NaOH

333

8.29 Attached syringe NpCa → 8.25

335

8.24 sample #2 41900-2

336

8.16 Added < 250 µl NaOH (0.32M) to T pit
pH → 8.28

339

8.20

341

8.14

349

8.08 T = 22.5°C sample #3 41900-3

351

8.08

355

8.08

358

8.08 22.5°C sample #4 41900-4

411

8.09 T = 22.8

420

8.10 22.9°C

421

8.10 sample #5 41900-5

425

8.1

429

8.11 22.7 MN 4/19/2000

430

STOP

14.949 ml added from both the Ca and HCl syringes

Mass of seed recovered: 0.8667

bad recovery - filter stuck to petri dish that it
was being stored in. Lost ~0.1g
Recovered 4/20/2000 by M. Nugent.

4/19/2000

con't
M. Nugent.

Calcite seed prep for LSA:

Bottle + cap (mass): 16.7094

Seed: 0.1011 g

Add: ~3 ml 1.0N HCl

Mass, Seed + Acid: 19.8123 g

3.1029 ml

(after dissolution)

Sample Name S5

Seed prep performed on 4/20/2000 by
M. Nugent.

Sample Name	Mass (all in grams) bottle+cap b+c+s	Sample b+c+s	a	TOTAL S+A	%sample	Subdivide the sample:			Calcium
						Name	b+c	b+c+s	
41900-1	16.7766	20.2911	3.5145	3.7634	93.3863	Caa47	4.0518	4.5509	11.071
41900-2	16.6132	25.4344	8.8212	9.4404	93.4410	Caa48	4.056	4.5528	11.037
41900-3	16.5722	24.4554	7.8832	8.3864	93.9998	Caa49	4.0413	4.526	11.016
41900-4	16.9006	25.4354	8.5348	9.0381	94.4314	Caa50	4.0625	4.5553	11.064
41900-5	16.5825	24.5163	7.9338	8.4360	94.0469	Caa51	4.037	4.5329	11.039
41900-6	16.834	24.9684	8.1344	8.6372	94.1787	Caa52	4.062	4.5558	11.06

mfg

4/19/2000 M. Nugent

4/21/00 9:39:04 AM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314Page # 1
User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00
Quench Set:
Count Time (min): 999.99
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off
Regions Half Life Units Reference Date Reference Time
Beta A
Beta B
Alpha

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS MESSAGES
1	999.99	34.04	1.08	40.68	0.99	2.02	4.46	841.53 Blank B
2	32.54	44.08	7.08	45.92	7.16	134.62	3.05	368.98 Np 48a
3	12.52	46.47	10.94	47.25	11.25	352.94	3.02	287.50 Np 49a
4	8.88	57.62	11.17	60.33	11.20	498.44	3.01	418.86 Np 50a
5	4.93	109.57	9.86	111.04	10.00	900.82	3.00	285.53 Np 51a
6	4.39	156.39	8.43	156.58	8.57	1011.42	3.00	293.42 Np 52a

M. Nugent 4/19/2000

4/28/00 11:40:09 PM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314Page # 1
User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00
Quench Set:
Count Time (min): 500.00
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off
Regions Half Life Units Reference Date Reference Time
Beta A
Beta B
Alpha

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS MESSAGES
1	500.00	37.28	1.46	45.46	1.33	2.18	6.05	890.56 Blank B
2	1.23	17447.27	1.37	17446.41	1.37	3623.02	3.00	210.13 SS
3	0.67	6455.26	3.05	6456.04	3.05	6676.92	2.99	221.18 SS
4	1.09	5253.54	2.65	5251.79	2.65	4112.49	2.99	230.25 SS
5	1.27	5940.67	2.31	5937.22	2.31	3502.54	3.00	246.44 SS

LST analysis of
Seeds (dissolved)
for 4/19/2000 page 1/2

M. Nugent

4/19/2000 M. Nugent

4/20/00 3:41:54 PM

SNC Protocol

QuantaSmart (TM) - 1.10

Serial# 405314

Page # 1

Page
2/2

Calibration Information

Software Version IC: 2.09

Software Version EC: 1.10

Instrument Model: Tri-Carb 3100TR

Instrument Serial Number: 405314

3H Chi Square: 16.94 Date Processed: 4/20/00 3:41:53 PM

14C Chi Square: 15.39 Date Processed: 4/20/00 3:41:53 PM

3H E^2/B (0-18.6 keV and 1-18.6 keV): 241.70 Date Processed: 4/20/00 3:41:53 PM

14C E^2/B (0-156 keV and 1-156 keV): 456.28 Date Processed: 4/20/00 3:41:53 PM

3H Efficiency (0-18.6 keV): 65.79 Date Processed: 4/20/00 3:41:53 PM

14C Efficiency (0-156 keV): 96.40 Date Processed: 4/20/00 3:41:53 PM

IPA Background Date Processed: 4/20/00 3:41:53 PM

3H Background CPM (0-18.6 keV): 17.75 Date Processed: 4/20/00 3:41:53 PM

14C Background CPM (0-156 keV): 26.08 Date Processed: 4/20/00 3:41:53 PM

3H Calibration DPM: 285000

3H Reference Date: 10/29/99

14C Calibration DPM: 134100

===== Errors and Warnings =====

===== End of Errors and Warnings =====

M. Nugent

M. Na

M. Nugent 4/19/2000

4/28/00 3:10:48 PM

SNC Protocol

QuantaSmart (TM) - 1.10

Serial# 405314

Page # 1

page 2/2

Calibration Information

Software Version IC: 2.09

Software Version EC: 1.10

Instrument Model: Tri-Carb 3100TR

Instrument Serial Number: 405314

3H Chi Square: 22.85 Date Processed: 4/28/00 3:10:48 PM

14C Chi Square: 28.30 Date Processed: 4/28/00 3:10:48 PM

3H E^2/B (0-18.6 keV and 1-18.6 keV): 244.90 Date Processed: 4/28/00 3:10:48 PM

14C E^2/B (0-156 keV and 1-156 keV): 452.29 Date Processed: 4/28/00 3:10:48 PM

3H Efficiency (0-18.6 keV): 65.56 Date Processed: 4/28/00 3:10:48 PM

14C Efficiency (0-156 keV): 96.48 Date Processed: 4/28/00 3:10:48 PM

IPA Background Date Processed: 4/28/00 3:10:48 PM

3H Background CPM (0-18.6 keV): 17.33 Date Processed: 4/28/00 3:10:48 PM

14C Background CPM (0-156 keV): 25.60 Date Processed: 4/28/00 3:10:48 PM

3H Calibration DPM: 285000

3H Reference Date: 10/29/99

14C Calibration DPM: 134100

===== Errors and Warnings =====

===== End of Errors and Warnings =====

M. Nugent
M. Naug

4/19/2000
M. Nugent

Notes: The syringe Np concentration was too high to achieve the desired results.

4/19/2000
M. Nugent

This
page
left
Blank

4/20/2000 M. Nugent

Summary: Experimental Conditions

Pre-Np spike:

Volume (L): $\frac{1L}{B}$
 Solution A/B: $\frac{B}{B}$
 ml Ca added: $\frac{4ml}{conc: 0.1 mL}$
 ml HCO₃ added: $\frac{4ml}{conc: 0.1 mL}$
 Mass calcite added (g): 0.998g

Post-Np spike:

ml Np added: 0.25 conc: 18 ppm
 ml 0.32m NaOH added: 0.25
 Syringe Np conc (m/l): 0.963 ppm
 Syringe Ca conc (m/l): 0.01
 Pump Infusion Rate: 0.0133 ml/min
 Steady State pH: 8.37
 Temp, init. (Celcius): 22°C
 Temp, final (Celcius): 26.6°C

Exp Date: 4/20/2000

Post-experiment:

Mass calcite recovered: 0.7646(g)
 AA - Ca: _____ units:
 LSA - Np: _____ units:
 Sample Numbers: 54 - 60
 Seed 56

Calculated:

Pttn Rate: _____
 Np/Ca aq: _____
 Np/Ca solid: _____
 Kd: _____
 35.097ml of Ca added,
 1.2133ml added before Np.

not necessary
 any
 calculate
 4/20/2000

Neptunium - Calcite Coprecipitation 4/20/2000

M. Nugent

Try logr = -4.5

1 ppm Np in Np-Ca syringe soln.

Infuse rate = 0.0133 ml/min

1g calcite seed

250 µl Np 118 ppm stock soln.

Objective: precipitate calcite at steady state with a steady state Np concentration. The procedure and equipment described on pages 2 and 3 of this notebook, CNWRA controlled copy #361, will be used.

Np-Ca syringe solution:

Want 60 ml @ 1 ppm. Want to add 0.5085g to 60 ml of 0.01m Ca syringe soln.

Added 0.5015g 118 Np solution to bottle

~~Bottle cap~~ Bottle = 19.2187g

Filled with 0.01m syringe solution to 61.4441 grams.

$$118 \times 0.5015 = C \times 61.4441$$

$$C = 0.963 \text{ ppm} \quad 4.0625 \times 10^{-6} \text{ ml}$$

Calcite Seeds:

10/14/2000 MN 4/20/2000

$$1.0044 \text{ g} - 0.0064 \text{ g} = 0.998 \text{ grams} \quad \text{Mass of calcite seed actually added to reaction vessel}$$

Mass of Calcite Seed
 in weigh boat before
 addition to reaction
 vessel

Mass of Calcite Seed
 in weigh boat after
 addition to the reaction
 vessel

106
M. Nugent 4/20/2000

4/25/00 6:21:05 PM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 1
User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00

Quench Set:
Count Time (min): 999.99
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results	S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	999.99	34.56	1.08	41.26	0.98	2.01	4.47	838.68	Blank	B
2	35.61	20.09	12.47	21.47	12.51	122.90	3.05	466.60	Np54a	
3	51.04	7.39	25.05	8.66	23.32	85.08	3.07	559.58	Np55a	
4	41.67	38.40	6.96	39.95	7.06	104.64	3.06	357.81	Np56a	
5	67.23	7.86	20.75	9.15	19.45	64.10	3.10	627.51	Np57a	
6	69.32	6.92	23.01	8.81	19.84	62.12	3.10	935.74	Np58a	
7	73.71	6.73	22.93	8.38	20.18	58.28	3.11	783.63	Np59a	
8	77.27	5.20	28.51	6.66	24.42	55.51	3.11	886.70	Np60a	
Missing vial 9.									EMPTY	
10	36.96	28.21	9.33	30.98	9.12	118.29	3.05	547.98	Np61a	
11	45.37	6.99	27.90	9.54	22.58	95.94	3.06	1156.58	Np62a	
12	64.12	4.20	38.09	5.92	29.80	67.32	3.09	1190.90	Np63a	
13	96.32	4.00	32.97	5.94	24.56	44.13	3.14	1354.62	Np64a	

4/20/2000 107
M. Nugent

4/28/00 11:40:09 PM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 1
User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00

Quench Set:
Count Time (min): 500.00
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results	S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	500.00	37.28	1.46	45.46	1.33	2.18	6.05	890.56	Blank	B
2	1.23	17447.27	1.37	17446.41	1.37	3623.02	3.00	210.13	SS	
3	0.67	6455.26	3.05	6456.04	3.05	6676.92	2.99	221.18	SS	
4	1.09	5253.54	2.65	5251.79	2.65	4112.49	2.99	230.25	SS	
5	1.27	5940.67	2.31	5937.22	2.31	3502.54	3.00	246.44	SS	

106
M. Nugent 4/20/2000

4/25/00 6:21:05 PM QuantaSmart (TM) - 1.10
Protocol 41 - Manual Np_Pa 3%2S.1:Serial# 405314

Page # 2
User: Nugent

Page 2/A

14 108.79 4.74 26.56 6.51 21.29 38.84 3.16 1088.77 Np65a

4/20/2000 107
M. Nugent

4/28/00 3:10:48 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/2

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 22.85 Date Processed: 4/28/00 3:10:48 PM
14C Chi Square: 28.30 Date Processed: 4/28/00 3:10:48 PM
3H E^2/B (0-18.6 keV and 1-18.6 keV): 244.90 Date Processed: 4/28/00 3:10:48 PM
14C E^2/B (0-156 keV and 1-156 keV): 452.29 Date Processed: 4/28/00 3:10:48 PM
3H Efficiency (0-18.6 keV): 65.56 Date Processed: 4/28/00 3:10:48 PM
14C Efficiency (0-156 keV): 96.48 Date Processed: 4/28/00 3:10:48 PM
IPA Background Date Processed: 4/28/00 3:10:48 PM
3H Background CPM (0-18.6 keV): 17.33 Date Processed: 4/28/00 3:10:48 PM
14C Background CPM (0-156 keV): 25.60 Date Processed: 4/28/00 3:10:48 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

M. Nugent

M. Nugent

106
M. Nugent 4/20/2000

4/25/00 7:52:54 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

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Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 23.16 Date Processed: 4/25/00 7:52:54 PM
14C Chi Square: 22.07 Date Processed: 4/25/00 7:52:54 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 263.59 Date Processed: 4/25/00 7:52:54 PM
14C E²/B (0-156 keV and 1-156 keV): 469.63 Date Processed: 4/25/00 7:52:54 PM
3H Efficiency (0-18.6 keV): 65.60 Date Processed: 4/25/00 7:52:54 PM
14C Efficiency (0-156 keV): 96.41 Date Processed: 4/25/00 7:52:54 PM
IPA Background Date Processed: 4/25/00 7:52:54 PM
3H Background CPM (0-18.6 keV): 16.20 Date Processed: 4/25/00 7:52:54 PM
14C Background CPM (0-156 keV): 24.93 Date Processed: 4/25/00 7:52:54 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

4/20/2000 107
M. Nugent

4/28/00 3:10:48 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/2

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 22.85 Date Processed: 4/28/00 3:10:48 PM
14C Chi Square: 28.30 Date Processed: 4/28/00 3:10:48 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 244.90 Date Processed: 4/28/00 3:10:48 PM
14C E²/B (0-156 keV and 1-156 keV): 452.29 Date Processed: 4/28/00 3:10:48 PM
3H Efficiency (0-18.6 keV): 65.56 Date Processed: 4/28/00 3:10:48 PM
14C Efficiency (0-156 keV): 96.48 Date Processed: 4/28/00 3:10:48 PM
IPA Background Date Processed: 4/28/00 3:10:48 PM
3H Background CPM (0-18.6 keV): 17.33 Date Processed: 4/28/00 3:10:48 PM
14C Background CPM (0-156 keV): 25.60 Date Processed: 4/28/00 3:10:48 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

M. Nugent

M. Nugent

106
M. Nugent 4/20/2000

4/20/2000 107
M. Nugent

4/24/00 12:35:09 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

Page 4/4

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 14.60 Date Processed: 4/24/00 12:35:09 PM
14C Chi Square: 15.16 Date Processed: 4/24/00 12:35:09 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 263.01 Date Processed: 4/24/00 12:35:09 PM
14C E²/B (0-156 keV and 1-156 keV): 485.01 Date Processed: 4/24/00 12:35:09 PM
3H Efficiency (0-18.6 keV): 65.96 Date Processed: 4/24/00 12:35:09 PM
14C Efficiency (0-156 keV): 96.77 Date Processed: 4/24/00 12:35:09 PM
IPA Background Date Processed: 4/24/00 12:35:09 PM
3H Background CPM (0-18.6 keV): 16.35 Date Processed: 4/24/00 12:35:09 PM
14C Background CPM (0-156 keV): 24.38 Date Processed: 4/24/00 12:35:09 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

4/28/00 3:10:48 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/2

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 22.85 Date Processed: 4/28/00 3:10:48 PM
14C Chi Square: 28.30 Date Processed: 4/28/00 3:10:48 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 244.90 Date Processed: 4/28/00 3:10:48 PM
14C E²/B (0-156 keV and 1-156 keV): 452.29 Date Processed: 4/28/00 3:10:48 PM
3H Efficiency (0-18.6 keV): 65.56 Date Processed: 4/28/00 3:10:48 PM
14C Efficiency (0-156 keV): 96.48 Date Processed: 4/28/00 3:10:48 PM
IPA Background Date Processed: 4/28/00 3:10:48 PM
3H Background CPM (0-18.6 keV): 17.33 Date Processed: 4/28/00 3:10:48 PM
14C Background CPM (0-156 keV): 25.60 Date Processed: 4/28/00 3:10:48 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

M. Nugent

M. Nugent

M. Nugent

M. Nugent

M. Nugent

M. Nugent

Pages 108 Through 110 Are Intentionally
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4/24/2000

Neptunium Calcite Coprecipitation

Try Log r = -3.2

H. N. agent

- syringe rate 0.0266 ml/min w/ 0.1m syringe
- try 3ppm Np in syringe
- lg calcite seeds
- 250 μ L Np 118ppm solution in rxn. vessel

Objective to obtain steady state Np_{aq} and steady state calcite growth rates using the procedure described on pages 2 and 3 of this notebook, controlled copy #361, CNWRA.

Np-Ca syringe solution

want 3ppm in 15ml, want to add 0.254 ml of 118ppm Np solution to 15ml total volume.

Bottle + cap: 7.8523g
tare

0.2470g of 118ppm Np sltn

Added 0.1m Ca/Nac2=0.05m sltn to 15.0126g.

mass, seeds before addition
to rxn vessel

Mass of seeds remaining in weight boat after addition to reaction vessel.

Calcite seeds: $(1.0074 - 0.0047)g$ added = 1.0027 grams.

Summary: Experimental Conditions

Exp Date: 4/24/2000

Pre-Np spike:

Volume (L): $\frac{1L}{A}$
Solution A/B:
ml Ca added: 7ml conc: 0.1mL
ml HCO3 added: 7ml conc: 0.1mL
Mass calcite added (g): 1.0027g

Post-Np spike:

ml Np added: 0.25 conc: 118ppm
ml 0.32m NaOH added: 0.25
Syringe Np conc (m/l): 0.2470ml 118ppm into 15.0126ml
Syringe Ca conc (m/l): 0.1mL
Pump Infusion Rate: 0.0266 ml/min
Steady State pH: 8.36
Temp, init. (Celcius): 23.0°C
Temp, final (Celcius): 25.8°C

Post-experiment:

Mass calcite recovered 0.9135(g)
AA - Ca: _____ units:
LSA - Np: _____ units:
Sample Numbers: 61-65
Seed 57

Calculated:

Ptm Rate: not necessary 4/28/2000
Np/Ca aq: to calculate
Np/Ca solid: _____
Kd: _____

3.1355ml added, 0.7853g Np.

M. N. agent

112

4/24/2000 M. Nugent

Filtered 1L Solution A - Stirring & bubbling air. Calibrate pH Electrode.

349 T = 23.0°C pH = 7.77

352 7ml 0.1M Ca → 7.69

353 8.53 7ml 0.1M HCO₃ → 8.57

402 8.38

→ 0.1M Ca and 0.1M HCO₃

403 8.38 Added seeds: Attached syringes @ 0.0266 ml/m

↳ pH → 8.35 after seeds.

406 8.32

408 8.32

411 pH = 8.32 T = 23.5°C

420 pH = 8.35

422 8.35

427 8.35

430 8.35

431 8.35 Added Np + NaOH pH → 8.48

432 8.36 Sample #1 42400-1

Stop syringes @ 0.7853 ml

changed to Np-Ca Syringe

434 8.35

435 T = 24.0 pH = 8.36

439 8.36

442 8.37 24.1°C

446 8.38 24.3°C

451 8.38 Sample #2 42400-2

457 8.37

527 25.3°C 8.36

529 8.36/8.37 Sample #3 42400-3

531 8.34 4/24/2000 M. Nugent 4/24/2000-4

553 Sample #4 42400-4 T = 25.8°C pH = 8.34

558 8.34

Np-Ca syringe may be leaking - will stop exp. early
and check sth Ca/Np concentrations

601 8.34/5 Sample #5 42400-5

603 8.35

STOP

filter sth and save solids, allow solids to dry in air.

4/24/2000 113

M. Nugent

3.1355 ml of solution added from Ca syringe and from HCO₃ syringe
Mass of seed recovered: 0.9135 g recovered on 4/25/2000.

Prep of Seed for LSA analysis 4/25/2000 Sample Name: S7

Bottle + Cap: 16.6094 M. Nugent
tare

Mass of calcite added: 0.1133 g

Added ~ 3ml of acid (1.0N HCl):

Total mass of calcite + acid after dissolution: 3.1326 g.

Sample Name	Mass (all in grams) bottle+cap b+c+s		Sample b+c+s+acid	a	TOTAL S+A	%sample	Subdivide the sample:		
							Name	b+c	b+c+s
42400-1	16.5962	23.8096	7.2134	24.3156	0.506	7.7194	93.4451 Npa61	16.8788	19.9051
42400-2	16.8092	23.846	7.0368	24.3495	0.5035	7.5403	93.3225 Npa62	16.734	19.7553
42400-3	16.7179	24.6802	7.9623	25.1795	0.4993	8.4616	94.0992 Npa63	16.781	19.7985
42400-4	16.9337	25.4245	8.4908	25.928	0.5035	8.9943	94.4020 Npa64	16.5947	19.6087
42400-5	16.72	25.8077	9.0877	26.3131	0.5054	9.5931	94.7316 Npa65	16.8873	19.9051

Solid sent for SEM analysis 4/28/2000

Results will be recorded after analysis is performed. Since this may take more than one day, experiments will be performed in the meantime.
See pages 145-150 for SEM results - in this notebook.

114
4/24/2000
M. Nugent

4/25/00 6:21:05 PM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 1
User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00

Quench Set:
Count Time (min): 999.99
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	999.99	34.56	1.08	41.26	0.98	2.01	4.47	838.68	Blank B
2	35.61	20.09	12.47	21.47	12.51	122.90	3.05	466.60	Np54a
3	51.04	7.39	25.05	8.66	23.32	85.08	3.07	559.58	Np55a
4	41.67	38.40	6.96	39.95	7.06	104.64	3.06	357.81	Np56a
5	67.23	7.86	20.75	9.15	19.45	64.10	3.10	627.51	Np57a
6	69.32	6.92	23.01	8.81	19.84	62.12	3.10	935.74	Np58a
7	73.71	6.73	22.93	8.38	20.18	58.28	3.11	783.63	Np59a
8	77.27	5.20	28.51	6.66	24.42	55.51	3.11	886.70	Np60a
Missing vial 9.									EMPTY
10	36.96	28.21	9.33	30.98	9.12	118.29	3.05	547.98	Np61a
11	45.37	6.99	27.90	9.54	22.58	95.94	3.06	1156.58	Np62a
12	64.12	4.20	38.09	5.92	29.80	67.32	3.09	1190.90	Np63a
13	96.32	4.00	32.97	5.94	24.56	44.13	3.14	1354.62	Np64a

LST analysis of
aqueous Np for
4/24/2000

page 1/4

4/24/2000 115
M. Nugent

4/28/00 11:40:09 PM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 1
User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00

Quench Set:
Count Time (min): 500.00
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	500.00	37.28	1.46	45.46	1.33	2.18	6.05	890.56	Blank B
2	1.23	17447.27	1.37	17446.41	1.37	3623.02	3.00	210.13	SS
3	0.67	6455.26	3.05	6456.04	3.05	6676.92	2.99	221.18	SS
4	1.09	5253.54	2.65	5251.79	2.65	4112.49	2.99	230.25	SS
5	1.27	5940.67	2.31	5937.22	2.31	3502.54	3.00	246.44	SS

LST analysis for
seeds (dissolved)
for 4/24/2000

page 1/2

114
4/24/2000
M. Nugent

4/25/00 6:21:05 PM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 2
User: Nugent

14 108.79 4.74 26.56 6.51 21.29 38.84 3.16 1088.77 Np65a

page 2/4

4/24/2000 115
M. Nugent

4/28/00 3:10:48 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/2

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 22.85 Date Processed: 4/28/00 3:10:48 PM
14C Chi Square: 28.30 Date Processed: 4/28/00 3:10:48 PM
3H E^2/B (0-18.6 keV and 1-18.6 keV): 244.90 Date Processed: 4/28/00 3:10:48 PM
14C E^2/B (0-156 keV and 1-156 keV): 452.29 Date Processed: 4/28/00 3:10:48 PM
3H Efficiency (0-18.6 keV): 65.56 Date Processed: 4/28/00 3:10:48 PM
14C Efficiency (0-156 keV): 96.48 Date Processed: 4/28/00 3:10:48 PM
IPA Background Date Processed: 4/28/00 3:10:48 PM
3H Background CPM (0-18.6 keV): 17.33 Date Processed: 4/28/00 3:10:48 PM
14C Background CPM (0-156 keV): 25.60 Date Processed: 4/28/00 3:10:48 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

114
4/24/2000
M. Nugent

4/25/00 7:52:54 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

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Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 23.16 Date Processed: 4/25/00 7:52:54 PM
14C Chi Square: 22.07 Date Processed: 4/25/00 7:52:54 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 263.59 Date Processed: 4/25/00 7:52:54 PM
14C E²/B (0-156 keV and 1-156 keV): 469.63 Date Processed: 4/25/00 7:52:54 PM
3H Efficiency (0-18.6 keV): 65.60 Date Processed: 4/25/00 7:52:54 PM
14C Efficiency (0-156 keV): 96.41 Date Processed: 4/25/00 7:52:54 PM
IPA Background Date Processed: 4/25/00 7:52:54 PM
3H Background CPM (0-18.6 keV): 16.20 Date Processed: 4/25/00 7:52:54 PM
14C Background CPM (0-156 keV): 24.93 Date Processed: 4/25/00 7:52:54 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

4/24/2000 115
M. Nugent

4/28/00 3:10:48 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

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Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 22.85 Date Processed: 4/28/00 3:10:48 PM
14C Chi Square: 28.30 Date Processed: 4/28/00 3:10:48 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 244.90 Date Processed: 4/28/00 3:10:48 PM
14C E²/B (0-156 keV and 1-156 keV): 452.29 Date Processed: 4/28/00 3:10:48 PM
3H Efficiency (0-18.6 keV): 65.56 Date Processed: 4/28/00 3:10:48 PM
14C Efficiency (0-156 keV): 96.48 Date Processed: 4/28/00 3:10:48 PM
IPA Background Date Processed: 4/28/00 3:10:48 PM
3H Background CPM (0-18.6 keV): 17.33 Date Processed: 4/28/00 3:10:48 PM
14C Background CPM (0-156 keV): 25.60 Date Processed: 4/28/00 3:10:48 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

114
4/24/2000
M. Nugent

4/24/2000 115
M. Nugent

4/24/00 12:35:09 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

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Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 14.60 Date Processed: 4/24/00 12:35:09 PM
14C Chi Square: 15.16 Date Processed: 4/24/00 12:35:09 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 263.01 Date Processed: 4/24/00 12:35:09 PM
14C E²/B (0-156 keV and 1-156 keV): 485.01 Date Processed: 4/24/00 12:35:09 PM
3H Efficiency (0-18.6 keV): 65.96 Date Processed: 4/24/00 12:35:09 PM
14C Efficiency (0-156 keV): 96.77 Date Processed: 4/24/00 12:35:09 PM
IPA Background Date Processed: 4/24/00 12:35:09 PM
3H Background CPM (0-18.6 keV): 16.35 Date Processed: 4/24/00 12:35:09 PM
14C Background CPM (0-156 keV): 24.38 Date Processed: 4/24/00 12:35:09 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

4/28/00 3:10:48 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/2

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 22.85 Date Processed: 4/28/00 3:10:48 PM
14C Chi Square: 28.30 Date Processed: 4/28/00 3:10:48 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 244.90 Date Processed: 4/28/00 3:10:48 PM
14C E²/B (0-156 keV and 1-156 keV): 452.29 Date Processed: 4/28/00 3:10:48 PM
3H Efficiency (0-18.6 keV): 65.56 Date Processed: 4/28/00 3:10:48 PM
14C Efficiency (0-156 keV): 96.48 Date Processed: 4/28/00 3:10:48 PM
IPA Background Date Processed: 4/28/00 3:10:48 PM
3H Background CPM (0-18.6 keV): 17.33 Date Processed: 4/28/00 3:10:48 PM
14C Background CPM (0-156 keV): 25.60 Date Processed: 4/28/00 3:10:48 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

116

4/26/2000

M Nugent

Summary: Experimental Conditions

Pre-Np spike:

Volume (L): $\frac{1L}{A}$
 Solution A/B:
 ml Ca added: 4 conc: 0.1 mL
 ml HCO₃ added: 4 conc: 0.1 mL
 Mass calcite added (g): 1.0046

Post-Np spike:

ml Np added: 0.25 conc: 118 ppm
 ml 0.32M NaOH added: 0.25
 Syringe Np conc (m/L): ~1.5 ppm
 Syringe Ca conc (m/L): 0.01M
 Pump Infusion Rate: 0.0133 ml/min
 Steady State pH: 8.35 to 8.38
 Temp, init. (Celcius): 21.6°C
 Temp, final (Celcius): 26.4°C

Exp Date: 4/26/2000

Post-experiment:

Mass calcite recovered 0.8337(g)

AA - Ca: units:

LSA - Np: units:

Sample Numbers: 66-71

Seed 58

Calculated:

Ptn Rate: not necessary to calculate

Np/Ca aq: 4/26/2000

Np/Ca solid:

Kd:

21.030 ml Ca added, 1.568 ml before Np.

M Nugent

117

4/26/2000

M Nugent

Neptunium Calcite Coprecipitation

Try $\log r = -4.5 \Rightarrow$ Repeat 4/20/2000 but with:
 - try 1.5 ppm in syringe solution

Objective: to coprecipitate Np and calcite @ $\log r = -4.5$
 moles/m²/hr with steady state Np_{aq} as described by the
 procedure on pages 3, 4 of this notebook, CNWRA Controlled Copy # 361,
 and the conditions above.

Np - Ca Syringe

Try 1.5 ppm in syringe

make 60 ml @ 1.5 ppm

 $18 \times V_1 = 60 \times 1.5 \Rightarrow$ add 0.7627 ml of 118 ppm Np soln.Instead - adjust extra syringe solution $\frac{118 \text{ ppm}}{4/26/2000}$ from 4/20/2000 exp

49.0112

~~49.9117~~

4/26/2000

to use for this exp. 29.7925

49.0253 - 19.2187 = 29.8066 g solution remaining

↑ total mass ↑ mass of bottle

This soln is $4.0625 \times 10^{-6} \text{ M/L} \Rightarrow 1.21089 \times 10^{-7} \text{ moles Np}$ Want ~30 ml @ 1.5 ppm = $\sim 1.898 \times 10^{-7} \text{ moles}$ \therefore add $6.87449 \times 10^{-8} \text{ moles}$.118 ppm Np soln is $4.9779 \times 10^{-4} \text{ M/L} \Rightarrow$ Add 0.138 ml

Actually added: Added 0.1378 grams of 118 Np soln to the
 solution from 4/20/2000.

Calcite seed added:

 $1.0055 - 0.0009 \text{ g} = 1.0046 \text{ g}$

Mass of calcite seed in
 weighboat before addition
 to reaction vessel

Mass of calcite seed left in weighboat
 after seed was added to reaction
 vessel.

4/26/2000
MN-gentCondition electrode. Calibrate electrode.
Filtered 1L of solution A

233 pm pH = 8.07. T = 21.6

236 4 ml of 0.1M Ca added pH = 8.08

237 4 ml of 0.1M H₂O₃ added pH = 8.60

240 8.52

305 8.39 T = 22.0°C

308 8.38 Added seeds → 8.37

Attached syringe @ 0.0133 ml/min
0.01M Ca, 0.01M HCO₃

309 8.36

311 pH = 8.35

319 8.36

326 pH = 8.36

331 8.36 T = 22.5°C

355 8.36

416 8.36

449 8.35 T = 24.9°C

506 8.34 T = 25.3°C

507 8.34 Added Np + NaOH → 8.47

508 8.35

509 8.35 Sample #1 42600-1

511 8.35

512 8.35 Stop syringe @ 1.5880 change to Np-Ca syringe

514 Start syringe pH = 8.35

549 8.36 T = 26.1

551 8.36 Sample #2 42600-2

4/27/2000

1004 T = 24.8°C pH = 8.39

1008 T = 24.8 pH 8.39 Sample #3 42600-3

1015 8.38

1056 pH = 8.37

4/26/2000

MN-gent

1206 pH = 8.38 T = 25.2

1207 8.38 Sample #4 42600-4

1249 pH = 8.38 T = 25.5°C

154 pH = 8.38 T = 26.1°C

325 T = 26.9 8.39 Sample #5 42600-5

420 8.39 T = 27.1°C

505 pH = 8.38/8.39 T = 26.7°C

534 8.39 26.4°C

537 8.39 STOP filter solution, same solids

• 21.030 ml added
from both Ca and H₂O₃ syringes.Mass of Solid
Recovered:
0.8337g

Prep of solids for LS+ analysis:

Date prepared: 4/28/2000 MN-gent

Mass, Bottle + Cap: 16.9335

tare

0.1011g calcite added to bottle + ~ 3ml acid + 0.5ml acid

Mass, bottle + cap + calcite + acid } 3.6438

Calcite + acid only!

SAMPLE NAME: S8

Date sent:

Solid sent for xray diffraction and SEM analyses 4/28/2000

Results will be recorded after analysis has
been performed. Approximately 0.1 grams used for SEM, 0.1g
used for XRD. See pages 145-150 of this notebook for SEM Results.

Sample Name	Mass (all in grams)		Sample	b+c+s+acid	a	TOTAL S+A	%sample	Subdivide the sample:		
	bottle+cap	b+c+s						Name	b+c	b+c+s
42600-1	16.8281	25.4109	8.5828	25.9201	0.5092	9.0920	94.3995	Npa66	16.9368	19.9637
42600-2	16.9041	24.553	7.6489	25.0628	0.5098	8.1587	93.7515	Npa67	16.9464	19.9718
42600-3	16.6809	25.2389	8.5580	25.7473	0.5084	9.0664	94.3925	Npa68	16.5308	19.5564
42600-5	16.9101	25.446	8.5359	25.9547	0.5087	9.0446	94.3756	Npa69	16.8569	19.8831
42600-6	16.939	24.6205	7.6815	25.1285	0.508	8.1895	93.7969	Npa70	16.7376	19.7604
42600-7	16.948	24.6945	7.7465	25.203	0.5085	8.2550	93.8401	Npa71	17.0513	20.0738

4/28/00 7:25:06 AM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 1
User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 500.00

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions Half Life

Units

Reference Date

Reference Time

Beta A

Beta B

Alpha

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS MESSAGES
1	500.00	33.15	1.55	39.74	1.42	2.17	6.07	853.84 Blank B
2	36.40	11.90	19.19	13.34	18.60	119.97	3.06	589.91 Np66a
3	39.73	5.96	34.39	7.98	28.35	109.76	3.06	942.16 Np67a
4	34.98	6.10	35.75	7.54	31.72	124.87	3.05	877.46 Np68a
5	32.04	6.77	33.86	9.35	27.14	136.59	3.05	1134.42 Np69a
6	32.21	5.50	40.92	8.10	30.88	135.89	3.05	1486.71 Np70a
7	31.79	8.21	28.48	9.65	26.49	137.62	3.05	757.83 Np71a

4/26/2000

Conclusion:

Np96 is steady!

page 1/4

Lst analysis for
aqueous Np for 4/26/2000
experiment
[Seed analysis, Page 3, 4]

4/27/00 7:30:13 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/4

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 9.68 Date Processed: 4/27/00 7:30:13 PM
14C Chi Square: 13.05 Date Processed: 4/27/00 7:30:13 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 255.57 Date Processed: 4/27/00 7:30:13 PM
14C E²/B (0-156 keV and 1-156 keV): 455.46 Date Processed: 4/27/00 7:30:13 PM
3H Efficiency (0-18.6 keV): 65.73 Date Processed: 4/27/00 7:30:13 PM
14C Efficiency (0-156 keV): 96.32 Date Processed: 4/27/00 7:30:13 PM
IPA Background Date Processed: 4/27/00 7:30:13 PM
3H Background CPM (0-18.6 keV): 16.77 Date Processed: 4/27/00 7:30:13 PM
14C Background CPM (0-156 keV): 25.28 Date Processed: 4/27/00 7:30:13 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

4/26/2000

Conclusion:

Np²³⁵ is steady!

4/28/00 11:40:09 PM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1:Serial# 405314

Page # 1
User: Nugent

Assay Definition-
Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00

Quench Set:
Count Time (min): 500.00
Count Mode: Normal
Assay Count Cycles: 1 Repeat Sample Count: 1
#Vials/Sample: 1 Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off
Regions Half Life Units Reference Date Reference Time
Beta A
Beta B
Alpha

Cycle 1 Results								SIS MESSAGES	
S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%		
1	500.00	37.28	1.46	45.46	1.33	2.18	6.05	890.56	Blank B
2	1.23	17447.27	1.37	17446.41	1.37	3623.02	3.00	210.13	55
3	0.67	6455.26	3.05	6456.04	3.05	6676.92	2.99	221.18	56
4	1.09	5253.54	2.65	5251.79	2.65	4112.49	2.99	230.25	57
5	1.27	5940.67	2.31	5937.22	2.31	3502.54	3.00	246.44	58

4/26/2000

Conclusion:

Np₂₆ is steady!

urgent
urgent
urgent

4/28/00 3:10:48 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 4/4

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 22.85 Date Processed: 4/28/00 3:10:48 PM
14C Chi Square: 28.30 Date Processed: 4/28/00 3:10:48 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 244.90 Date Processed: 4/28/00 3:10:48 PM
14C E²/B (0-156 keV and 1-156 keV): 452.29 Date Processed: 4/28/00 3:10:48 PM
3H Efficiency (0-18.6 keV): 65.56 Date Processed: 4/28/00 3:10:48 PM
14C Efficiency (0-156 keV): 96.48 Date Processed: 4/28/00 3:10:48 PM
IPA Background Date Processed: 4/28/00 3:10:48 PM
3H Background CPM (0-18.6 keV): 17.33 Date Processed: 4/28/00 3:10:48 PM
14C Background CPM (0-156 keV): 25.60 Date Processed: 4/28/00 3:10:48 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

urgent

4/26/2000

Conclusion:

Np²³⁵ is steady!

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5/21/2000

M. Nugent

Summary: Experimental Conditions

Pre-Np spike:

Volume (L): 1L
 Solution A/B: A
 ml Ca added: 5 conc: 0.1 mL
 ml HCO₃ added: 3 conc: 0.1 mL/L
 Mass calcite added (g): 1.0005g

Post-Np spike:

ml Np added: 0.25 conc: 118 ppm
 ml 0.32M NaOH added: 0.25
 Syringe Np conc (m/l): 2.02 ppm
 Syringe Ca conc (m/l): 0.01 mL
 Pump Infusion Rate: 0.0133 ml/min
 Steady State pH: 8.33
 Temp, init. (Celsius): 20.7°C
 Temp, final (Celsius): 23.2°C

Exp Date: 5/21/2000

Post-experiment:

Mass calcite recovered: 0.865g
 AA - Ca: _____ units:
 LSA - Np: _____ units:
 Sample Numbers: 72-75
 Seed - NO seed analysis

Calculated:

Pttm Rate: _____
 Np/Ca aq: _____
 Np/Ca solid: _____
 Kd: _____

22.821 mL added from Ca syringe,
 4.6307 mL before Np

Seed couldn't be filtered - filtration broke.

123

MN 5/21/2000

5-2-2000

M. Nugent

Neptunium Calcite Coprecipitation

Try Wgr = -3.5

NpCa syringe @ 2 ppm

0.133 mL/min

0.01M Ca, H₂O₃

Objective: to coprecipitate Np and calcite @ $\log r = -3.5$ mol/m²/sec hr
 with steady state Np aq. Perform experiment according to
 the procedure outlined on pages 2-3 of this notebook, CNWBT
 controlled copy #361.

Np-Ca syringe solution:

Make 30 mL @ 2 ppm

MN 5/21/2000

$$118 \times V = 30 \text{ mL} \times 2 \text{ ppm} \Rightarrow V = \frac{1.96 \text{ mL}}{118} = 0.0166 \text{ mL}$$

B+Cap: 11.4006g

0.5143g of 118 ppm stock solution

0.01M Ca sln added to 30.0335 grams.

$$\text{Conc: } 0.5143 \text{ g} \times 118 \text{ ppm} = C_2 \times 30.0335 \text{ g} \quad C_2 = 2.0206 \text{ ppm}$$

Calcite Seeds

$$1.0023 - 0.0018 = 1.0005 \text{ g added}$$

Mass calcite seeds
 before addition to
 the reaction vessel.

Mass calcite seeds remaining in the
 weigh boat after calcite seed addition
 to reaction vessel.

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5/2/2000

MNugent

Filter 1L SLHA, bubbling, stirring solution. Calibrate pH electrode.

233 pm pH = 7.84 T = 20.7°C

242 8.09

243 5ml of 0.1M Ca added → 8.08

244 5ml of 0.1M H₂O₃ added → 8.50 T = 20.8°C

253 8.42

255 Added seeds 8.40

256 Started syringes 0.01M Ca, H₂O₃ @ 0.133ml/min 8.38

257 8.37 T = 20.9°C

258 8.36

301 8.35

306 pH = 8.35 T = 21.0°C

326 8.26 T = 21.1°C

328 Add Np + NaOH → 8.62

329 8.38 Sample #1 5200-1

330 8.36

331 STOP syringe 8.36 (4.6307 ml added)

change to Np-Ca syringe, start

332 ~~8.35~~ MNugent 8.35 T = 21.6°C

358 8.33 T = 22.1°C

400 8.33 Sample #2 5200-2 T = 22.0°C

417 8.32 T = 22.4°C

435 8.32 22.8°C

439 8.33 Sample #3 5200-3

459 8.34 T = 23.2°C

542 8.33 23.8 Sample #4 5200-4

549 STOP 8.34

22.821 ml added from both the Ca syringe and the H₂O₃ syringe

Note:

Unfortunately, the filtration glassware broke during the exp. The calcite cannot be filtered. Allowed calcite to settle. Will still analyze the aqueous samples by LSA, and try to recover seeds using a less efficient filtration apparatus.

125

5/2/2000

MNugent

Calcite, Mass recovered: 0.8859g

Date recovered: 5/3/2000

Recovered by: M. Nugent

5/3/00 5:51:16 AM

QuantaSmart (TM) - 1.10

Protocol# 41 - Manual Np_Pa 3%2S.1:Serial# 405314

Page # 1

User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta

Report Name: Manual Np_Pa 3% 2sigma

Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa

Quench Indicator: SIS

External Std Terminator (sec): n/a

Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 500.00

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions Half Life

Units

Reference Date

Reference Time

Beta A

Beta B

Alpha

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	500.00	33.32	1.55	39.98	1.41	2.21	6.02	861.47	B
2	4.78	406.85	4.72	409.60	4.74	927.50	3.01	222.98	1
3	0.47	7828.39	3.30	7832.36	3.30	9723.33	2.96	173.09	2
4	35.53	44.98	6.70	46.59	6.81	122.90	3.06	372.90	Np72a
5	31.54	11.10	21.88	12.49	21.15	138.73	3.05	532.56	Np73a
6	25.81	15.23	18.33	16.47	18.29	170.09	3.04	411.46	Np74a
7	19.64	17.04	19.04	17.96	19.39	224.27	3.03	378.53	Np75a

LST analysis for
aqueous samples
for 5/2/2000

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5/2/2000

M Nugent

Filter 1L SLHA, bubbling, stirring solution. Calibrate pH electrode.

233 pm pH = 7.84 T = 20.7°C

242 8.09

243 5 ml of 0.1M Ca added → 8.08

244 5 ml of 0.1M H₂O₃ added → 8.50 T = 20.8°C

253 8.42

255 Added seeds 8.40

256 Started syringes 0.01M Ca, H₂O₃ @ 0.133 ml/min 8.38

257 8.37 T = 20.9°C

258 8.36

301 8.35

306 pH = 8.35 T = 21.0°C

326 8.26 T = 21.1°C

328 Add NP + NaOH → 8.62

329 8.38 Sample #1 5200-1

330 8.36

331 STOP syringe 8.36 (< 4.6307 ml added)

change to NP-Ca syringe, start

332 ~~8.35~~ 8.35 T = 21.6°C

358 8.33 T = 22.1°C

400 8.33 Sample #2 5200-2 T = 22.0°C

417 8.32 T = 22.4°C

435 8.32 22.8°C

439 8.33 Sample #3 5200-3

459 8.34 T = 23.2°C

542 8.33 23.8 Sample #4 5200-4

549 STOP 8.34

22.821 ml added from both the Ca syringe and the H₂O₃ syringe

Note:

Unfortunately, the filtration glassware broke during the exp. The calcite cannot be filtered. Allowed calcite to settle. Will still analyze the aqueous samples by LSA, and try to recover seeds using a less efficient filtration apparatus.

125

5/2/2000

M Nugent

Calcite, Mass recovered: 0.8859 g

Date recovered: 5/3/2000

Recovered by: M. Nugent

5/2/00 7:26:50 PM
SNC ProtocolQuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/2

Calibration Information

Software Version IC: 2.09

Software Version EC: 1.10

Instrument Model: Tri-Carb 3100TR

Instrument Serial Number: 405314

3H Chi Square: 11.29 Date Processed: 5/2/00 7:26:49 PM

14C Chi Square: 26.17 Date Processed: 5/2/00 7:26:49 PM

3H E²/B (0-18.6 keV and 1-18.6 keV): 254.81 Date Processed: 5/2/00 7:26:49 PM14C E²/B (0-156 keV and 1-156 keV): 463.38 Date Processed: 5/2/00 7:26:49 PM

3H Efficiency (0-18.6 keV): 66.01 Date Processed: 5/2/00 7:26:49 PM

14C Efficiency (0-156 keV): 96.43 Date Processed: 5/2/00 7:26:49 PM

IPA Background Date Processed: 5/2/00 7:26:49 PM

3H Background CPM (0-18.6 keV): 17.02 Date Processed: 5/2/00 7:26:49 PM

14C Background CPM (0-156 keV): 25.80 Date Processed: 5/2/00 7:26:49 PM

3H Calibration DPM: 285000

3H Reference Date: 10/29/99

14C Calibration DPM: 134100

===== Errors and Warnings =====

===== End of Errors and Warnings =====

M Nugent

M Nugent

5/2/2000

M Nugent

Sample Name	Mass (all in grams)		Sample	b+c+s+acid	a	TOTAL S+A	%sample	Subdivide the sample:			
	bottle+cap	b+c+s						Name	b+c	b+c+s	Sample
5200-1	16.7172	24.6745	7.9573	25.1836	0.5091	8.4664	93.9868	Npa72	16.819	19.8267	3.0077
5200-2	16.9298	25.5513	8.6215	26.0558	0.5045	9.1260	94.4718	Npa73	16.6412	19.6541	3.0129
5200-3	16.9157	25.168	8.2523	25.6788	0.5108	8.7631	94.1710	Npa74	16.6643	19.6794	3.0151
5200-4	16.7463	24.8215	8.0752	25.3332	0.5117	8.5869	94.0409	Npa75	16.917	19.9296	3.0126

Summary:

Np_{ac} increases during the experiment - therefore, try again with lower Np_{ac} in syringe.

5/2/2000

User: Nugent

5/8/00 12:42:58 PM
Protocol# 41 - Manual Np_Pa 3%2S.1
Serial# 405314

QuantaSmart (TM) - 1.10

LST analysis for
Seeds (dissolved)
for 5/2/2000

page 1/2

Assay Definition- Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00
Quench Set:
Count Time (min): 600.00
Count Mode: Normal
Assay Count Cycles: 1
#Vials/Sample: 1
Repeat Sample Count: 1
Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On
Colored Samples: n/a
Coincidence Time (nsec): 18
Luminescence Correction: Off
Heterogeneity Monitor: n/a
Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off
Regions Half Life
Beta A
Beta B
Alpha

Units Reference Date Reference Time

Cycle	1 Results	CPMA	A:2S%
S#	Count Time		
1	600.00	32.62	1.43
2	34.54	60.08	5.51
3	33.63	15.10	16.07
4	25.12	12.64	21.56
5	22.85	15.12	19.36
6	22.59	15.01	19.60
7	329.88	3.87	21.01
8	1.84	7804.88	1.67
9	4.02	4354.69	1.52
10	2.01	8335.54	1.55

CPMB	B:2S%	CPMA	alpha2S%
39.15	1.30	2.13	5.59
61.75	5.60	126.53	3.05
16.96	15.52	130.04	3.05
14.28	20.74	174.82	3.04
16.74	18.93	192.40	3.03
17.74	18.12	194.68	3.03
5.55	16.16	0.00	0.00
7808.68	1.67	2419.06	3.00
4358.12	1.52	1105.08	3.00
8339.96	1.55	2263.54	2.97

SIS	MESSAGES
851.12	Blank B
333.78	Np81a
610.14	Np82a
671.77	Np83a
610.40	Np84a
815.40	Np85a
1288.81	Seed Blank
243.61	Sa (5200)
235.82	S10 (5600)
246.03	S11 (5700)

5/8/00

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5/2/2000 M Nugent

Sample Name	Mass (all in grams)		Sample	b+c+s+acid	a	TOTAL S+A	%sample	Subdivide the sample:			
	bottle+cap	b+c+s						Name	b+c	b+c+s	Sample
5200-1	16.7172	24.6745	7.9573	25.1836	0.5091	8.4664	93.9868	Npa72	16.819	19.8267	3.0077
5200-2	16.9298	25.5513	8.6215	26.0558	0.5045	9.1260	94.4718	Npa73	16.6412	19.6541	3.0129
5200-3	16.9157	25.168	8.2523	25.6788	0.5108	8.7631	94.1710	Npa74	16.6643	19.6794	3.0151
5200-4	16.7463	24.8215	8.0752	25.3332	0.5117	8.5869	94.0409	Npa75	16.917	19.9296	3.0126

Summary:

Npac increases during the experiment - therefore, try again with lower Npac in syringe.

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5/2/2000 M Nugent

5/7/00 6:35:15 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/2

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 31.04 Date Processed: 5/7/00 6:35:14 PM
14C Chi Square: 31.06 Date Processed: 5/7/00 6:35:14 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 253.02 Date Processed: 5/7/00 6:35:14 PM
14C E²/B (0-156 keV and 1-156 keV): 469.05 Date Processed: 5/7/00 6:35:14 PM
3H Efficiency (0-18.6 keV): 65.87 Date Processed: 5/7/00 6:35:14 PM
14C Efficiency (0-156 keV): 96.65 Date Processed: 5/7/00 6:35:14 PM
IPA Background Date Processed: 5/7/00 6:35:14 PM
3H Background CPM (0-18.6 keV): 16.97 Date Processed: 5/7/00 6:35:14 PM
14C Background CPM (0-156 keV): 25.07 Date Processed: 5/7/00 6:35:14 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

5/6/2000 M Nugent

Summary: Experimental Conditions

Pre-Np spike:

Volume (L): 1L
 Solution A/B: B
 ml Ca added: 5 conc: 0.1m/L
 ml HCO₃ added: 5 conc: 0.1m/L
 Mass calcite added (g): 0.996 0.9996 grams
 MNS 5/6/2000

Post-Np spike:

ml Np added: 0.25 conc: 118ppm
 ml 0.32m NaOH added: 0.25
 Syringe Np conc (m/l): 17ppm
 Syringe Ca conc (m/l): 0.01m
 Pump Infusion Rate: 0.133 ml/min
 Steady State pH: 8.34
 Temp, init. (Celcius): 21.3°C
 Temp, final (Celcius): 22.9

Exp Date: 5/6/2000

Post-experiment:

Mass calcite recovered 0.9146 (g)
 AA - Ca: _____ units: _____
 LSA - Np: _____ units: _____
 Sample Numbers: 76-80
 Seed SL0
 Calculated:
 Pttm Rate: _____
 Np/Ca aq: _____
 Np/Ca solid: _____
 Kd: _____

13.605me added / 1.9354 w/o Np.

not
 necessary
 to calcul-
 ate
 5/7/2000

Neptunium-Calcite Coprecipitation 5/6/2000
 Try logr = -3.5
 Syringe Np = 1.7 ppm
 M Nugent

Objective: to coprecipitate up and calcite @ logr = -3.5
 mol/m²/hr with steady state Np_{aq}. Because the experiment
 with Np = 2 ppm in the Np-Ca syringe failed to produce steady state Np
 (see pages 123-127 of this notebook, CNWRA controlled copy #361),
 the experiment will repeated with less Np (Np = 1.7 ppm) in the
 Np-Ca syringe solution.

Np-Ca syringe solution:

Using the Np-Ca syringe solution from sl2 low experiment, dilute
 it to ~ 1.7 ppm. This solution is recorded on page 123 of this notebook, CNWRA
 Mass of sltn + Bottle + Cap = 23.9887g
 Bottle + Cap = 11.4006g
 Therefore 12.5881g of 2.02066 ppm sltn.
 calculate volume to add for 1.7 ppm:
 $12.5881 \times 2.02066 = 1.7 \text{ ppm} \times V_2 \Rightarrow V_2 = 14.8095$
 so, $14.8095 + 11.4006 = 26.2101 = \text{total mass of sltn + Bottle + Cap.}$

I added 0.01m Ca syringe solution to 26.2966g

calculate concentration:
 $12.5881 \times 2.02066 = (26.2966 - 11.4006) \times C_2$
 $C_2 = 1.70759 \text{ ppm.}$

The Np concentration in the Np-Ca
 syringe solution is approximately 1.708 ppm.

Calcite Seed:

1.0022g - 0.0026g = 0.9996 grams
 actual mass added to reaction vessel.

mass of seed
 before addition
 to the reaction
 vessel

mass of seed left in weighboat
 after seed addition to reaction
 vessel

5/16/2000
M. NugentCondition electrode Filtered 1L of solution B
Calibrate pH electrode.

4:31 pm 8.26 T=21.3

4:32 5x MN 1ml of 0.1M Ca → 8.23

8/14/2000 4:33 5x MN 1ml of 0.1M HCO₃ → 8.51

4:52 8.44 T=21.4°C

4:54 Added Calcite pH → 8.42

Attached syringes (0.01M Ca, 0.01M HCO₃) @ 0.133 ml/min

4:55 pm: pH = 8.40

4:57 pH = 8.37

4:58 8.36

5:00 8.34 T=21.5°C

5:02 pH = 8.33

5:04 8.32

5:06 8.32 T=21.6°C

5:07 8.31/8.32

5:08 Added Np + NaOH → 8.50

0.25 ml of 118 ppm spike + 0.25 ml 0.32 M NaOH

5:08 1/2 Sample #1 pH = 8.33 5600-1

5:09 Stopped pump @ 1.9354 ml added.

changed to np-ca syringe & restarted 1.9354

pump

5:10 8.32

5:12 8.32

5:24 8.34 T=21.9°C

5:24 MN 5:32 pH = 8.34 Sample #2 5600-2

5:51 8.36 T=22.3°C (recalibrated electrode)

5:57 8.36 Sample #3 5600-3

6:17 pH = 8.36 T=22.6°C

6:23 8.36 Sample #4 5600-4

6:32 8.36 T=22.9°C

6:36 8.36 Sample #5 5600-5

6:37 STOP!

Calcite Seed:

Mass recovered: 0.9142g

Recorded M. Nugent 5/17/2000

13.605 ml added from both the
Ca and HCO₃⁻
Syringes.M. Nugent 5/16/2000
cm⁴

Sample Name	Mass (all in grams)		Sample	b+c+s+acid	a	TOTAL STA	%sample	Subdivide the sample:			
	bottle+cap	b+c+s						Name	b+c	b+c+s	Sample
5600-1	16.7142	25.4255	8.7113	25.9332	0.5077	9.2190	94.4929	Npa76	16.6646	19.68	3.0154
5600-2	16.9486	24.477	7.3284	24.9834	0.5064	8.0348	93.6974	Npa77	16.7025	19.7197	3.0172
5600-3	16.7172	23.6251	6.9079	24.1235	0.4984	7.4063	93.2706	Npa78	16.8869	19.905	3.0181
5600-4	16.9344	24.7222	7.7878	25.228	0.5058	8.2936	93.9013	Npa79	16.7565	19.7737	3.0172
5600-5	16.9189	24.6889	7.7700	25.1942	0.5053	8.2753	93.8939	Npa80	16.814	19.8327	3.0187

Seed prep for LST

Seed Sample Name: S10

prepared 5/16/2000

by M. Nugent.

Mass of Bottle + Cap: 16.8811 grams

Mass of seed: 0.1045g

Mass, Seed + 1m HCl: 3.6634 grams

↑ mass measured after dissolution of seed was

Complete.

132 M Nugent

5/6/2000

5/7/00 7:22:09 AM

QuantaSmart (TM) - 1.10

Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 1

User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta

Report Name: Manual Np_Pa 3% 2sigma

Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa

Quench Indicator: SIS

External Std Terminator (sec): n/a

Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 500.00

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions Half Life

Beta A

Beta B

Alpha

Units

Reference Date

Reference Time

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS MESSAGES
1	500.00	33.01	1.56	39.53	1.42	2.15	6.10	850.83 Blank B
2	35.44	55.87	5.74	58.16	5.79	123.33	3.05	371.57 Np 76a
3	37.82	11.30	19.69	14.26	17.19	115.41	3.06	1016.09 Np 76a
4	28.54	84.79	4.83	86.96	4.88	153.56	3.04	332.48 Np 76a
5	30.56	12.70	19.68	14.92	18.28	143.30	3.05	778.48 Np 76a
6	28.05	10.59	24.05	12.88	21.67	156.28	3.04	670.84 Np 76a

LST analysis of
aqueous samples for
experiment 5/6/2000.

[Seed analysis page 3 and page 4]

page 1/4

133

5/6/2000

M. Nugent

Conclusion: Np ag ↑ with time. Syringe Np is too
high. Try again, with ~~10~~ 5/12/2000 with lower Np
concentration in syringe.

132 MNugent
5/6/00

5/6/00 8:14:29 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

Page 2/4

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 15.23 Date Processed: 5/6/00 8:14:29 PM
14C Chi Square: 21.28 Date Processed: 5/6/00 8:14:29 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 272.52 Date Processed: 5/6/00 8:14:29 PM
14C E²/B (0-156 keV and 1-156 keV): 499.79 Date Processed: 5/6/00 8:14:29 PM
3H Efficiency (0-18.6 keV): 66.07 Date Processed: 5/6/00 8:14:29 PM
14C Efficiency (0-156 keV): 96.36 Date Processed: 5/6/00 8:14:29 PM
IPA Background Date Processed: 5/6/00 8:14:29 PM
3H Background CPM (0-18.6 keV): 15.77 Date Processed: 5/6/00 8:14:29 PM
14C Background CPM (0-156 keV): 23.32 Date Processed: 5/6/00 8:14:29 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

with
Mugent
out

133

5/6/2000
M. Nugent

Conclusion: Np²³⁵ ↑ with time : Syringe Np is too
high. Try again, with ~~100~~ 50% with lower Np
concentration in syringe.

132 MNugent
5/6/2000

5/8/00 12:42:58 PM

QuantaSmart (TM) - 1.10

Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 1

User: Nugent

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta

Report Name: Manual Np_Pa 3% 2sigma

Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa

Quench Indicator: SIS

External Std Terminator (sec): n/a

Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 600.00

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CFM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions Half Life

Units

Reference Date

Reference Time

Beta A

Beta B

Alpha

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	600.00	32.62	1.43	39.15	1.30	2.13	5.59	851.12	Blank B
2	34.54	60.08	5.51	61.75	5.60	126.53	3.05	333.78	Np85a Np81a
3	33.63	15.10	16.07	16.96	15.52	130.04	3.05	610.14	Np85a Np82a
4	25.12	12.64	21.56	14.28	20.74	174.82	3.04	671.77	Np85a Np83a
5	22.85	15.12	19.36	16.74	18.93	192.40	3.03	610.40	Np85a Np84a
6	22.59	15.01	19.60	17.74	18.12	194.68	3.03	815.40	Np85a
7	329.88	3.87	21.01	5.55	16.16	0.00	0.00	1288.81	Seed Blank
8	1.84	7804.88	1.67	7808.68	1.67	2419.06	3.00	243.61	89 (3200)
9	4.02	4354.69	1.52	4358.12	1.52	1105.08	3.00	235.82	810 (5600)
10	2.01	8335.54	1.55	8339.96	1.55	2263.54	2.97	246.03	S11 (5700)

LST analysis of
Seeds (dissolved) for
experiment 5/6/2000.

Page 3/4

133

5/6/2000

M. Nugent

Conclusion: Np85 ↑ with time : Syringe Np is too
high. Try again, with 1/2 syringe with lower Np
concentration in syringe.

132 M Nugent

5/6/2000

5/7/00 6:35:15 PM
SNC Protocol

QuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

Page 4/4

Calibration Information
Software Version IC: 2.09
Software Version EC: 1.10
Instrument Model: Tri-Carb 3100TR
Instrument Serial Number: 405314
3H Chi Square: 31.04 Date Processed: 5/7/00 6:35:14 PM
14C Chi Square: 31.06 Date Processed: 5/7/00 6:35:14 PM
3H E²/B (0-18.6 keV and 1-18.6 keV): 253.02 Date Processed: 5/7/00 6:35:14 PM
14C E²/B (0-156 keV and 1-156 keV): 469.05 Date Processed: 5/7/00 6:35:14 PM
3H Efficiency (0-18.6 keV): 65.87 Date Processed: 5/7/00 6:35:14 PM
14C Efficiency (0-156 keV): 96.65 Date Processed: 5/7/00 6:35:14 PM
IPA Background Date Processed: 5/7/00 6:35:14 PM
3H Background CPM (0-18.6 keV): 16.97 Date Processed: 5/7/00 6:35:14 PM
14C Background CPM (0-156 keV): 25.07 Date Processed: 5/7/00 6:35:14 PM
3H Calibration DPM: 285000
3H Reference Date: 10/29/99
14C Calibration DPM: 134100
===== Errors and Warnings =====
===== End of Errors and Warnings =====

print
out
print
merged

133

5/6/2000

M. Nugent

Conclusion: Np²³⁵ ↑ with time. Syringe Np is too
high. Try again, with ~~10~~ 5/12/2000 with lower Np
concentration in syringe.

5/7/2000 M. Nugent

Neptunium Calcite Coprecipitation

5-7-2000

M. Nugent

Try $\log r = -3.5$

Try 1.6 ppm in syringe

Objective: to coprecipitate calcite and Np at steady state.
 Npase. Because the experiment performed with 1.7 ppm in the Np-Ca syringe solution had too much Np in solution (see experiment dated 5/6/2000 on pages 129-133 of this notebook, CNWRA controlled copy #361), try to repeat that experiment, but with less (Np=1.6 ppm) Np in the syringe solution. Therefore, use the same methods and equipment as experiment 5/6/2000, pages 129-133 of this notebook.

Np-Ca syringe solution:

30 ml \times 1.6 ppm = 118 \times $V_2 \Rightarrow V_2 = 0.4068$ g of sltn added
 to 30 ml of 0.01 M Ca syringe sltn.

Bottle + cap: 11.5128

Grams of 118 spike Np: 0.4041

Grams of Np spike + 0.01 M Ca syringe sltn: 30.0371 g

Record Mass of Calcite Seed Added to reaction vessel.

Calcite Seed [Mass seed before addition - Mass seed in weight boat after addition]

1.0037 g - 0.0008 g = 1.0029 g

Summary: Experimental Conditions

Exp Date: 5/7/2000

Pre-Np spike:

Volume (L): 1L
 Solution A/B: B
 ml Ca added: 5 conc: 0.1 M/L
 ml HCO₃ added: 5 conc: 0.1 M/L
 Mass calcite added (g): 1.0029

Post-Np spike:

ml Np added: 0.25 conc: 118 ppm
 ml 0.32 M NaOH added: 0.25
 Syringe Np conc (m/l): 1.59 ppm
 Syringe Ca conc (m/l): 0.01
 Pump Infusion Rate: 0.133 ml/min
 Steady State pH: 8.36
 Temp, init. (Celsius): 21.2
 Temp, final (Celsius): 25.0

Post-experiment:

Mass calcite recovered: 0.9073 (g)

AA - Ca: units:

LSA - Np: units:

Sample Numbers: 81-85

Seed 311

Calculated:Ptn Rate: ~~not necessary to calculate~~Np/Ca aq: ~~5/8/2000~~Np/Ca solid: ~~5/8/2000~~Kd: ~~5/8/2000~~

26.823 ml Ca added, 3.4348 before Np

M. Nugent

136

5/7/2000

MNugent

Filtered 1L of solution B, started bubbling air
 & stirring. Calibrate pH electrode.

106 pm pH = 7.75 $T = 21.2^{\circ}\text{C}$

108 pm Added 5 ml of 0.1 M Ca $\rightarrow 7.61$

109 pm Added 5 ml 0.1 M $\text{HCO}_3^- \rightarrow \text{pH} = 8.14$

121 8.23

127 8.28

130 8.31

131 8.31/8.32 Added calcite seeds and

attached 0.01 M Ca and 0.01 HCO_3^- syringes
 @ 0.133 ml/min

132 pH = 8.29 $T = 21.5^{\circ}\text{C}$

151 8.35 $T = 21.7$

156 pH = 8.35 Added Np + NaOH pH $\rightarrow 8.42$

157 8.37 Sample #1 5700-1

157 1/2 STOP Syringe @ 3.4348 ml added
 change to Np-Ca syringe. 3.4348

158 1/2 8.35

201 8.35

220 8.36 $T = 22.3^{\circ}\text{C}$

229 8.36

231 8.36 $T = 22.4^{\circ}\text{C}$ Sample #2 5700-2
~~5600-2~~
 MN 5/7/2000

298 8.36

400 8.36 $T = 24.4^{\circ}\text{C}$

403 8.36 Sample #3 ~~5600-3~~ 5700-3
 MN 5/7/2000

431 8.36/8.37 $T = 24.9^{\circ}\text{C}$

437 8.36 Sample #4 ~~5600-4~~ 5700-4
 MN 5/7/2000

446 8.36

452 8.36 Sample #5 ~~5600-5~~ 5700-5
 MN 5/7/2000

455 8.36 $T = 25.0^{\circ}\text{C}$ STOP

26.823 ml added from Ca syringe

26.823 ml added from HCO_3^- syringe.

Calute Seed:

Mass Recovered: 0.9073g

137

Seed Prep for LST:

Bottle + cap: 16.9762

Sample Name S11

5/7/2000

Mass of seed added: 0.1129

MNugent

Mass of seed + 1 M HCl acid: 3.3967

Sample Name	Mass (all in grams)		Sample	b+c+s+acid	a	TOTAL S+A	%sample	Subdivide the sample:			
	bottle+cap	b+c+s						Name	b+c	b+c+s	Sample
5700-1	17.0415	24.6829	7.6414	25.1914	0.5085	8.1499	93.7607	Npa81	16.8984	19.9228	3.0244
5700-2	16.9569	25.0732	8.1163	25.5821	0.5089	8.6252	94.0998	Npa82	16.922	19.9468	3.0248
5700-3	16.9003	23.929	7.0287	24.4372	0.5082	7.5369	93.2572	Npa83	16.9362	19.9621	3.0259
5700-4	16.7324	24.3913	7.6589	24.8925	0.5012	8.1601	93.8579	Npa84	16.9371	19.9631	3.0260
5700-5	16.7731	24.3094	7.5363	24.8142	0.5048	8.0411	93.7223	Npa85	16.7589	19.7199	2.9610

mugent

5/7/2000

MNugent

5/8/00 12:42:58 PM QuantaSmart (TM) - 1.10
Protocol# 41 - Manual Np_Pa 3%2S.1Serial# 405314

Page # 1
User: Nugent

Assay Definition-
Assay Description:

Assay Type: Alpha/Beta
Report Name: Manual Np_Pa 3% 2sigma
Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S
Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa
Quench Indicator: SIS
External Std Terminator (sec): n/a
Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 600.00

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial
Low CPM Threshold: Off
2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On Luminescence Correction: Off
Colored Samples: n/a Heterogeneity Monitor: n/a
Coincidence Time (nsec): 18 Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS	MESSAGES
1	600.00	32.62	1.43	39.15	1.30	2.13	5.59	851.12	Blank B
2	34.54	60.08	5.51	61.75	5.60	126.53	3.05	333.78	Np82a Np81a
3	33.63	15.10	16.07	16.96	15.52	130.04	3.05	610.14	Np85a Np82a
4	25.12	12.64	21.56	14.28	20.74	174.82	3.04	671.77	Np87a Np83a
5	22.85	15.12	19.36	16.74	18.93	192.40	3.03	610.40	Np85a Np84a
6	22.59	15.01	19.60	17.74	18.12	194.68	3.03	815.40	Np85a
7	329.88	3.87	21.01	5.55	16.16	0.00	0.00	1288.81	Seed Blank
8	1.84	7804.88	1.67	7808.68	1.67	2419.06	3.00	243.61	S9 (5200)
9	4.02	4354.69	1.52	4358.12	1.52	1105.08	3.00	235.82	S10 (5600)
10	2.01	8335.54	1.55	8339.96	1.55	2263.54	2.97	246.03	S11 (5700)

MN 5/8/2000

5/7/2000

Np ac ↑ in solution during experiment. Syringe MNugent
Np concentration is too high. Try again, with lower
Np concentration in the Np-2a syringe.

138

5/7/2000

MNugent

5/7/00 6:35:15 PM

QuantaSmart (TM) - 1.10

Page # 1

SNC Protocol

Serial# 405314

page 2/2

Calibration Information

Software Version IC: 2.09

Software Version EC: 1.10

Instrument Model: Tri-Carb 3100TR

Instrument Serial Number: 405314

3H Chi Square: 31.04 Date Processed: 5/7/00 6:35:14 PM

14C Chi Square: 31.06 Date Processed: 5/7/00 6:35:14 PM

3H E²/B (0-18.6 keV and 1-18.6 keV): 253.02 Date Processed: 5/7/00 6:35:14 PM

14C E²/B (0-156 keV and 1-156 keV): 469.05 Date Processed: 5/7/00 6:35:14 PM

3H Efficiency (0-18.6 keV): 65.87 Date Processed: 5/7/00 6:35:14 PM

14C Efficiency (0-156 keV): 96.65 Date Processed: 5/7/00 6:35:14 PM

IPA Background Date Processed: 5/7/00 6:35:14 PM

3H Background CPM (0-18.6 keV): 16.97 Date Processed: 5/7/00 6:35:14 PM

14C Background CPM (0-156 keV): 25.07 Date Processed: 5/7/00 6:35:14 PM

3H Calibration DPM: 285000

3H Reference Date: 10/29/99

14C Calibration DPM: 134100

===== Errors and Warnings =====

===== End of Errors and Warnings =====

139

5/7/2000

MNugent

Np ac ↑ in solution during experiment. Syringe
Np concentration is too high. Try again, with lower
Np concentration in the Np-239 syringe.

140

5/11/2000

MNugent

Summary: Experimental Conditions

Exp Date: 5/11/2000

Pre-Np spike:

Volume (L): 1L
 Solution A/B: B
 ml Ca added: 5 conc: 0.1 ml/L
 ml HCO₃ added: 5 conc: 0.1 ml/L
 Mass calcite added (g): 1.0017g

Post-Np spike:

ml Np added: 0.25 conc: 118 ppm
 ml 0.32M NaOH added: 0.25
 Syringe Np conc (m/l): 1.397 ppm
 Syringe Ca conc (m/l): 0.01
 Pump Infusion Rate: 0.133 ml/min
 Steady State pH: 8.30
 Temp, init. (Celcius): 22.5°C
 Temp, final (Celcius): 25.4°C

Post-experiment:

Mass calcite recovered: _____ (g)
 AA - Ca: _____ units: _____
 LSA - Np: _____ units: _____
 Sample Numbers: _____

Seed

Calculated:

Pttm Rate: not necessary
 Np/Ca aq: to calculate
 Np/Ca solid: not 5/11/2000
 Kd: _____

Ca: 2.9069 me added, 8.4304 me before Np.

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Neptunium - Calcite Coprecipitation 5/11/2000

Try log_r = -3.5

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try 1.4 ppm in syringe.

Objective: trying to get coprecipitation @ log_r = -3.5 ml/m³.hr with steady state Np aq.

Therefore, repeat the experiment performed on 5/7/2000 and recorded on pages 135-139 of this notebook (CNWRA Controlled copy # 361), except decrease the amount of neptunium in the Ca-Np syringe.

Therefore, use the same materials and methods as described by ^{MN 5/11/2000} experiment the 5/7/2000 experiment.

Np-Ca Syringe Solution:

Try 0.3559 ml of 118 ppm set into 30 ml 0.01 M Ca.

Bottle #22 MN 5/11/2000 11.2284g (no cap)

Mass of 118 ppm Np solution: 0.3614

Mass of 118 ppm Np sltn + 0.01 M Ca sltn: 30.5372

~ 1.397 ppm

Weigh out approximately 1 gram of calcite seed.

Calcite seed

1.0027 - 0.0010g = 1.0017 grams added.

↑
Mass of seed in weighboat

↑
Mass of seed remaining in weighboat after seed is added to reaction vessel.

5/11/2000 MNugent

Filtered 1L Solution B. Bubbling and stirring. Calibrate Electrode.

1040 am pH = 7.79

1102 am pH = 8.16 T = 22.5°C

1103 5ml of 0.1M Ca added → pH = 8.13

1104 5ml of 0.1M H₂O₃ added → pH = 8.48

1117 8.37, 22.8°C

1121 pH = 8.36 Added calcite seeds, Attached

syringes - 0.01M Ca and 0.01M HCO₃ @ 0.133 ml/min

1122 pH = 8.32

1124 8.31/8.30

1129 8.28 T = 23.0°C

1133 8.27 T = 23.2°C

1137 8.26

1141 8.26

1203 pH = 8.26 T = 23.9

1220 pH = 8.29 T = 24.2

MN still 2000 → 8.25

0.25 ml of

1222 8.25 Added Np + NaOH → 8.48

0.25 ml 118 ppm soln + 0.32 M NaOH

1223 8.24 Sample #1 51100-1

1224 MN 1204 pm: 8.23 24.2°C

Stopped syringes @ 8.5 MN 8.4304 ml 8.4304
and changed to Np-Ca syringe.

1226 8.24

1230 pH = 8.26 T = 24.4°C

1236 8.27 24.4°C Sample #2 51100-2

112 8.29 T = 24.8°C

148 8.30 T = 25.2°C

151 8.30 Sample #3 51100-3

223 T = 25.3°C pH = 8.31

224 pm 8.31 Sample #4 51100-4

238 8.31 T = 25.3°C

247 8.31 T = 25.4°C

249 8.31 Sample #5 51100-5

301 8.31 24 MN still 2000 25.4°C STOP!

29.069 ml added from both the Ca syringe and the H₂O₃ syringe

5/11/2000

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Solution was filtered and the calcite seeds were stored for LSA analysis.

Sample Name	Mass (all in grams) bottle+cap b+c+s		Sample	b+c+s+acid	a	TOTAL SFA	%sample	Subdivide the sample:			
								Name	b+c	b+c+s	Sample
51100-1	17.0066	25.7361	8.7295	26.2453	0.5092	9.2387	94.4884	Npa86	16.6457	19.6715	3.0258
51100-2	16.7493	23.7098	6.9605	24.2181	0.5083	7.4688	93.1944	Npa87	16.8573	19.8835	3.0262
51100-3	16.9441	25.9544	9.0103	26.4631	0.5087	9.5190	94.6560	Npa88	16.9322	19.9624	3.0302
51100-4	16.8798	25.1074	8.2276	25.6147	0.5073	8.7349	94.1923	Npa89	16.8655	19.8945	3.0290
51100-5	16.937	25.7787	8.8417	26.2871	0.5084	9.3501	94.5626	Npa90	16.7665	19.7962	3.0297

5/11/2000

MNugent

5/12/00 5:48:34 AM

QuantaSmart (TM) - 1.10

Page # 1

Protocol 41 - Manual Np_Pa 3%2S.1Serial# 405314

User: Nugent

Page 1/2

Assay Definition-

Assay Description:

Assay Type: Alpha/Beta

Report Name: Manual Np_Pa 3% 2sigma

Output Data Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Raw Results Path: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S

Comma-Delimited File Name: C:\Packard\Tricarb\Results\Nugent\Manual Np_Pa 3%2S\Manual Np_Pa 3%2S.txt

Count Conditions-

Nuclide: Manual Np/Pa

Quench Indicator: SIS

External Std Terminator (sec): n/a

Pre-Count Delay (min): 0.00

Quench Set:

Count Time (min): 400.00

Count Mode: Normal

Assay Count Cycles: 1

#Vials/Sample: 1

Repeat Sample Count: 1

Calculate % Reference: Off

Background Subtract: On - 1st Vial

Low CPM Threshold: Off

2 Sigma % Terminator: On - Any Region

In Use Discriminator: 137

Regions	LL	UL	Bkg Subtract	2Sigma % Terminator
Beta A	0.0	400.0	1st Vial	0.00
Beta B	0.0	2000.0	1st Vial	0.00
Alpha	100.0	400.0	1st Vial	3.00

Count Corrections-

Static Controller: On

Colored Samples: n/a

Coincidence Time (nsec): 18

Luminescence Correction: Off

Heterogeneity Monitor: n/a

Delay Before Burst (nsec): 75

Half Life-

Half Life Correction: Off

Regions	Half Life	Units	Reference Date	Reference Time
Beta A				
Beta B				
Alpha				

Cycle 1 Results

S#	Count Time	CPMA	A:2S%	CPMB	B:2S%	CPMa	alpha2S%	SIS MESSAGES
1	400.00	33.44	1.73	40.07	1.58	2.13	6.85	851.81 Blank B
2	36.30	36.73	7.73	38.58	7.80	120.32	3.06	434.85 Np 86a
3	37.87	11.74	19.24	13.22	18.57	115.22	3.06	705.89 Np 87a
4	30.21	8.80	27.66	12.27	22.07	145.07	3.04	1167.91 Np 88a
5	26.71	10.10	25.91	11.75	24.32	164.25	3.04	604.38 Np 89a
6	24.94	13.11	21.30	15.27	19.95	176.10	3.04	659.10 Np 90a

LST analysis
of aqueous samples
for 5/11/2000 experiment

5/14/2000

M. Nugent

SEM Analysis of Calcite Seeds, before and after reaction.

Calcite seeds were analyzed in the Scanning Electron Microscope (SEM) in Division 1. Samples were carbon-coated before analysis.

Unreacted calcite seeds were analyzed, as well as calcite seeds which were precipitated, with ~~uranium~~ ^{neptunium} MN 5/14/2000, as overgrowths onto the seeds. The unreacted calcite seeds are called 'calcite' and the reacted calcites are called '42400' and '42600' for the experiments performed on 4/26/00 and 4/24/00 and recorded on pages 116-121 and 111-115 of this notebook, ENWRA Controlled Copy #361. The samples and their reaction conditions, are described below.

Note, analyses were performed by Fawn Daby.

Unreacted Calcite: Calcite seeds used in all reactions described in this notebook were analysed. A ~~representative~~ MN 5/14/2000 representative sampling is presented on the ~~see~~ MN 5/14/2000 next page. In Figure 1 and 2.
No chemical analyses were performed.

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5/11/2000

M. Nugent

5/11/00 8:26:06 PM
SNC ProtocolQuantaSmart (TM) - 1.10
Serial# 405314

Page # 1

page 2/2

Calibration Information
 Software Version IC: 2.09
 Software Version EC: 1.10
 Instrument Model: Tri-Carb 3100TR
 Instrument Serial Number: 405314
 3H Chi Square: 18.69 Date Processed: 5/11/00 8:26:05 PM
 14C Chi Square: 21.21 Date Processed: 5/11/00 8:26:05 PM
 3H E²/B (0-18.6 keV and 1-18.6 keV): 268.17 Date Processed: 5/11/00 8:26:05 PM
 14C E²/B (0-156 keV and 1-156 keV): 462.42 Date Processed: 5/11/00 8:26:05 PM
 3H Efficiency (0-18.6 keV): 65.86 Date Processed: 5/11/00 8:26:05 PM
 14C Efficiency (0-156 keV): 96.26 Date Processed: 5/11/00 8:26:05 PM
 IPA Background Date Processed: 5/11/00 8:26:05 PM
 3H Background CPM (0-18.6 keV): 15.98 Date Processed: 5/11/00 8:26:05 PM
 14C Background CPM (0-156 keV): 25.10 Date Processed: 5/11/00 8:26:05 PM
 3H Calibration DPM: 285000
 3H Reference Date: 10/29/99
 14C Calibration DPM: 134100
 ===== Errors and Warnings =====
 ===== End of Errors and Warnings =====

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5/14/2000

M. Nugent

SEM Analysis of Calcite Seeds, before and after reaction.

Calcite seeds were analyzed in the Scanning Electron Microscope (SEM) in Division 1. Samples were carbon-coated before analysis.

Unreacted calcite seeds were analyzed, as well as calcite seeds which were precipitated, with ~~uranium~~ ^{neptunium} MN 5/14/2000, as overgrowths onto the seeds. The unreacted calcite seeds are called 'calcite' and the reacted calcites are called '42400' and '42600' for the experiments performed on 4/26/00 and 4/24/00 and recorded on pages 116-121 and 111-115 of this notebook, ENWRA Controlled Copy #361. The samples and their reaction conditions, are described below.

Note, analyses were performed by Fawn Daby.

Unreacted Calcite: Calcite seeds used in all reactions described in this notebook were analyzed. A ~~representative~~ MN 5/14/2000 representative sampling is presented on the ~~see~~ MN 5/14/2000 next page in Figure 1 and 2.
 No chemical analyses were performed.

5/14/2000
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Figure 1
Unreacted
Calcite
Seeds.

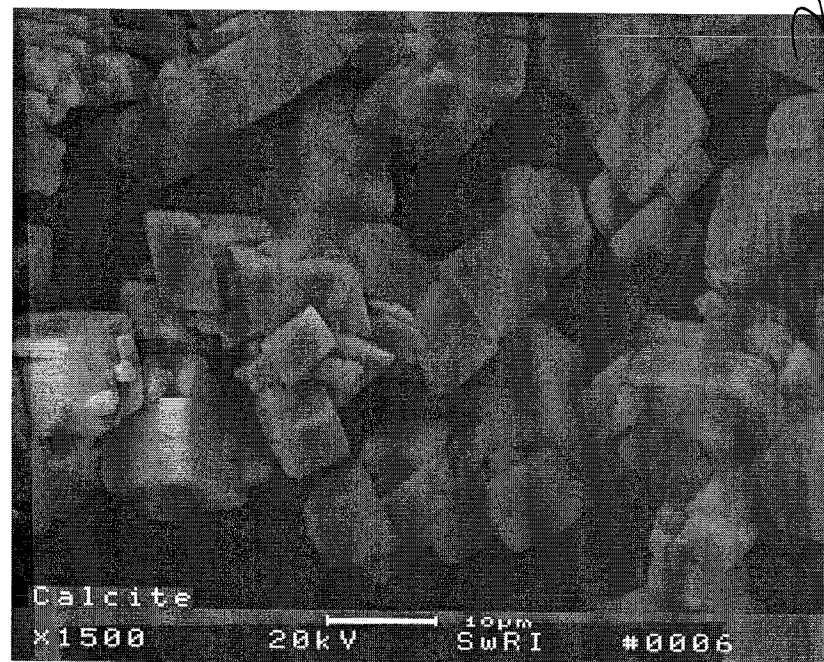
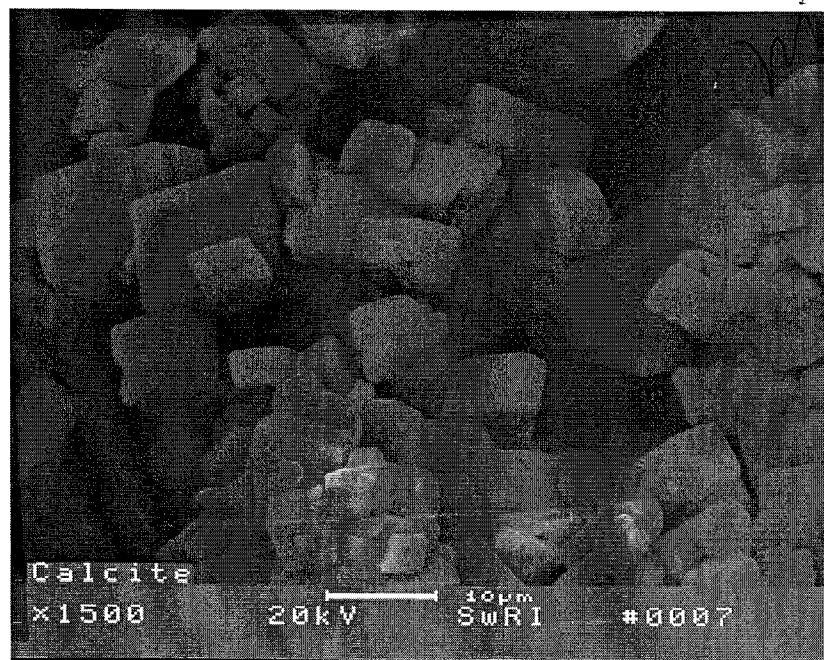


Figure 2
Unreacted
Calcite
Seeds.



5/14/2000 M. Nugent

Calcite Seeds with Np-bearing overgrowths

Sample name, 4/24/2000 = 42400.

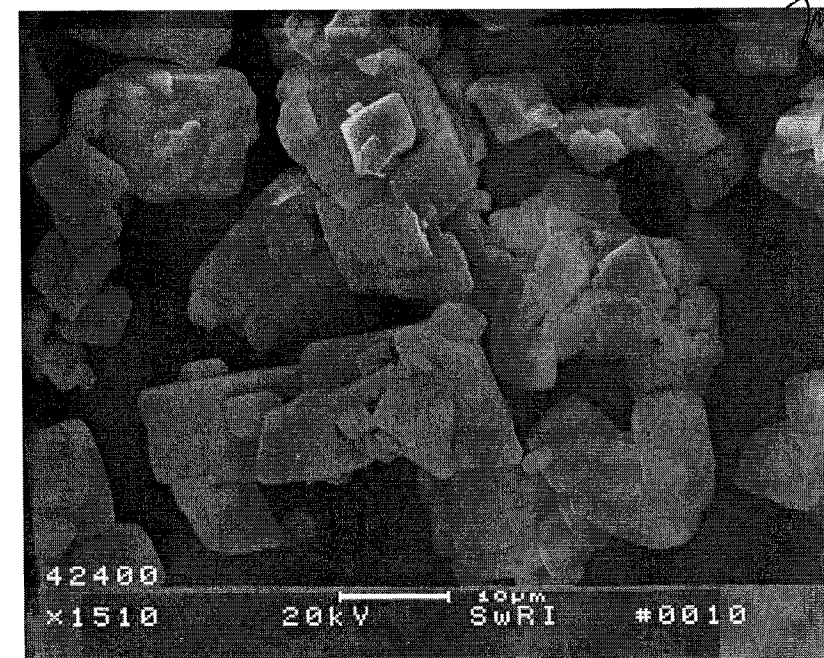
Experimental conditions:

- log rate of precipitation = -3.2 (fast)
- ~3 ppm Np in Np-Ca syringe, ~28 ppb Np in reaction vessel, initially. Np as was not steady in experiment but this experiment should be acceptable for analysis, since Np in solution was not completely depleted.

Representative samples of the reacted calcite seed are presented on pages 147 and 148 of this notebook, CNWRA controlled copy #361, as Figures 3 and 4.

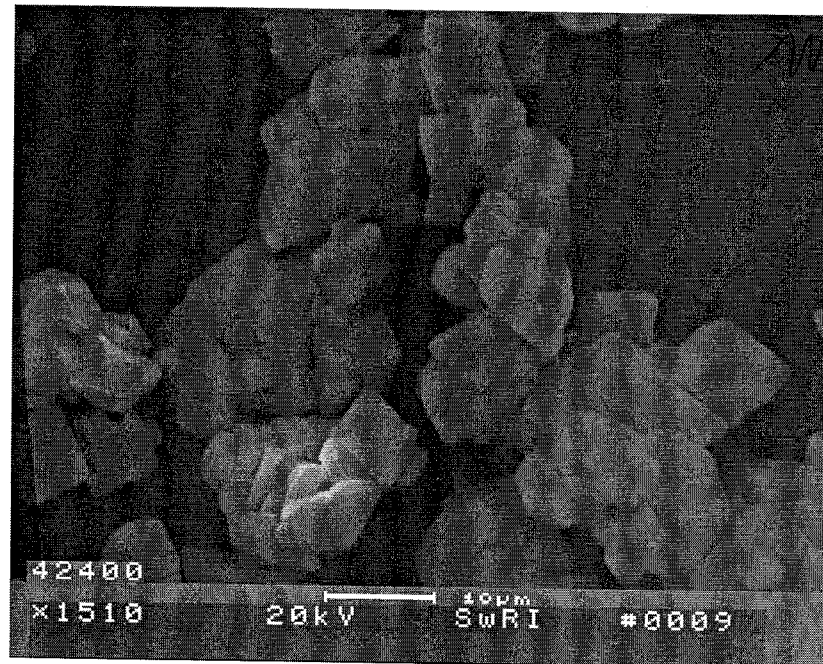
No chemical analyses were performed.

Figure 3. Calcite seeds with Np-bearing overgrowths, from Experiment 42400 performed on 4/24/2000.



5/14/2000
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Figure 4. Calcite seeds with Np-bearing overgrowths,
from Experiment 42400 performed on 4/24/2000.



Note, The calcite samples from Experiment 42400
do not appear to be different from the samples labeled
'Calcite'.

5/14/2000
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Sample Name: 42600 = 4/26/2000

Experimental Conditions:

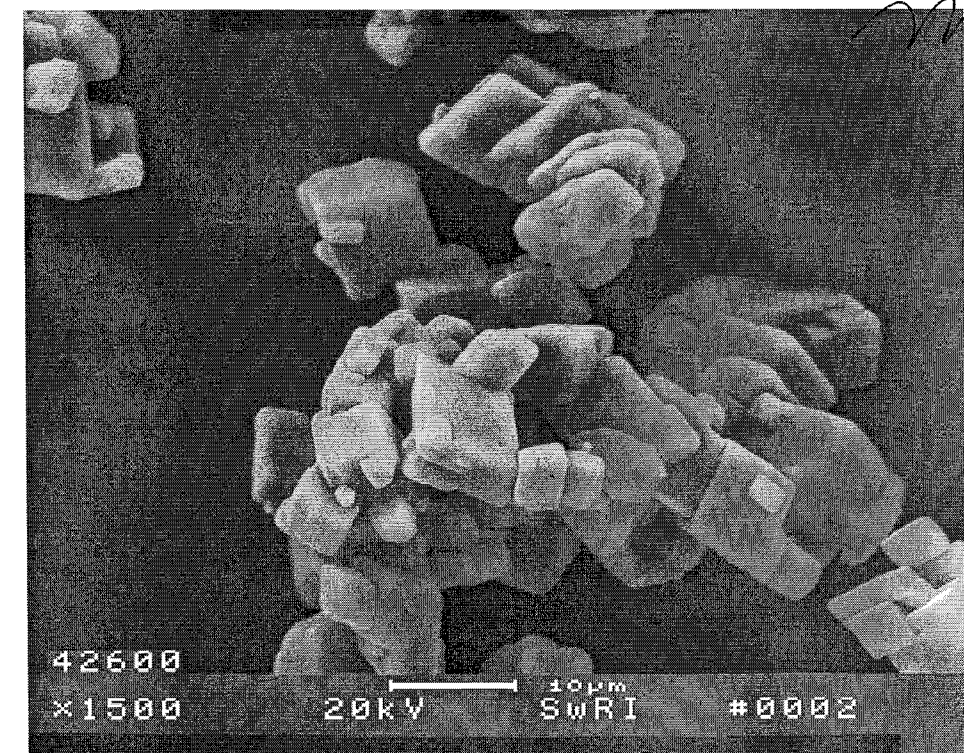
- log precipitation rate = -4.5 (~~slow~~) MN 5/14/2000 (slow)
- 1.5 ppm Np in Np-Ca syringe, and ~28 ppb

Np in reaction vessel initially.

Representative pictures are presented as Figures 4 and
Figures 5 and 6 on pages 149 and 150 of this notebook,
controlled copy #361, CNWRA.

Note, no chemical analyses were performed.

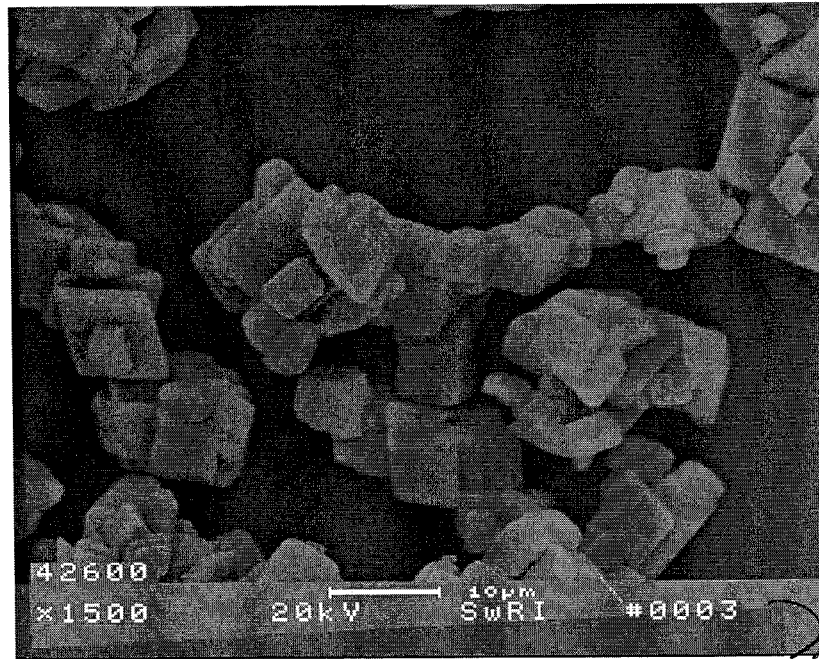
Figure 5. Calcite seeds with Np-bearing calcite overgrowths,
taken from Experiment 42600 performed on 4/26/2000.



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5/14/2000
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Figure 6. SEM image of calcite seeds with
Np-bearing calcite overgrowths, taken from
Experiment 42600, performed on 4/26/00.

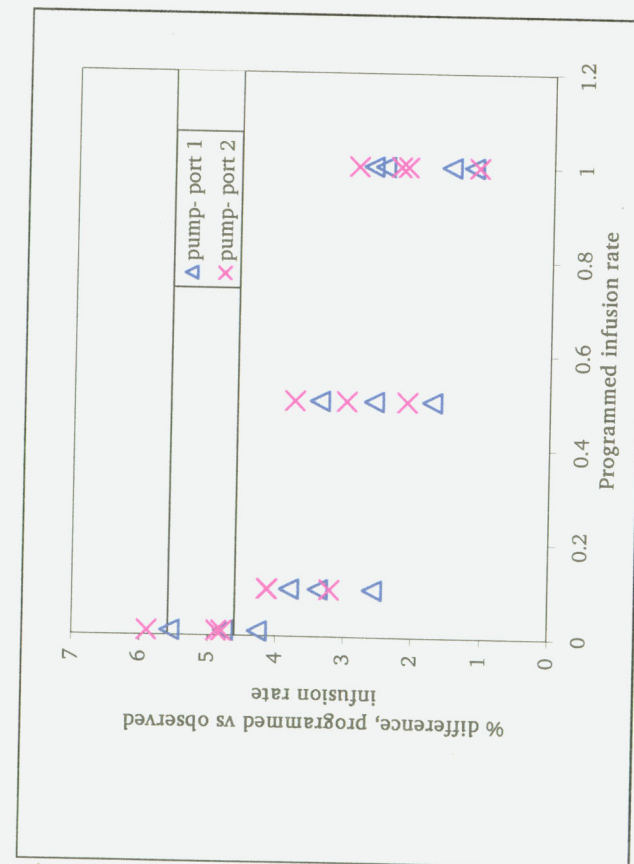
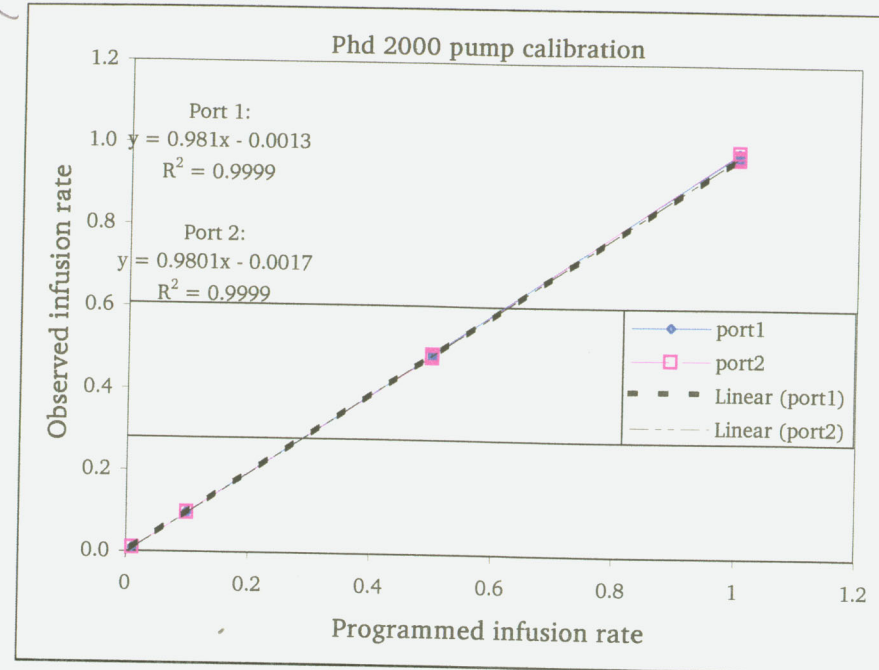


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Note, These calcites from 42600 are not different
in appearance from the samples ~~inspected~~ 5/14/2000
inspected by SEM labeled 42400 and calcite.

Pages 151 Through 155 Are Intentionally
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11/14/1999
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programmed infuse ml/min	% differences:				
	port1 ml/min	port2 ml/min	port1- port2	pump- port 1	pump- port 2
0.01	0.0095	0.0095	-0.197	4.77	4.80
0.01	0.0094	0.0094	0.011	5.56	5.90
0.01	0.0096	0.0095	0.399	4.27	4.88
0.1	0.0962	0.0960	0.171	3.81	4.14
0.1	0.0966	0.0960	0.609	3.39	4.13
0.1	0.0974	0.0969	0.554	2.58	3.22
0.5	0.4870	0.4854	0.325	2.60	3.00
0.5	0.4830	0.4819	0.240	3.39	3.76
0.5	0.4913	0.4897	0.328	1.74	2.10
1	0.9882	0.9890	-0.082	1.18	1.11
1	0.9750	0.9789	-0.396	2.50	2.16
1	0.9734	0.9720	0.140	2.66	2.88
1	0.9849	0.9779	0.717	1.51	2.26

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11/14/1999
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Pump Calibration

Objective:

Calibrate the pump infusion rate and the ml of solution delivered per unit time.

Materials:

- Harvard Apparatus PHD 2000 Infusion/Withdrawal Dual Syringe pump.
- 2 30ml Luer lock syringes + tubing(per run)
- Nanopure water
- Bottles to hold water.

Identify 'front' syringe port as "port #1" and 'back syringe port as "port #2". Fill syringes, taking care to get rid of any air bubbles. Attach syringes to pump. Before each run, run pump at the desired rate for a few minutes - until 3 drops of water exit the syringes - to eliminate any potential initial start-up errors. Repeat calibration annually.

programmed infuse ml/min	Port 1 bottle1 mass g	Port 2 bottle1+h2o mass g	Port 2 bottle2 mass g	Port 2 bottle2+h2o mass g	time sec	ml pump reading	port 1 ml measured	port 2 ml measured	port 1 rate calculated ml/min	port 2 rate calculated ml/min
1	19.7205	29.7766	11.1973	21.2617	610.59	10.176	10.0561	10.0644	0.988169	0.988984
0.5	11.2093	19.2164	11.2091	19.1903	986.47	8.2081	8.0071	7.9812	0.487015	0.48544
0.1	11.8161	20.4731	11.1974	19.8396	5400	8.9975	8.657	8.6422	0.096189	0.096024
0.01	11.1787	21.1538	11.0973	21.0328	62521	10.42	9.9751	9.9355	0.009573	0.009535
1	11.2347	20.5075	11.1837	20.4934	570.62	9.5518	9.2728	9.3097	0.975024	0.978904
0.5	20.5075	29.2048	20.4934	29.1699	1080.32	9.0018	8.6973	8.6765	0.48304	0.481885
0.1	29.2048	38.3864	29.1699	38.2959	5702	9.5003	9.1816	9.126	0.096615	0.096029
1	38.3864	56.1976	38.2959	56.0822	1097.89	18.295	17.8112	17.7863	0.973387	0.972026
0.5	11.1715	21.1579	11.1843	21.1381	1219.58	10.161	9.9864	9.9538	0.491304	0.4897
0.01	11.1646	20.5259	11.2165	20.5963	58980	9.8261	9.3613	9.3798	0.009523	0.009542
1	20.3859	28.271	20.4263	28.2553	480.36	8.0065	7.8851	7.829	0.984899	0.977892
0.1	28.271	36.6494	28.2553	36.5875	5160.36	8.5975	8.3784	8.3322	0.097416	0.096879
0.01	11.2003	20.8555	11.169	20.8231	61341	10.223	9.6552	9.6541	0.009444	0.009443

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12/13/99
M Nugent

liter stock solution: 0.006m/l Na:
IN PPM!!!

Ca		Na		Vol ppm St ml ug/l		Vol. Ca St to add:	Actual Mass Added:		Calculated concentrations:	
ppm	m/l	ppm	m/l				Ca mass (G)	H2O: mass (G)	Ca m/l	ppm
Blank	0	0.0E+0	143 0.0062	100	1000	0.000	Blank		0	0
			0							
1	0.5	1.2E-5	143 0.0062	100	1000	0.050	1	0.0505	101.415	1.24E-5 0.498
2	1	2.5E-5	143 0.0062	100	1000	0.100	2	0.1017	100.0006	2.54E-5 1.017
3	2	5.0E-5	143 0.0062	100	1000	0.200	3	0.2022	99.8569	5.05E-5 2.025
4	4	1.0E-4	143 0.0062	250	1000	1.000	4	1.0106	252.0808	1.00E-4 4.009
5	5	1.2E-4	143 0.0062	100	1000	0.500	5	0.5016	104.0839	1.20E-4 4.819

Standards are prepared using Fisher ACS Reagent
grade NaCl (Lot #) and Fisher AA
Standard for Calcium, Lot # 986835-24, expiration
date Oct. 2000.

0.006m NaCl solution prepared - This solution will be
used as the AA Standard Stock Solution.

EC
6/13/00

AA standards for Calcium analysis:

M Nugent
12/13/99

AA standards for calcium analysis
Atomic Adsorption Standards

IN PPM!!!		Na		Vol ppm St ml ug/l		Vol. Ca St to add:	Actual Mass Added:		Calculated concentrations:	
Ca ppm	m/l	ppm	m/l				Ca mass (G)	H2O: mass (G)	Ca m/l	ppm
Blank	0	0.0E+0	143 0.01	60	1000	0.000	Blank		0	0
High St.	10	2.5E-4	143 0.01	60	1000	0.600				
1	0.5	1.2E-5	143 0.01	30	1000	0.015	1	0.015	101.415	1.24E-5 0.498
2	1	2.5E-5	143 0.01	30	1000	0.030	2	0.030	100.0006	2.54E-5 1.017
3	2	5.0E-5	143 0.01	30	1000	0.060	3	0.060	99.8569	5.05E-5 2.025
4	4	1.0E-4	143 0.01	30	1000	0.120	4	0.120	252.0808	1.00E-4 4.009
5	5	1.2E-4	143 0.01	30	1000	0.150	5	0.150	104.0839	1.20E-4 4.819

1 liter stock solution: 0.006m/l Na:
use 0.351g/L nanopure

0.3526 g NaCl per 1000.0 g nanopure water used.

M Nugent

Surface Area Analysis

Coulter SA 3100 Surface Area and Pore Size Analyzer
Analysis Report

page 1/7

MT Nugent

Serial No.	W46020	Software Version	2.11
Sample ID	CAL-1	Start Date	07/31/89
Customer	CNWRA	Start Time	23:50:26
Operator	NUGENT	Elapsed Time	19 min
Sample Wt	3.0374 g	Outgas Time	720 min
Profile	BET5	Outgas Temperature	350 C

Summary

Surface Area Report

BET Surface area 0.257 sq.m/g
Correlation Coefficient 0.99987

This notebook appears
to comply with QAP-001.

E.C. Pen
6/13/2000

Average the 3 Surface
area analyses:

0.257
0.257
0.246

I have reviewed this AVE. 0.253 m²/g
notebook again. It still complies with
QAP-001. There is still sufficient
information for another qualified person
to repeat the activities.

E.C. Pen
8/28/2000

Surface Area Analysis

Coulter SA 3100 Surface Area and Pore Size Analyzer
Analysis Report

page 2/7

Serial No.	w46020	Software Version	2.11
Sample ID	CAL-1	Start Date	07/31/89
Customer	CNWRA	Start Time	23:50:26
Operator	NUGENT	Elapsed Time	19 min
Sample Wt	3.0374 g	Outgas Time	720 min
Profile	BET5	Outgas Temperature	350 C

Surface Area Report

BET Surface area 0.257 sq.m/g

Slope	16.849601
Intercept	0.090214
C_value	187.774
Monolayer Volume	0.0590 cc/g (STP)
Correlation Coefficient	0.99987

One Point BET Surface Area (Ps/Po=0.3) 0.254 sq.m/g

Analysis Data Ps/Po	BET Function	Vads cc/g(STP)
0.0506	0.929330	0.057
0.0685	1.242188	0.059
0.0965	1.551438	0.061
0.1044	1.860112	0.063
0.1203	2.166105	0.064
0.1761	3.044414	0.070

Interpolated Data Ps/Po	BET Function	Vads cc/g(STP)
0.0500	0.932694	0.056
0.0800	1.438182	0.060
0.1200	2.112166	0.065
0.1600	2.786153	0.069

This notebook appears
to comply with QAP-001.

E.C. Pen
6/13/2000

Average the 3 surface
area analyses:

6.257
0.257
0.246

I have reviewed this AVE. 0.253 m²/g
notebook again. It still complies with
QAP-001. There is still sufficient
information for another qualified person
to repeat the activities.

E.C. Pen
8/28/2000

Surface Area Analysis

page 3/7

Coulter SA 3100 Surface Area and Pore Size Analyzer
Analysis Report

Serial No.	W46020	Software Version	2.11
Sample ID	CAL-1	Start Date	07/31/89
Customer	CNWRA	Start Time	23:50:26
Operator	NUGENT	Elapsed Time	19 min
Sample Wt	3.0374 g	Outgas Time	720 min
Profile	BET5	Outgas Temperature	350 C

Isotherm Data

Freespace Calculation

Slope 0.03144
Intercept -0.005536
Correlation Coefficient 1.00000

Isotherm Data Table

Ps/Po	Vads cc/g(STP)	Ps mmHg	Po mmHg
0.0000	0.000	0.001	746.36
0.0098	0.047	7.311	746.42
0.0231	0.053	17.238	746.68
0.0410	0.056	30.611	746.77
0.0506	0.057	37.755	746.76
0.0685	0.059	51.193	746.81
0.0865	0.061	64.585	746.83
0.1044	0.063	77.945	746.69
0.1223	0.064	91.364	746.76
0.1761	0.070	131.540	746.80
0.2297	0.078	171.539	746.87

This notebook appears
to comply with QAP-001.

E.C. Pen
6/13/2000

Average the 3 surface
area analyses:

6.257
0.257
0.246

I have reviewed this AVE. 0.253 m²/g
notebook again. It still complies with
QAP-001. There is still sufficient
information for another qualified person
to repeat the activities.

E.C. Pen
8/28/2000

Surface Area Analysis

Coulter SA 3100 Surface Area and Pore Size Analyzer
Analysis Report

page 4/7

Serial No.	W46020	Software Version	2.11
Sample ID	CAL-2	Start Date	08/01/89
Customer	CNWRA	Start Time	00:24:52
Operator	NUGENT	Elapsed Time	19 min
Sample Wt	3.0055 g	Outgas Time	720 min
Profile	BET5	Outgas Temperature	350 C

Summary

Surface Area Report

BET Surface area 0.257 sq.m/g
Correlation Coefficient 0.99966

This notebook appears
to comply with QAP-001.

E.C. Per
6/13/2000

Average the 3 surface
area analyses:

0.257
0.257
0.246

I have reviewed this AVE. 0.253 m²/g
notebook again. It still complies with
QAP-001. There is still sufficient
information for another qualified person
to repeat the activities.

E.C. Per
8/28/2000

Surface Area Analysis

Coulter SA 3100 Surface Area and Pore Size Analyzer
Analysis Report

page 517

Serial No. W46020 Software Version 2.11

Sample ID CAL-2 Start Date 08/01/89
Customer CNWRA Start Time 00:24:52
Operator NUGENT Elapsed Time 19 min
Sample Wt 3.0055 g Outgas Time 720 min
Profile BET5 Outgas Temperature 350 C

Surface Area Report

BET Surface area 0.257 sq.m/g

Slope 16.772084
Intercept 0.134657
C_value 125.554
Monolayer Volume 0.0591 cc/g (STP)
Correlation Coefficient 0.99966

One Point BET Surface Area (Ps/Po=0.3) 0.253 sq.m/g

Analysis Data

Ps/Po	BET Function	Vads cc/g(STP)
0.0506	0.960831	0.055
0.0623	1.169452	0.057
0.0803	1.485528	0.059
0.0982	1.796273	0.061
0.1162	2.104226	0.062
0.1341	2.405966	0.064
0.1878	3.255938	0.071

Interpolated Data

Ps/Po	BET Function	Vads cc/g(STP)
0.0500	0.973261	0.054
0.0800	1.476424	0.059
0.1200	2.147307	0.064
0.1600	2.818190	0.068
0.2000	3.489074	0.072

This notebook appears
to comply with QAP-001.

E.C. Pen
6/13/2000

Average the 3 Surface
area analyses:

6.257
0.257
0.246

I have reviewed this AVE. 0.253 m²/g
notebook again. It still complies with
QAP-001. There is still sufficient
information for another qualified person
to repeat the activities.

E.C. Pen
8/28/2000

Surface Area Analysis

Coulter SA 3100 Surface Area and Pore Size Analyzer
Analysis Report

page 6/7

Serial No. W46020 Software Version 2.11

Sample ID CAL-2 Start Date 08/01/99
 Customer CNWRA Start Time 00:24:52
 Operator NUGENT Elapsed Time 19 min
 Sample Wt 3.0055 g Outgas Time 720 min
 Profile BET5 Outgas Temperature 350 C

Isotherm Data

Freespace Calculation

Slope 0.03141
 Intercept -0.004417
 Correlation Coefficient 1.00000

Isotherm Data Table

Ps/Po	Vads cc/g(STP)	Ps mmHg	Po mmHg
0.0000	0.000	0.001	747.28
0.0099	0.044	7.430	747.13
0.0232	0.051	17.313	747.19
0.0410	0.054	30.675	747.37
0.0506	0.055	37.809	747.36
0.0623	0.057	46.589	747.27
0.0803	0.059	60.014	747.31
0.0982	0.061	73.359	747.19
0.1162	0.062	86.832	747.36
0.1341	0.064	100.240	747.34
0.1878	0.071	140.357	747.42
0.2412	0.079	180.259	747.35

This notebook appears
 to comply with QAP-001.

E.C. Pen
 6/13/2000

Average the 3 surface
 area analyses:

6.257
 0.257
0.246

I have reviewed this AVE. 0.253 m²/g
 notebook again. It still complies with
 QAP-001. There is still sufficient
 information for another qualified person
 to repeat the activities.

E.C. Pen
 8/28/2000

Surface Area Analysis

page 7/7

Coulter SA 3100 Surface Area and Pore Size Analyzer
Analysis Report

Serial No.	W46020	Software Version	2.11
Sample ID	CAL-3	Start Date	08/01/89
Customer	CNWRA	Start Time	01:11:08
Operator	NUGENT	Elapsed Time	20 min
Sample Wt	3.0218 g	Outgas Time	720 min
Profile	BET5	Outgas Temperature	350 C

Summary

Surface Area Report

BET Surface area	0.246 sq.m/g
Correlation Coefficient	0.99980

This notebook appears
to comply with QAP-001.

E.C. Pen
6/13/2000

Average the 3 surface
area analyses:

6.257
0.257
0.246

I have reviewed this AVE. 0.253 m²/g
notebook again. It still complies with
QAP-001. There is still sufficient
information for another qualified person
to repeat the activities.

E.C. Pen
8/28/2000

Equations used in spreadsheets

M. Nugent
6/9/2020

This page belongs with CNWRA Controlled Copy #361, and describes the equations used in the spreadsheets on the disk in this envelope.

Filename: sampling.xls

Worksheet: sample prep

$$\% \text{ sample} = \frac{(\text{mass of sample})}{(\text{mass, sample} + \text{mass acid})} \times 100.$$

Worksheet: LSA-Np

- Efficiency Correction = (CPMA counts * ϕ) + CPMA counts

Efficiency correction is assumed to be 1.

- # atoms = $\frac{\text{efficiency}}{6.16 \times 10^{-13}}$ where $\lambda = \frac{0.693}{T_{1/2}} = 6.16 \times 10^{-13}$

- # moles = # atoms $\times \frac{1 \text{ mole}}{6.022 \times 10^{23} \text{ atoms}}$

- m/L = # moles $\times \frac{1000 \text{ mL}}{L} \times \frac{1}{\text{Sample mass, g}}$

- ppb = (conc., m/L) $\times 237.048 \text{ g/mL} \times 1000^2$

Worksheet: LSA-Tc

Same equations as for worksheet LSA-Np

but using $T_{1/2}$ for Tc, so $\lambda = 6.19 \times 10^{-12}$

and 98.907 g/mol ^{MW} 6/9/2020

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