UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

MAR 30 1984

MEMORANDUM FOR: M. Silberberg

FROM: R. O. Meyer

SUBJECT: VAPORIZATION AND AEROSOL GENERATION FROM SILVER ALLOY CONTROL RODS

Parts I and II of the enclosure describe basic concepts and available experimental evidence on this subject. Part III gives some related discussion. Conclusions are scattered throughout the enclosure, but they are indented and numbered for easy reference. Surprisingly, I have made some progress in understanding control-rod aerosol behavior, but there is still a long way to go. I think you will find the conclusions interesting.

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Enclosure:
As stated

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Enclosure

VAPORIZATION AND AEROSOL GENERATION FROM SILVER ALLOY CONTROL RODS

R. O. Meyer

Accident Source Term Program Office

U.S. Nuclear Regulatory Commission

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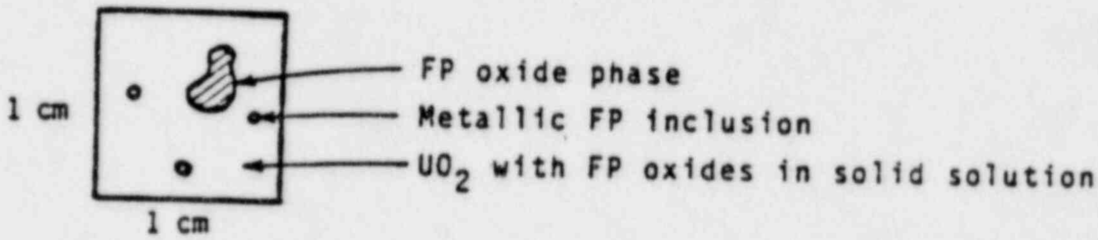
Part I Basic Concepts of Vaporization

Fission Products

Consider a square centimeter of exposed surface area of a piece of fuel.



If N_i is the atom fraction of species "i" in the fuel, then approximately N_i of the surface lattice sites on this square centimeter will be occupied by "i" atoms whether they are in solid solution or not. Since vapor pressure is a measure of the number of atoms or molecules impinging on a surface or jumping off a surface, the number of atoms or molecules of species "i" leaving the square centimeter of fuel will be proportional to its partial pressure p_i. At equilibrium, the number of atoms returning would equal the number leaving with no net loss. But the reactor atmosphere does not provide an equilibrium back-pressure nor is it a vacuum. Therefore, n_i the net number of species "i" leaving the surface is

$$n_i = \text{cons't}\ p_i, \tag{1}$$

where the constant contains some mass-transfer parameters as well as kinetic parameters like Boltzmann's constant.

Assuming Raoult's law holds, the partial pressure is just N_i times the pressure p_i^o over the pure species, so Eq. (1) becomes

$$n_i = \text{cons't}\ N_i p_i^o. \tag{2}$$

CORSOR-M was based on fission product data rather than data for structural materials, so the pre-exponential constant for CORSOR-M corresponds to S/V for fuel rather than S/V for cladding or structural materials. To assess the relative difference, I compared S/V for a typical cracked fuel geometry with S/V for a typical piece of LWR cladding. For a fuel pellet with six pie-shaped wedges, each broken across the middle,* I calculated an S/V of about 25 cm^{-1}. For a piece of cladding (or sheet metal) with a typical thickness of 0.7 mm, S/V is 29 cm^{-1}.

Conclusion No. 3: Prior to melting, cracked fuel has about the same S/V as cladding and structural materials.

This apparent similarity may explain why CORSOR-M agreed well with SASCHA data on structural material releases for Sn and why it agreed approximately with the data for Mn, Cr, and Fe. In fact, the modest overprediction for Mn, Cr, and Fe might have resulted from molten stainless steel (M.P. approx. 1450°C) assuming a lower S/V in the SASCHA test.

Ag-In-Cd Control Rod Material

While S/V for fuel fragments and structural metals is constant or perhaps mildly varying, S/V for the Ag-alloy control material cannot be expected to be well behaved. Silver is violently expelled from its cladding at about 1400°C (see Part II); in some cases it has wetted adjacent surfaces (presenting a large S/V); in other cases it has run down and frozen in cooler regions (presenting a smaller S/V). One would therefore expect to see a wildly varying release rate coefficient k for control-rod Ag because of the marked variations with time in S/V. This is precisely the kind of behavior seen in the SASCHA control-rod Ag tests in sharp contrast to the steady k value for fission-product Ag in the SASCHA tests (see Fig. 2 in Part II).

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*Based on a discussion with L. A. Neimark of ANL.

is reduced, but this brings the estimated value down to only 1.5% whereas Parker measured a low 0.34%.

Conclusion No. 4: S/V for control-rod silver appears to vary widely in the tests available to date.

Conclusion No. 5: Major variations in release of control-rod silver in available tests appear to be explainable on the basis of S/V effects.

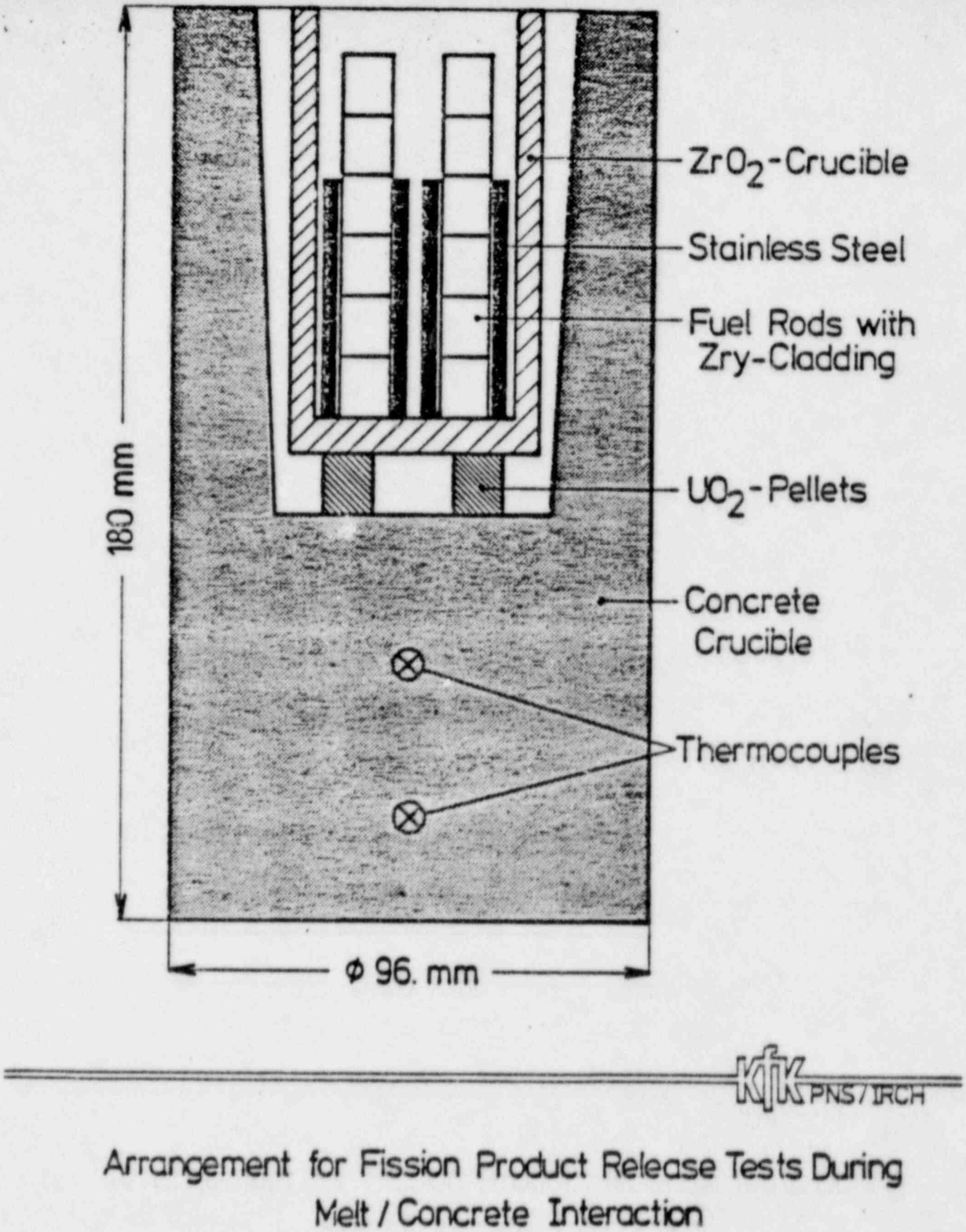


Fig. 1. Diagram of SASCHA test apparatus.

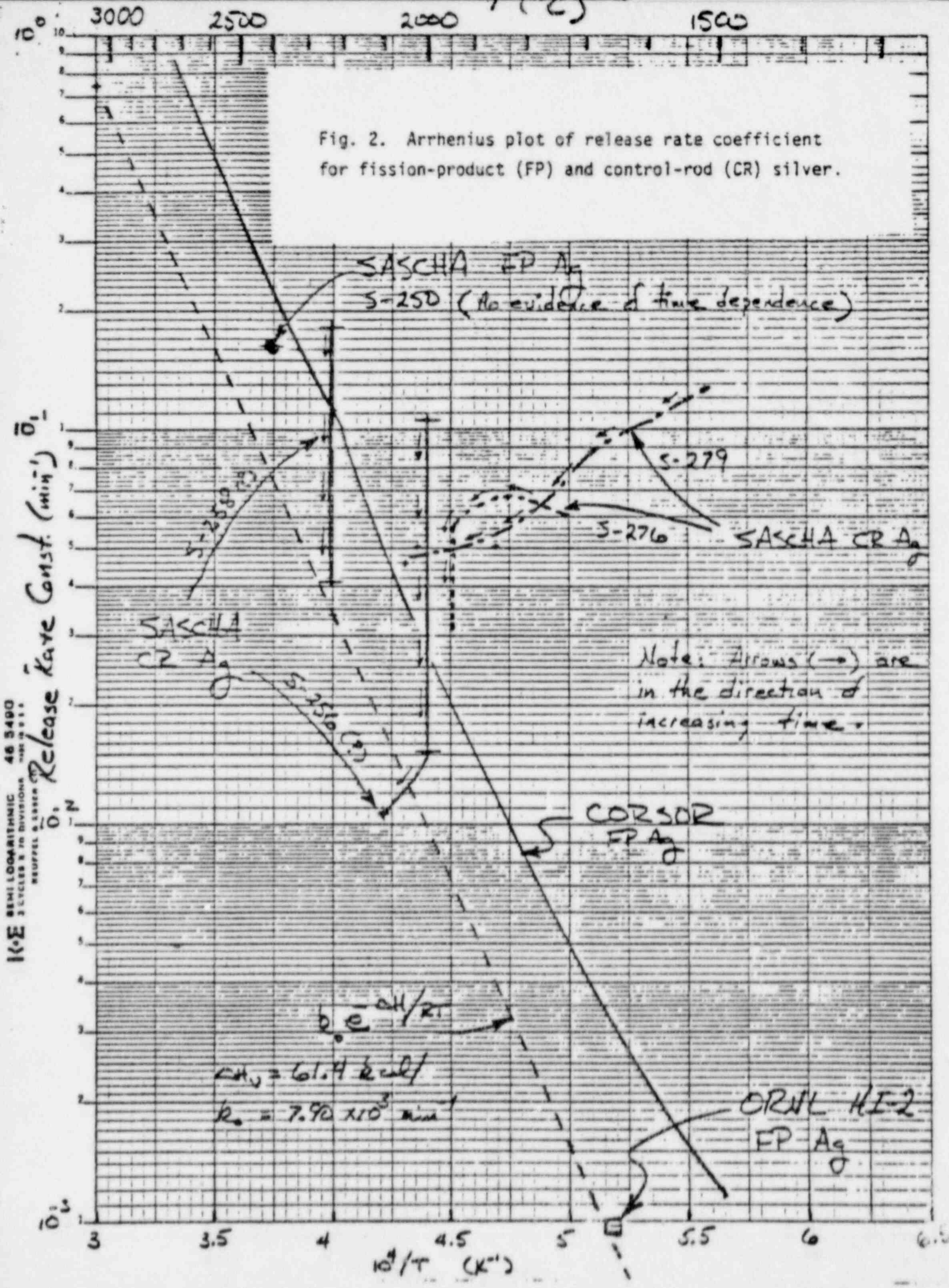


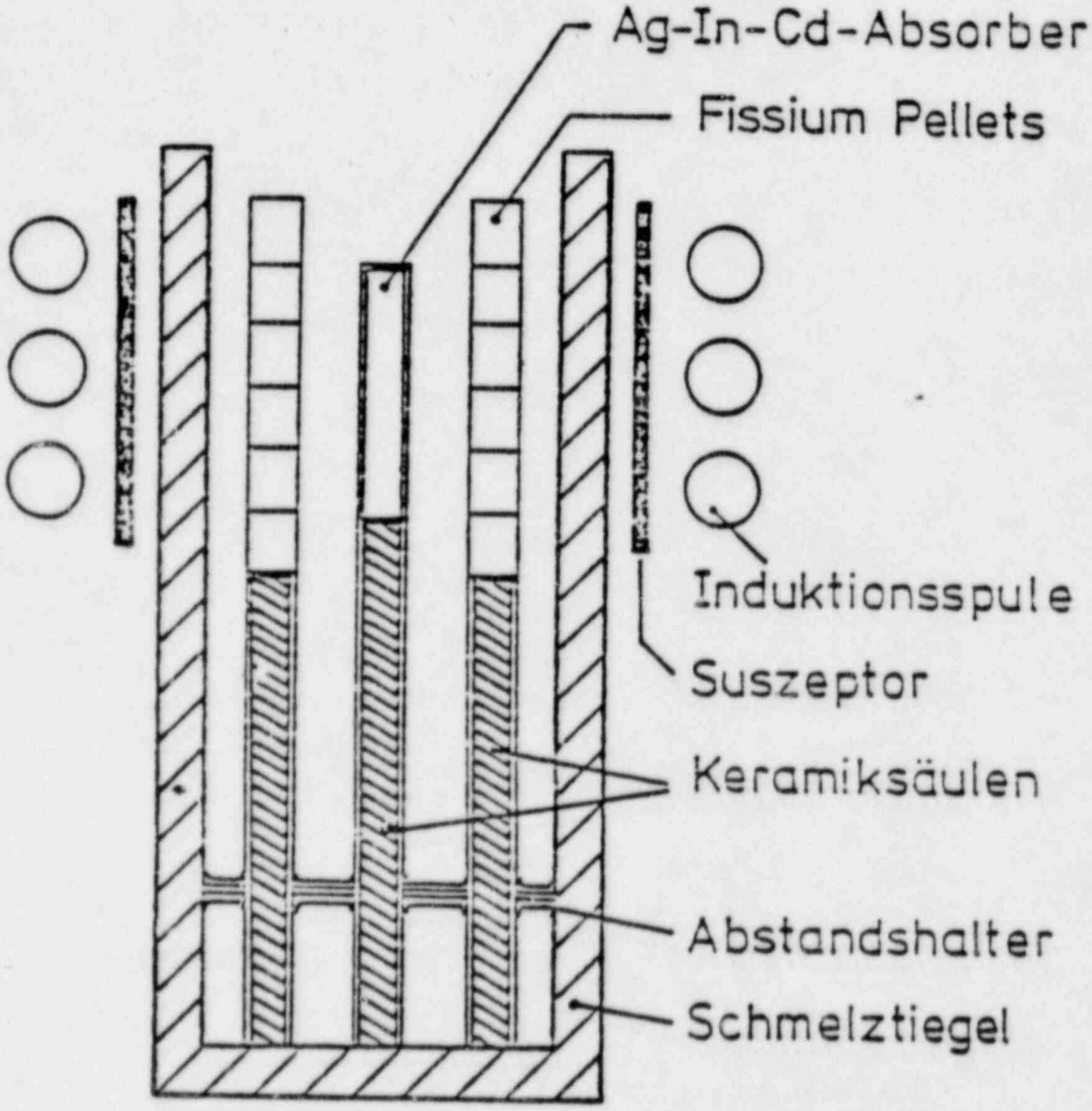
Fig. 2. Arrhenius plot of release rate coefficient for fission-product (FP) and control-rod (CR) silver.

Albrecht has concluded that "there is no doubt that the evaporation of Ag from real control rods does not proceed in the same way as in the SASCHA crucible." To remedy this deficiency, Albrecht has designed a large crucible (Fig. 3, from Ref. 6) for tests in 1984. This crucible will allow the liquid Ag alloy to run down the test rods toward colder regions of the "core" in a more prototypical manner.

Parker

In 1981, 1982, and 1983, Parker conducted three tests (CM-16, CM-18, and CM-30) involving Ag-In-Cd control rods (Refs. 8-10). His test setup was similar to Albrecht's inasmuch as test rods were contained in a crucible, heated by induction heating, and contained fission product simulants. However, Parker's test specimens appear to be more prototypical than Albrecht's (see Fig. 4, from Ref. 11), and the Ag alloy control material is sealed in a stainless steel tube (welded end caps) as it would be in an LWR. Parker's induction coil is on an elevator so he can move the heated zone axially. Thus it appears that his temperatures have less axial uniformity than Albrecht's; this is especially true in test CM-18, in which the test bundle was almost twice as long as in the other tests and "caused a significant part of the bundle to be outside the influence of the RF induction field."

Parker's temperatures are, unfortunately, poorly known. In all cases the tests were heated on a ramp until the fuel bundle specimen was completely melted down, thus fixing the final test temperatures at about 2400°C. However, intermediate temperatures were often estimated by visual sightings since pyrometer difficulties were experienced in the smoky chamber. The time required to reach these estimated temperatures was usually recorded to establish the heating rate. From approximate time and temperature values reported, I have constructed approximate temperature histories in Fig. 5. Total reported releases are given in Table 1. It is clear from these results that the 7-in.-long CM-18 bundle produced a much smaller release of Ag than did the 4-in.-long CM-16 and CM-30 bundles. It is important, however, to remember that in all three cases the silver was eventually raised to about 2400°C as each bundle melted down into the crucible.



Konzeption für die Messung der AgJ-Bildung

Fig. 3. Diagram of new SASCHA crucible for tests with longer specimens.

Table 1. Total percentage released in Parker's tests

	Ag	In	Cd
CM-16	6.1	5.4	53
CM-18	0.34	0.47	44.9
CM-30	2.98	0.14	67.6

Parker made several important visual observations during these tests:

(a) The control rod cladding ruptured at around 1400°C with a forceful ejection of molten Ag alloy. This obsevation is in agreement with Hagen's observation.

(b) The dramatic rupture produces a cloud of dense black smoke that is nearly pure Cd, and this smoke is promptly carried away by the hydrogen being generated by the Zr-water reaction.

(c) Zircoloy cladding forms a low-melting alloy with silver and sloughs off or melts off in a process that Parker refers to as "candling."

Like Albrecht, Parker is concerned about scale effects and the ability of liquid materials to flow naturally down a long bundle rather than being artificially held in a high temperature zone in a crucible. Therefore, Parker is planning to run 10 kg tests in the near future in contrast to the nominal 1 kg tests described above.

TMI-2

Some of the filter debris from TMI-2 have already been examined (Ref. 12) and found to contain significant concentrations (approx. 15 w/o) of Ag, In, and Cd from control rods. However, the principal metal component of the debris was Zr, and about 6 w/o of the debris was uranium (presumably as UO_2). Zirconium and UO_2 have very low volatilities even at core melt temperatures, so these debris probably reached the filters by being flushed out of the primary system rather

Part III Discussion

As of early 1984, the behavior of Ag-alloy control rod materials in a severe accident is poorly understood and very important. On one hand, IDCOR believes that the liquid Ag alloy runs down to the lower head where it is quenched thereby contributing nothing to the aerosol. On the other hand, NRC's contractors assume (a) that 5% of the Ag is released at 1400°C, (b) that this value is increased linearly to 50% at 2300°C, and (c) that this value is again increased linearly to 100% at 2800°C. For the Surry reanalysis in BMI-2104, Vol V, this assumption causes Ag to dominate the aerosol mass (60 to 80% of the aerosol is Ag, Ref. 14).

Complete answers are not available yet, but several firm conclusions seem to follow from the experimental work summarized in Part II.

> Conclusion No. 6. The stainless steel cladding of Ag-In-Cd control rods ruptures at about 1400°C and is accompanied by an energetic expulsion of the liquid Ag alloy.

> Conclusion No. 7. Cadmium, by virtue of its extremely high vapor pressure (B.P. = 767°C), flashes when the cladding ruptures producing a dense, black, smoky aerosol that contains a large fraction of the total Cd.

> Conclusion No. 8. After the control rod cladding ruptures, liguid Ag alloy will flow down adjacent surfaces to cooler regions.

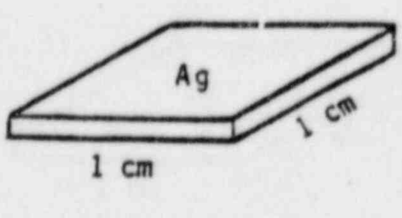
Of course this last conclusion is of little value unless one knows how far down the Ag alloy flows and if it leaves a wetted film or alloyed surface (large S/V) behind. Though qualitative, Parker's results provide rather convincing evidence that the Ag alloy leaves the heated zone rather completely without forming large amounts of aerosol. Albrecht's results do not appear to be in contradiction since the liquid silver in SASCHA was apparently directly heated by the induction field and did not find a colder region to flow to. At this time the TMI-2

to permit a credible result. Because the ORNL interim recommendation was based on only part of the data now available and was influenced by a questionable calculation, I believe a re-evaluation would be appropriate.

Finally, there is a popular belief that if control-rod Ag becomes alloyed with other materials, its vapor pressure will be reduced and hence its release rate will be suppressed. The following three quotations reveal the wide-spread nature of this belief.

> Parker, NUREG/CP-0027, 1982, p. 1083.
>
> The same processes [e.g., alloying and fuel and cladding ($Zr-ZrO_2-UO_2$) eutectic melting] which lead to the lower melting temperatures also lead to lower vaporization rates of many of the otherwise volatile species by inherent vapor suppression of a dilute species in a less volatile one.
>
> Wichner (Lorenz) letter to Jankowski, February 28, 1983, p. 29.
>
> Therefore, <u>if conditions allow dissolution in Zr</u>, the vapor pressure of these materials would be reduced approximately by its local mole fraction in the resulting alloy (Raoult's law).
>
> BMI-2104, Vol. I, 1983, p. 5-10.
>
> Preliminary experimental evidence suggests that when the control rods burst, the liquefied silver will be expelled from the rods and will form a solution with the zirconium cladding which has a lower vapor pressure and thereby greatly reduces the participation of silver in the aerosol formation.

It is, of course, true that the partial pressure of Ag over an alloy would be only a fraction (approximately its atom fraction in the alloy) of the vapor pressure over pure Ag. But this does not lead to the conclusion that the Ag release rate would be reduced as can be seen with the following simplified example. Consider a square centimeter of Ag sheet metal.



$$p_{Ag} = p^o_{Ag}$$

Part IV References

1. S. Hagen, Projekt Nukleare Sicherheit Jahresberichte, KfK-2700, November 1978, p. 89 and p. 4300-29.

2. S. Hagen, Projekt Nukleare Sicherheit Jahresberichte, KfK-2750, October 1979, p. 90 and p. 4300-46.

3. H. Albrecht et al., Projekt Nukleare Sicherheit Jahresberichte, KfK-3250, June 1982, p. 48 and p. 4300-36.

4. H. Albrecnt and H. Wild, Proc. ANS Cambridge Conf., August 1983, p. 4.2-1.

5. H. Albrecht et al., Projekt Nukleare Sicherheit Jahresberichte, KfK-3350, July 1983, p. 4300-20.

6. S. O. Peck and D. K. Peck (NRC delegates at KfK) letter to H. Scott (NRC), November 27, 1983.

7. M. F. Osborne et al., Proc. ANS Cambridge Conf., August 1983, p. 4.1-1.

8. G. W. Parker et al., ORNL Aerosol Release and Transport Program Quarterly Progress Report, NUREG/CR-2299, Vol. 4, May 1982, p. 15.

9. G. W. Parker et al., ORNL Aerosol Release and Transport Program Quarterly Progress Report, NUREG/CR-2809, Vol. 1, August 1982, p. 11.

10. G. W. Parker (ORNL) private communication with R. O. Meyer (NRC), March 9, 1984.

11. G. W. Parker et al., Proc. Chicago Conf., NUREG/CP-0027, Vol. 2, February 1983, p. 1078.

STATUS OF SOURCE TERM CODE PACKAGE

- CODE PACKAGE NOW BEING USED IN SARRP
- OBSOLETE MODEL OPTIONS BEING REMOVED IN PREPARATION FOR PUBLIC RELEASE
- RELEASE OF DOCUMENTATION AND TAPES EXPECTED IN DECEMBER
- MANUAL FOR CODE PACKAGE WILL RELY ON MANUALS FOR INDIVIDUAL CODES

ASSESSMENT OF STCP

- o DEVELOPMENT
  DESCRIBED IN INDIVIDUAL CODE MANUALS, WHICH HAVE BEEN PUBLISHED. MANUAL DESCRIBING RECENT CHANGES TO BE ISSUED SOON.

- o VERIFICATION
  QA CHECKING OF STCP (LINE BY LINE) AND OF SARRP CALCULATIONS (INPUT AND OUTPUT) IS IN PROGRESS AT AN INDEPENDENT LABORATORY (BNL).

- o VALIDATION
  ADDITIONAL DATA COMPARISONS FOR INDIVIDUAL CODES IN STCP MADE IN CONNECTION WITH OUR EXPERIMENTAL PROGRAMS. SEE ALSO ORNL/TM-8842. DATA COMPARISONS FOR REFERENCE VERSION OF STCP SCHEDULED TO START AT BNL IN EARLY CY86. STEP-BY-STEP COMPARISON OF STCP WITH CONSERVATION LAWS TO BE DONE AT BCL IN FY 86.

- o STANDARD PROBLEMS
  STANDARD PROBLEMS WITH REFERENCE VERSION OF STCP PLANNED AT BNL FOR CY 86.

- o UNCERTAINTY STUDIES
  SCOPING STUDY CALLED QUEST DESCRIBED IN SAND84-0410. FOLLOW-ON STUDY UNDERWAY AT BNL.

- o PEER REVIEW
  DESCRIBED IN NUREG-0956. ACRS REVIEW AND PUBLIC COMMENT EVALUATION IN PROGRESS.

ADDITIONAL INFO. ON
STATUS SOURCE TERM CODE PACKAGE

(1) CODE PACKAGE BEING USED IN SARRP
- o 6 PEACH BOTTOM SEQUENCES COMPLETED
- o 9 SEQUENCES IN PROGRESS FOR SEQUOYAH, GRAND GULF, AND SURRY
- o APPROX. 10 MORE SEQUENCES SCHEDULED

(2) NUMEROUS OBSOLETE OR UNUSED MODEL OPTIONS ARE BEING REMOVED FROM THE CODE PACKAGE IN PREPARATION FOR PUBLIC RELEASE.

(3) COMPLETION OF CODE MANUAL AND RELEASE OF TAPES EXPECTED IN DECEMBER. CODE PACKAGE WILL BE AVAILABLE DIRECTLY FROM BCL AND THROUGH THE NATIONAL ENERGY SOFTWARE CENTER.

(4) CODE MANUAL FOR THE PACKAGE WILL REFER TO CODE MANUALS FOR INDIVIDUAL CODES FOR BASIC DESCRIPTION OF THE MODELS. THE MANUAL FOR THE CODE PACKAGE WILL DESCRIBE CHANGES THAT HAVE BEEN MADE, TELL HOW TO USE THE PACKAGE, AND PROVIDE SAMPLE PROBLEM INPUT AND OUTPUT.

RECENT CHANGES* IN SOURCE TERM CODE PACKAGE

(1) BWR MODELS IMPROVED

- CHANNEL BOX AND CONTROL BLADES MODELED EXPLICITLY
- REALISTIC RELIEF VALVE MODEL INCORPORATED
- CREDIT FOR RETENTION IN REACTOR BUILDING

(2) AG-IN-CD RELEASES IN PWRS REDUCED IN AGREEMENT WITH RECENT DATA

(3) INCONSISTENT DECAY HEAT MODELS IN MARCH AND CORCON CORRECTED

(4) CONSISTENT SET OF FISSION PRODUCTS TRACKED THROUGHOUT CODES

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* CHANGES MADE PRIOR TO INITIATING CALCULATIONS FOR NUREG-1150.

NOTES FOR SLIDE B6 ON STCP CHANGES

(1) CHANNEL BOX AND CONTROL BLADE MODELS ARE IN MARCH 2, BUT WERE NOT USED IN BMI-2104. CREDIT WAS NOT GIVEN FOR REACTOR BUILDINGS IN BMI-2104. BMI-2104 ASSUMED THAT RELIEF VALVES HELD PRESSURE AT CONSTANT VALUE: NEW MODEL HAS A 50 PSI WINDOW, OR "DEAD BAND," RESULTING IN A VARYING PRESSURE.

(2) AG AND IN REDUCED BY 0.1x (TEN-FOLD REDUCTION) CD REDUCED BY 0.7x. STILL A CRUDE MODEL, WHICH WILL BE IMPROVED WHEN NEW DATA ARE FULLY EVALUATED. BIG REDUCTION IN AEROSOL MASS, BUT PROBABLY SMALL EFFECT ON FP TRANSPORT.

(3) MARCH2 WAS 8% TOO HIGH (SIMPLE VERSION OF ANS STANDARD) AND CORCON MOD2 WAS 12% TOO LOW (ORIGEN). HOWIE RICHINGS ENGINEERED THE FIX. TAL ENGLAND (LASL) AND DAVID BENNETT (SNL) CONCUR.

(4) EACH CODE'S I/O NOW COMBINED OR EXPANDED INTO 11 CAREFULLY CHOSEN GROUPS.

CORSOR:	13 SPECIES
TRAP-MELT:	9 SPECIES
VANESA:	22 GROUPS
NAUA:	1 GROUP