MEDICAL RADIATION CONSULTANTS, Inc.
2101 Prudence Drive
Dayton, Ohio 45431

Bruce T. Austin, Ph.D.
Medical Physicist

Ms. B.J. Holt
Regional Materials Licensing Section
USNRC Region III
799 Roosevelt Road
Glen Ellyn, Il 60137

October 29, 1985

Dear Ms. Holt;

This letter is forwarded directly to you at the request of Mr. Michael D. Huard, President of the Bradley - Thompson Tool Company, Southfield, Michigan and is in response to the letter of October 18, signed by Patricia Vacherlon for Dr. Bruce S. Mallett.

This information is in support of the Source Material License application pending for the Bradley - Thompson Tool Company, assigned control number 79420, and differs from a similar response, dated October 11, in that it responds to the order of the above letter rather than to my notes of our telephone call of August 26.

1. Mr. Huard has in excess of 20 years of experience in the processing of Magnesium-thorium alloys under the provisions of the general license issued under 10 CFR part 40. He is personally expert in precision machine tool operation and in management of machine tool operations of the Company. Since submission of the application, he has received training in the previously specified topics relevant to his role as Corporate Radiation Safety Officer.

2. Initial training of material users was conducted by Mr. Solari and myself as adjuncts to Mr. Huard. Subsequent training, particularly that associated with daily operating procedures required for compliance with license conditions has been conducted by Mr. Huard. Future training will be conducted by Mr. Huard with assistance by Mr. Solari or myself as he deems necessary.

3. Bioassays will be performed and evaluated by Mr. Solari or myself when indicated. Assay procedures will be those indicated by the exposure necessitating evaluation, rather than being limited to urinalysis.

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4. Air sampling procedures, instrumentation, radioassay and

calculation procedures are to be conducted in accordance with the enclosed description by Mr. Solari.

5. Survey instrument calibration will be performed by the instrument manufacturer or other person or organization authorized to calibrate survey instruments by the Nuclear Regulatory Commission.

6. Waste materials will be transferred to a licensee that is authorized to receive radioactive waste for disposition.

7. The consulting physicists will spend time on site as indicated by the scope of licensed activities and in accordance with the request of the Corporate Radiation safety Officer. On site time presently exceeds 20 hours and is anticipated to total in excess of 25 hours prior to initiation of licensed activities. Future time commitment are anticipated to be reduced as Mr Huard becomes familiar with license requirements and establishes routine operational procedures.

I regret the delay in your receipt of this information and the impact that the delay had had on the operations of the Tool Company. Should any question arise concerning matters pertaining to this application, please do not hesitate to contact me through the service at (513) 229-8933.

Sincerely,

Bruce T. Austin, Ph.D.
Consultant Physicist

cc. M. Huard

Thorium Particulate Activity Monitoring

Breathing Zone Sample

Samples for particulate radioactivity are taken in the breathing zone area of the worker. The samples are taken through a membrane filter for a period of 4 or more hours. The pump is capable of pulling 0.5 cfm. The sampling head is placed near the machinist so as to sample the air in his breathing zone but will not interfere with his normal working habits. At the end of the sampling period, the membrane filter is removed and placed in a small envelope marked with the date, location, and air volume. The sample will be stored for 48 hours or more to permit the decay of the short lived daughter products of radium and thorium.

Counting Procedure

Remove filters from envelope and place in new planchet.
Count filter on Gas flow proportional counter for 30 minutes.
(The voltage must be set on the alpha plateau.)
Count the background for an equivalent period of time.

Calculation Procedure

Calculate net count rate (net cpm) for the membrane filter.

Calculate the concentration of activity as follows:

Concentration = (net cpm)/(Eff) (2.22E6) (Air volume in milliliters)
where Eff = counter efficiency and 2.22E6 = dpm/microCurie

Calculate % MPC = concentration x 100 / (6E-11 uCi/ml)
where 6E-11 is the MPC for natural thorium (appendix B, 10CFR20)

Equipment required:

Membrane filter: Gelman or equivalent
Air Pump: WISA model DBGM
Gas flow detector: Nuclear Measurements Corporation

SENSITIVITY OF MEASUREMENTS

Air volume collected at 0.5 cubic feet per minute

Sampling Time	Air Volume
1 hours	14158 ml
2 hours	28316 ml
3 hours	42475 ml
4 hours	56632 ml
6 hours	84948 ml
8 hours	113264 ml

Background counting rate: 0.5 cpm
Counting efficiency: 50 %
Collection efficiency: 100 %

COUNTING AND REPORTING PRACTICES

Critical Level

The critical level is used only to determine if a measurement is statistically different from background. For equal sample and background counting time the critical level becomes: (ref 1)

Critical level = 1.65 x square root(2xRb/T) where:
1.65 is the one sided confidence level
Rb is the background counting rat and
T is the counting time.

The critical level is 0.30 cpm for Rb=0.5 and T=30 minutes.

The detection limit (LD) is:
LD = 2 x 1.65 x (square root (2xRb/T)) (ref 2)

The detection limit is 0.6 cpm for the conditions described above. This corresponds to a limit 1.20 dpm or 5.405E-7 uCi on the filter as a lower limit of detection. The overall limit of detection therefore varies with the volume of air sampled. Sampling for 4 hours gives a lower limit of 1.91E-11 uCi/ml or 0.315 MPC. Sampling for 8 hours results in a lower limit of detection of 9.55E-12 uCi/ml or 0.16 MPC.

Ref 1) Currie L.A. Limits for Qualitative Detection and Quantitative Determination. Analytical Chemistry, Vol 40, No. 3, Mar 1969

Ref 2) Hartwell, J.K. Detection limits f· Radioisotopic counting techniques ARH-2537, Jun 22, 1972

INTRODUCTION

Environmental surveillance and bioassay are two health physics disciplines in which it is necessary to measure trace quantities of radioactivity. A clear understanding of the statistical limit of radioactivity measurement (the "minimum detectable activity," or MDA) is therefore indispensable. Unfortunately, a careful review of the literature reveals numerous, and often discordant definitions relating to the detection limit of a counting instrument. This paper, tutorial in nature, presents several definitions (following principally Currie (1))

the critical level
the detection level
the less-than level
the determination limit

and discusses their application. In addition, the Loevinger-Berman criterion for optimizing counter performance, and the chi-square test for assessing counter performance are discussed.

NOTATION AND ASSUMPTIONS

The following notation will be used:

n = number of counts accumulated in a counting time T
σ_n = standard deviation in $\underline{n}$ counts
R = count rate = n/T
σ_R = standard deviation in count rate R

T_b = time to count background
R_b = background count rate
σ_b = standard deviation in R_b

T_t = time to count total (source plus background)
R_t = total count rate
σ_t = standard deviation in R_t

R_s = count rate from source alone
σ_s = standard deviation in R_s

K_o = ratio of observed to expected (Poisson) standard deviation

K_1 = normal distribution multiplier for one-sided confidence interval (e.g., for 95% confidence, $K_1 = 1.65$)

K_2 = normal distribution multiplier for two-sided confidence interval (e.g., for 95% conficence, $K_2 = 1.96$)

L_c = the critical level

L_d = the detection limit

L_ℓ = the less-than level

L_q = the determination limit

Counts occurring in a time interval are assumed to be Poisson-distributed (later, this requirement will be relaxed somewhat). The number of accumulated counts is assumed to be sufficiently large to permit the probability distribution of accumulated counts to be adequately approximated by a normal distribution having mean and variance equal to the expected number of counts. This is a reasonable assumption in almost all cases of practical interest: even when the number of accumulated counts is as small as twenty, the normal approximation is acceptable. The usual assumptions are made (e.g., independence of errors) which permit application of first-order propagation of errors theory (2).

With the above assumptions, the standard deviation σ, of n counts is

$$\sigma = \sqrt{n}$$

so

$$\sigma_R = \frac{\sqrt{n}}{T} = \sqrt{\frac{R}{T}} .$$

Also, if

$$c = a - b$$

$$\sigma_c^2 = \sigma_a^2 + \sigma_b^2$$

THE CRITICAL LEVEL

The Critical Level is defined as the net count rate which must be exceeded before the sample is said (at some degree of confidence) to contain measurable radioactivity above background. The Critical Level

$$L_c = K_1 \sigma_o$$

where K_1 is the one-sided confidence factor and σ_o is the standard deviation of zero net count rate. If we wish a 95 percent confidence level (five percent of background counts will be judged to have radioactivity above background), then $K_1 = 1.65$. The factor K_o is, for the present, taken equal to 1, and will be discussed later.

Since $R_s = R_o = R_t - R_b = 0$

$$\sigma_o^2 = \sigma_t^2 + \sigma_b^2$$

$$\sigma_t^2 = \frac{R_t}{T_t} = \frac{R_b}{T_t}$$

$$\sigma_b^2 = \frac{R_b}{T_b}$$

So $$\sigma_o^2 = \frac{R_b}{T_t} + \frac{R_b}{T_b} = R_b \frac{(T_t + T_b)}{T_t T_b}$$

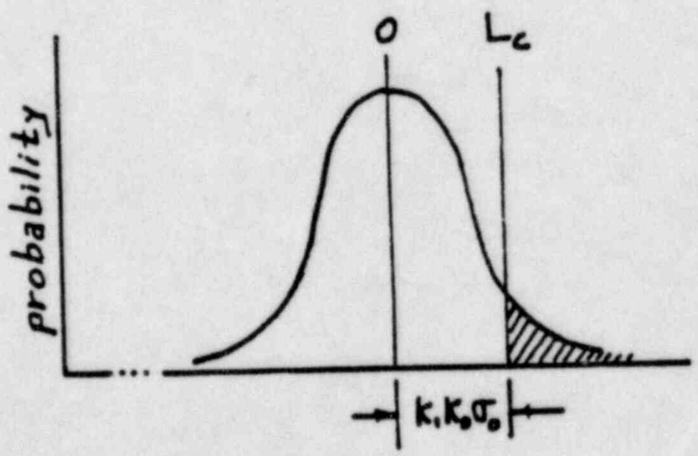

Figure 1. The Critical Level L_c.

$$L_c = K_0 K_1 \left[\frac{R_b}{T_b} \left(1 + \frac{T_b}{T_t}\right) \right]^{1/2} \qquad (1)$$

If sample and background counting times are equal, $T_b = T_t \equiv T$

$$\text{then } L_c = K_0 K_1 \left[\frac{2R_b}{T} \right]^{1/2} \qquad (2)$$

THE DETECTION LIMIT

The Detection Limit is defined as the smallest count rate which can be detected with a specified degree of confidence.

The Detection Limit could be taken to be equal to the critical level, L_c. See Figure 2. If this is done, there is one chance in two that true count rates equal to L_c will go undetected. It is not very satisfactory to specify a detection limit which can be detected only half the time; it is preferable to set the Detection Limit at some higher count rate such that the observed count rate will rarely be below the Critical Level, for example, not more than 5 percent of the time. The Detection Limit, as defined by equation (3), satisfies the requirement for the confidence level specified by K_1.

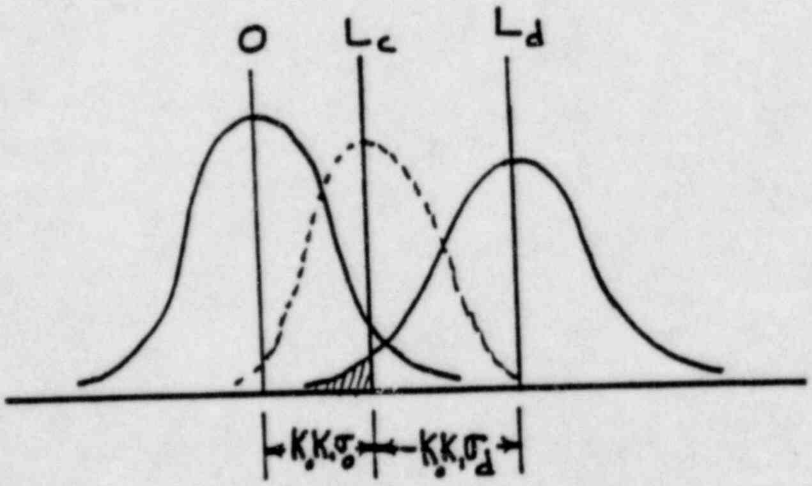

Figure 2. The Detection Limit L_d.

$$L_d = L_c + K_1 \sigma_d$$

Again, K_1 is the <u>one-sided</u> confidence factor. Since

$$L_d = R_d = R_t - R_b$$

$$\sigma_d^2 = \sigma_t^2 + \sigma_b^2$$

$$= \frac{R_t}{T_t} + \frac{R_b}{T_b} = \frac{R_d + R_b}{T_t} + \frac{R_b}{T_b} \; .$$

But $R_d = L_d$ so $$\sigma_d^2 = \frac{L_d}{T_t} + \frac{R_b}{T_b} \left(1 + \frac{T_b}{T_t}\right)$$

and

$$L_d = L_c + K_1 \left[\frac{L_d}{T_t} + \frac{R_b}{T_b} \left(1 + \frac{T_b}{T_t}\right) \right]^{1/2}$$

Solving for L_d and substituting the expression for L_c:

$$L_d = \frac{K_o^2 K_1^2}{T_t} + 2K_o K_1 \left[\frac{R_b}{T_b} \left(1 + \frac{T_b}{T_t}\right) \right]^{1/2} = \frac{K_o^2 K_1^2}{T_t} + 2L_c \qquad (3)$$

If the sample and background counting times are equal, $T_b = T_t \equiv T$, then

$$L_d = \frac{K_o^2 K_1^2}{T} + 2K_o K_1 \left[\frac{2R_b}{T} \right]^{1/2} \qquad (4)$$

In most cases where one is attempting to minimize the Detection Limit, counting times are long and the first term is negligible compared to the second:

$$\frac{K_o^2 K_1^2}{T} << 2K_o K_1 \left(\frac{2R_b}{T} \right)^{1/2}$$

The Detection Limit is then given by the following approximate formula often seen in the literature.

$$L_d = 2K_1 \left(\frac{2R_b}{T}\right)^{1/2} \quad . \qquad (5) \quad *$$

Choosing $K_1 = 1.65$ will result in only 5 percent of true count rates equal to L_d being missed (classified as background).

Again, a factor K_o has been inserted into equations (3) and (4) and will be discussed later.

THE LESS-THAN LEVEL

Suppose we have a sample with a count rate at or above the background count rate, but less than the Critical Level. We will conclude that we have not detected net radioactivity, but how large could the true count rate be and still produce a count rate not more than was observed? Neither L_c nor L_d answers this question. The Less-Than Level is defined as the maximum true net count rate which a sample could have (at a specified confidence level), based on a measured R_s, where R_s is less than L_c. The Less-Than Level is developed similarly to the Detection Limit, except with $R_s < L_c$. The Less-Than Level

$$L_\ell = R_s + K_1\sigma_\ell$$

$$\sigma_\ell^2 = \sigma_t^2 + \sigma_b^2$$

$$= \frac{R_t}{T_t} + \frac{R_b}{T_b}$$

$$= \frac{R_L + R_b}{T_t} + \frac{R_b}{T_b} = \frac{L_\ell}{T_t} + \frac{R_b}{T_b} \left(1 + \frac{T_b}{T_L}\right)$$

So, $\sigma_\ell^2 = \frac{L_\ell}{T_t} + \left(\frac{L_c}{K_o K_1}\right)^2$

Thus, $L_\ell = R_s + K_o K_1 \left[\frac{L_\ell}{T_t} + \frac{L_c^2}{K_o^2 K_1^2}\right]^{1/2}$

Solving for L_ℓ yields

$$L_\ell = R_s + \frac{K_o^2 K_1^2}{2T_t} + \left[\frac{R_s K_o^2 K_1^2}{T_t} + L_c^2 + \frac{K_o^4 K_1^4}{4T_t^2} \right]^{1/2} \tag{6}$$

If $R_s = L_c$, then $L_\ell = L_d$.

If $R_s = 0$, then $L_\ell > L_c$.

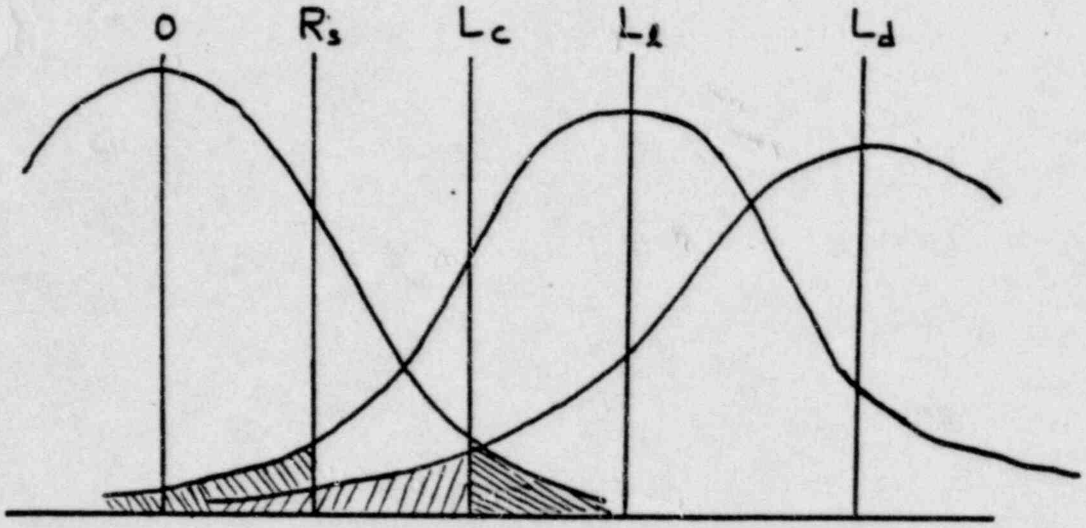

Figure 3. The Less-Than Level, L_ℓ
and its relationship to L_c and L_d.

THE DETERMINATION LIMIT

One last quantity of occasional importance is the <u>Determination</u> <u>Limit</u>, defined as the smallest net count rate which can be measured with a pre-specified relative standard deviation (i.e., coefficient of variation). Let

> f_q = the reciprocal relative standard deviation (e.g., if the coefficient of variation is to be 0.05, f_q = 20)

then the Determination Limit is given by

$$L_q = \frac{K_o f_q^2}{2T_t}\left[1 + \left(1 + \frac{4T_t R_b (T_t + T_b)}{f_q^2 T_b}\right)^{1/2}\right] \qquad (7)$$

When $T_t = T_b \equiv T$

$$L_q = \frac{K_o f_q^2}{2T}\left[1 + \left(1 + \frac{8R_b T}{f_q^2}\right)^{1/2}\right]$$

See the paper by Currie for the derivation.

REPORTING PRACTICES

The following reporting practices are recommended by Lochamy (3).

1. The Critical Level is used only to determine if a measurement is statistically different than background. It should not be used as a Detection Limit or Less-Than Level.

2. The Detection Limit and Determination Limit are not used for routine counting and reporting. In those cases where you are required to specify a minimum detectable activity (e.g., to a regulatory agency), it is recommended that the Detection Limit be given as the practical reporting limit.

3. The Determination Limit is useful when "sensitivity" with a specified relative standard deviation is required.

4. For routine low-level counting, only the Critical Level and Less-Than Level are of interest. Their use is:
   a. If $R_s > L_c$, the result is reported as positive, with the two-sided confidence interval desired, $R_s \pm K_2\sigma_s$, where for example $K_2 = 1.96$ at the 0.05 level, and
   $$\sigma_s = K_o\left[\frac{R_t}{T_t} + \frac{R_b}{T_b}\right]^{1/2} .$$
   b. If $R_s \leq L_c$, L_ℓ is calculated using the one-sided confidence interval and the result reported as less than L_ℓ.

THE CHI-SQUARE TEST

It has been assumed that the observed counts are distributed according to the Poisson distribution, although the inclusion of the factor K_0 permits application of the formulae if this is not the case. In real-life situations this is a hypothesis which must be tested. The appropriate test is a special form of the Chi-square test, tailored to the Poisson distribution (8). In general,

$$X = \sum_{i=1}^{n} \frac{(O_i - E_i)^2}{E_i}$$

where O_i and E_i are the observed and expected numbers, respectively, of occupants in each of the subdivisions in the distribution. For the Poisson distribution

$$X = \sum_{i=1}^{n} \frac{(x_i - \bar{x})^2}{\bar{x}}$$

$$= \frac{\sum_i x_i^2}{\bar{x}} - n\bar{x}$$

where x_i is one of <u>n</u> observed number of counts (<u>not</u> count rate), and $\bar{x}$ and s are the experimental mean and standard deviation of the x_i. Each of the counts x_i are taken for the same counting interval, i.e., preset time is used. After considerable disagreement on the subject, statisticians now seem agreed that the test statistic X is distributed according to the chi-square distribution with n-1 degress of freedom. The author's preference is to use an alternate test statistic,

$$\frac{X}{n-1}$$

and compare it to the chi-square over degrees of freedom distribution (9). The advantage of this latter statistic is that the expected value is 1, independent of degrees of freedom.

The procedure for conducting the test is as follows. Acquire twenty to fifty replicate counts x_i. The counting time and source activity should be similar to those employed when assaying unknowns. In the case under discussion, background samples are appropriate. To realistically simulate the background standard deviation, replicate background samples should be prepared and counted once each. The statistic $X/(n-1)$ is computed and compared to the chi-square over degrees of freedom distribution at the 95 or 98% confidence level. If the data pass this test, Poisson statistics may be assumed and $K_0 = 1$ in the formulae. If the chi-square test is failed, K_0 is calculated

$$K_0 = \frac{\text{observed standard deviation}}{\text{expected (Poisson) standard deviation}}$$

$$= \sqrt{\frac{X}{n-1}} \quad .$$

The author's experience is that many instruments will not regularly pass the chi-square test for background samples counted for long times. Any phenomenon which adds to the variability of the randomness of radioactive decay can cause failure of the test. Examples include instrument instability, diurnal variation in natural background, variation in background count rate due to movement of sources within the laboratory, variations between background samples, and variation of sample positioning.

OPTIMAL TIME PARTITION

Given an unknown sample and a background sample, what is the optimal way to partition a fixed, total counting time T between the background counting time T_b and the sample counting time T_t?

Consider the detection limit, for which

$$\sigma_d^2 = \frac{R_t}{T_t} + \frac{R_b}{T_b} \quad .$$

Since $T_t = T - T_b$, and T is constant, σ_d^2 is minimized when

$$\frac{\partial}{\partial T_b}\left(\sigma_d^2\right) = \frac{\partial}{\partial T_b}\left(\frac{R_t}{T-T_b} + \frac{R_b}{T_b}\right) = 0 \quad .$$

This yields

$$\frac{T_b}{T_t} = \sqrt{\frac{R_b}{R_t}} .$$

For very weak samples, $R_t \approx R_b$ so the time is divided most efficiently when $T_b = T_t$, that is, when sample and background counting times are equal. This discussion has presumed one unknown sample and one background sample. When there are many unknown samples to be assayed, more than one background sample may be employed (preset time), but since the detection limit is proportional to

$$(1 + \frac{1}{m})^{1/2}$$

where m is the number of background samples counted, there is little to be gained by using more than half-dozen background samples.

COUNTER SET UP

A final question should be addressed. It is readily apparent that both the counting efficiency and the background count rate depend upon the particulars of the instrument adjustments (high voltage, amplifier gain, window location and width, etc.). How should these adjustments be made so as to minimize the detection limit? In 1951 Lovenger and Berman found the elegant answer to this question. The detection limit is minimized when adjustments are made such that

$$\frac{R_s^2}{R_b} = \frac{(R_t - R_b)^2}{R_b}, \text{ or } \frac{(\text{efficiency})^2}{R_b}$$

is minimized. R_s^2/R_b is the proper quantity to be minimized when one is counting weak samples, $R_s << R_b$. When the count rates are not such that $R_s << R_b$, different quantities should be minimized or maximized to maximize counting efficiency.

This simple procedure is not without difficulties and cautions, which are:

1. The efficiency must be measured at various instrument settings, which is not difficult, because an active source may be used. The background count rate must also be accurately measured at these same instrument settings, which can be very time-consuming if the background count rate is low.

2. The background must be fixed and stable during the time-consuming measurements of the preceding paragraphs, and the background during sample counting should not be much different than this value.

3. Instrument settings for which the chi-square test is failed should be avoided in favor of settings for which the test is passed. That is, regions of instrument instability should be avoided.

4. The Lovenger-Berman procedure does not address certain problems. For example, it cannot be used with mixtures of isotopes of variable composition, as the efficiency is not a fixed number. The procedure cannot be used to optimize instrument settings when it is intended to most accurately differentiate between two radionuclides in a composite sample. This latter problem must be solved using mathematical considerations quite different than those described in this paper.

ACKNOWLEDGEMENT

This paper is primarily tutorial in nature. The author has drawn freely from material given by the references.

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CONVERSATION RECORD

TIME 2:30

DATE 8/26/85

TYPE

☐ VISIT ☐ CONFERENCE ☒ TELEPHONE

☐ INCOMING

☒ OUTGOING

Location of Visit/Conference:

ROUTING	
NAME/SYMBOL	INT

NAME OF PERSON(S) CONTACTED OR IN CONTACT WITH YOU	ORGANIZATION (Office, dept., bureau, etc.)	TELEPHONE NO.
Bruce Austin	Bradley-Thorpe	(513) 226-7824

SUBJECT

New License Application
Control No. 79420

SUMMARY

I requested the following additional info:

1) Description of Heard's previous experience working with magnesium-thorium alloys.

2) Clarification on who will train persons working under the supervision of Heard and the approximate length of the training.

3) Statement that bioassays, if warranted, will be performed by the method most appropriate to determine internal deposition – urinalysis, whole body

ACTION REQUIRED

NAME OF PERSON DOCUMENTING CONVERSATION	SIGNATURE	DATE
	B J Holt	8/26/85

ACTION TAKEN

SIGNATURE	TITLE	DATE

country, etc.

4) Complete protocol for collecting and analyzing air samples.

5) Instrument Calibration Procedures.

6) Re: Waste Disposal Procedures:

I informed Bruce that if G.E. is not authorized to receive waste material for disposal ~~will~~ we will not authorize Bradley-Thompson to send the mag-thorium shavings, etc. to them.

7.) Amount of time Medical Radiation Consultants will spend at the site of Bradley-Thompson Tool Company.