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NOTES ON CONFERENCE ON REACTOR POISONS. CONFERENCE HELD AT WAPD JANUARY 3, 1952

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Atomic Power Division
Westinghouse Electric Corporation
Pittsburgh, Pennsylvania

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INTRODUCTION

About a year ago various members of the WAPD staff became disturbed by informal reports that the fission yield of xenon-135 had been incorrectly measured and was, in fact, at least 10% higher than had been assumed. If true, these reports had a considerable significance insofar as the reactor program was concerned. In an effort to get the facts clarified, a meeting of radiochemists was proposed to be held as a joint Canadian-American conference. After many delays, due in large part to the difficulty of arranging a joint Canadian-American meeting, the conference was held in New York on September 10, 1951.

At this conference no definite conclusions could be drawn from the available data. It was, however, quite clear that further radiochemical studies were necessary. At the same time, it was suggested that attacking the problem from the reactor physics point of view, utilizing data from operating reactors, might be significant. The conference on reactor poisoning held at WAPD on January 3, 1952, was the outgrowth of this suggestion.

The attached notes are intended to summarize the information discussed at this latter conference. Except for the Summary of Radiochemical Data by R. A. Brightsen, none of the participants mentioned has had an opportunity to edit these notes and should not be held responsible for any errors or misstatements in the notes. The general result of the conference shows that a set of parameters can be developed for a given reactor which explains the poison behavior of that reactor. It is not easy, however, to reconcile the results obtained from different reactors. Because of the inconclusive nature of the results, it was not considered worthwhile to issue a formal set of minutes; instead these notes are being issued to indicate the general nature of the discussion.

The WAPD staff is deeply indebted to the conference participants for their time and effort in assisting us with this particular problem; we are especially grateful to Prof. C. D. Coryell for his sponsorship of the radiochemical conference and for his continuing interest. We hope that the studies projected at these conferences will ultimately lead to a complete understanding of the reactor poison problem.

For the benefit of the participants at the conferences, notes on both are being distributed together.

S. Krasik

NOTES ON CONFERENCE ON REACTOR POISONS

Conference held at WAPD
January 3, 1952

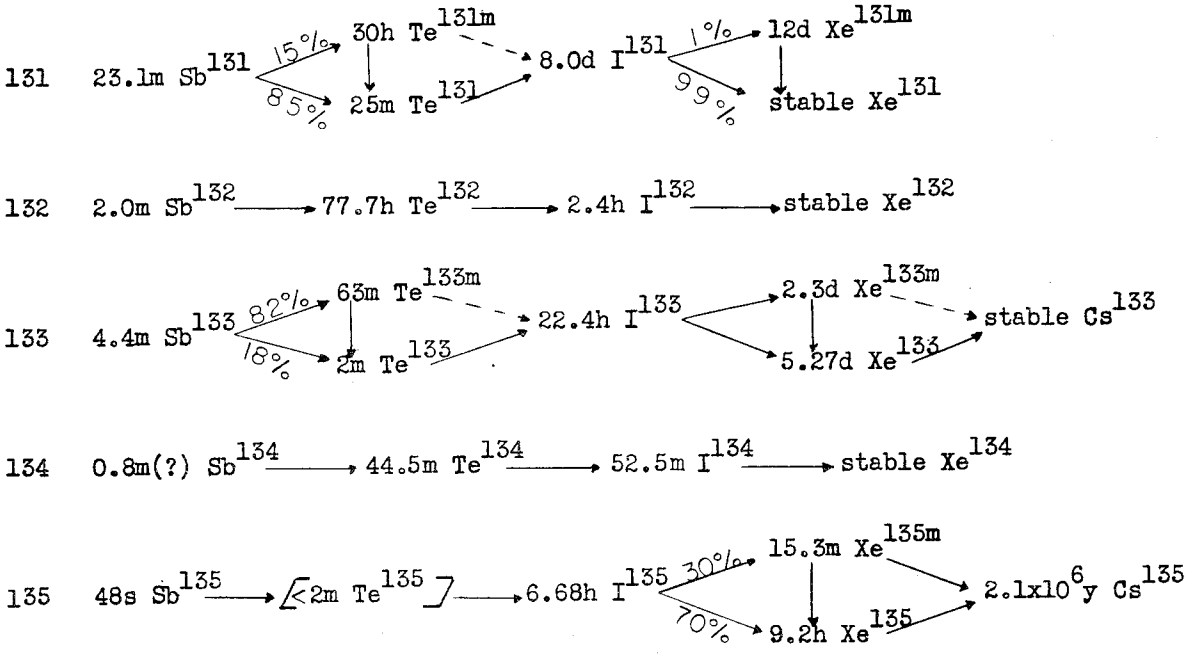
Notes by R. R. Schiff

SUMMARY OF RADIOCHEMICAL DATA (R. A. Brightsen, WAPD)

Introduction

The information presented at the Reactor Poison Conference of September 10, 1951, on the fission yield of Xe-135 has been summarized in part in WAPD-RM-95; information not available at the time WAPD-RM-95 was written is outlined in brief form here.

The mass chains of interest are:



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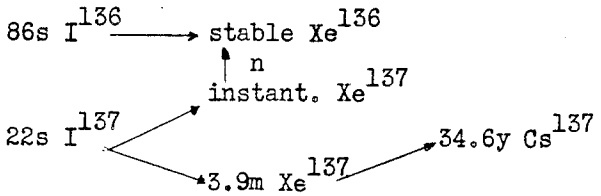


Summary of Radiochemical Measurements on Chain 135

(1) No information is available on the fission yields of the 48-second Sb-135 and the hypothetical < 2m Te-135.

(2) Early Project measurements on the fission yield of the 6.68h I-135 by Glendenin and Metcalf and by Katcoff and co-workers gave values of 5.6 ± 0.3% for both sets of experiments. These values were obtained using absorption correction factors corresponding to I-135 β half-thicknesses of 45 mg/cm^2 and 47 mg/cm^2, respectively. These workers had found by absorption measurements only a single β particle in the 6.68h I-135 beta spectrum, with an energy of 1.4 mev. Since that time, beta-ray spectrometer measurements have shown that the β-spectrum of I-135 is complex; 0.47 mev (35%), 1.00 mev (40%) and 1.40 mev (25%). Hence most betas have a lower energy than the 1.4 mev used by early workers; they therefore should have used a higher correction factor, leading to a higher fission yield. Pappas has re-evaluated the original data and obtains a new value of 6.10%. He has also made two new measurements, obtaining values of 6.10 and 6.20% based on 6.17% for 12.80d Ba-140.

(3) Hoagland and Sugarman in five experiments found a value of 4.1% of the total chain yield of 6.68h I-135 for the fraction of 9.2h Xe-135 and 15.3m Xe-135m formed promptly in fission, but considered a value of 4.9 ± 0.6% the best value for the independent yield of these two species. Taking the I-135 yield as 6.10%, the independent yield of the two Xe-135 species is 0.30% [= (.049)(6.10)]; hence the available radiochemical data indicate a fission yield of 6.40% (= 6.10 + 0.30) for the 9.2h Xe-135.

Fission Yield of 9.2h Xe-135 from Mass Spectrometer Measurements

Survey of method and recent developments

The yield of the 9.2h Xe-135 is obtained in the following manner:

$$\text{(I)} \qquad Y\,(Xe^{133}) = \left[\frac{Xe^{133}}{Xe^{131}}\right]\left[Y\,(I^{131})\right]$$

$$\text{(II)} \qquad Y\,(Xe^{135}) = \left[\frac{Cs^{135}}{Cs^{133}}\right]\left[Y\,(Xe^{133})\right]$$

where

$$\left[\frac{Xe^{133}}{Xe^{131}}\right] \quad \text{and} \quad \left[\frac{Cs^{135}}{Cs^{133}}\right]$$

are ratios obtained from mass spectrometric analysis of fission xenon and and cesium and $Y\,(I^{131})$ is the absolute fission yield of 8.0d I-131 obtained radiochemically and based on a 6.17% yield for 12.80d Ba-140.

The following points should be noted:

(a) Implicit in equations (I) and (II) is the assumption that the independent yields of Xe-131 and Cs-133 are negligible; this assumption is very probably a good one. However, any appreciable independent yields for Xe-131 and Cs-133 would increase the value of the Xe-135 yield.

(b) The ratio of (Cs^{135}/Cs^{133}) is discussed in detail in WAPD-RM-95. This ratio is rather well known from Thode's recent work, and probably is the most accurately known of the three parameters necessary to obtain a value for the absolute yield of 9.2h Xe-135. The value obtained by Thode is 1.057/1.096 = 0.9644.

(c) The ratio of (Xe^{133}/Xe^{131}) obtained by Thode is complicated by the recent discovery of 2.30d Xe-133m. Thode corrected his observed ratio for the

decay of Xe-133, using a value of 5.27d for the Xe-133. This may not be the correct value of the half-life; on the other hand, the 5.27 day value measured by Thode for Xe-133 was obtained on a sample containing some 2.30 d Xe-133m. Hence the error may be negligible. The value of (Xe^{133}/Xe^{131}) obtained by Thode is 6.30/2.80 = 2.250.

(d) The fission yield of 8.0d I-131, discussed in WAPD-RM-95, has recently been re-measured by Yaffee at Chalk River, counting in a 4π methane flow counter. According to Yaffee (private communication, December 7, 1951) the counting errors were negligible. The values he obtained are, relative to 6.17% for 12.80d Ba-140:

	3.17
	3.21
	3.16
average	3.18

This is to be compared to the Pappas value of 3.0 and the old Project value of 2.8 obtained by Katcoff and co-workers.

The absolute fission yield of 9.2h Xe-135

Taking the average value of 3.09 ± 0.09 [i.e., weighting equally the measurements of Pappas (3.00) and Yaffee (3.18)] as the best available figures for the fission yield of 8.0d I-131, one can compute the fission yield of 9.2h Xe-135 to be

$$Y\,(Xe^{135}) = \left[\frac{Xe^{133}}{Xe^{131}}\right]\left[\frac{Cs^{135}}{Cs^{133}}\right]\left[Y\,(I^{131})\right]$$

$$Y\,(Xe^{135}) = (2.250)\,(0.9664)\,(3.09\pm0.09) = 6.70 \pm 0.20\%$$

The radiochemical value of 6.40% is certainly not better than ± 5%; hence one can estimate 6.4 ± 0.3% from this type of experiment. The available data can thus be summarized as follows:

Type of Measurement	Absolute Yield of 9.2h Xe^{135}
Mass spectrometry and radiochemistry	6.7 ± 0.2%
Radiochemistry	6.4 ± 0.3%

There seems to be no way at present to make a definite choice between these two values. It is the author's opinion, however, that until data are available on the absolute fission yield of Cs-133, the value of 6.7 ± 0.2 ought to be used, in view of the importance of the Xe-135 override in power reactors.

PROPOSED CRYSTAL SPECTROMETER MEASUREMENT OF XENON CROSS SECTION (J. Faulkner, HW)

It is planned to perform a crystal spectrometer measurement on the Xe-135 cross section up to 1 ev using the Hanford production pile. Close attention will be paid to the xenon assay. Gases will continuously be swept out by helium from a specially prepared uranium hydroxide precipitate in the

reactor and be passed through charcoal columns to separate the xenon from other gases, mainly krypton. It is hoped to get at least 10^{17} atoms of xenon in this manner. It is planned to determine the number of atoms by absolute γ-β counting on the xenon itself. The experiment is now being set up on a pilot basis. Possibly a year will suffice for completing the experiment.

DATA FROM BROOKHAVEN PILE OPERATION (I. Kaplan, BNL)

An experiment at Brookhaven measured the excess reactivity required to compensate for equilibrium 9h Xe-135. The formula

$$\delta k = -\frac{f}{k} Y \frac{\sigma_f}{\sigma_a^u} \frac{\int \frac{\varphi^3}{\lambda_x/\sigma_x + \varphi} dV}{\int \varphi^2 dV}$$

can be derived for low burnup from the one group perturbation theory. In this expression σ_a^u is the microscopic absorption cross section of uranium, V is the volume of the reactor, and the other quantities are defined as in the previous section. Borst evaluated $Y\sigma_x$ from this expression and found $Y\sigma_x$ = 16 to 19x10^4 b. Using 5.9% for Y, this gives σ_x = 2.7 to 3.2x10^6 b.

The Canadians integrated the same expression and found that it could be replaced by

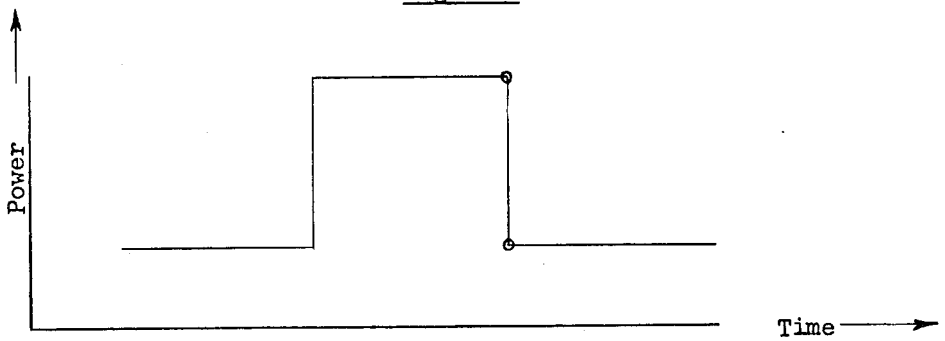
$$\delta k = -\frac{f}{k} Y \frac{\sigma_f}{\sigma_a^u} \frac{1}{1 + \text{const.} \left(\frac{P}{U}\right)^* \frac{U}{P}}$$

where P is the power, U the mass of uranium, and the starred ratio refers to the power for which there is equal probability of decay or burnout.

The constant depends on the geometry and on σ_x. When a value of 2.9×10^6 b is taken for the pseudo-average cross section of xenon, δk is 65 to 70 inhours. This compares with an experimental value of about 75 inhours.

At the time, it was thought that the Xe-135 cross section was quite well determined, and more interest was taken in establishing the xenon decay scheme. To this extent "flash experiments" were performed, i.e., the reactor was run at a low power level for a considerable period, then suddenly "flashed" to a much higher level for several hours and finally taken down to the low power level again. (See Figure 2 below.) If some 9h Xe-135 were formed from a 15-20 minute precursor, the effect of this should be noted just after the pile had returned to low level operation. However, in order that such an effect be observable, at least 10% of the 9h Xe-135 would have to be formed in this manner. No effect was found, thus placing an upper limit of 10% on the 9h Xe-135 which might be formed from a short-lived precursor.

Figure 2



Furthermore, if as much as 10% of the Xe-135 had been formed as a direct fission product, that too would have been observed. Negative results here also indicate an upper limit of 10% on the direct fission yield. Chalk River experiments reduce this upper limit to 2%.

It is no longer practical to do more xenon work on the BNL reactor. Furthermore, the effects of xenon on the reactivity were never large in this reactor, and complete xenon override was always achieved. The problem of temperature distribution presents another serious obstacle since the pile is coolest at the center. Temperature effects are a large part of the total xenon effect. It appears that large graphite piles are simply not well suited to the measurement of xenon data.

USE OF THE MTR AT ARCO

The MTR will be available within a few months and the ORNL group there is eager to carry out experiments when it first is brought to power. The MTR has advantages for getting fundamental data over such reactors as STR because it is flexible, fairly regular in shape, and without large holes.

It was suggested that the MTR might be used for getting complete xenon data. Certainly the question of the direct yield and the yield from a short-lived precursor could be settled.

Consideration was given to methods of calibrating the reactor, such as changing the size of the core, moving the rods, and introducing uniform poison. The possibility of avoiding the necessity for calibration by allowing the reactor to run away and regain criticality because of temperature effects was mentioned, but the feasibility of this method was questioned.

It was decided that ANL and WAPD should jointly make arrangements with the MTR group concerning possible experimental work at Arco.

PROPOSED CHALK RIVER CONFERENCE

Dr. Lewis of NRC has suggested a conference at Chalk River on reactor poisons and related problems such as long term reactivity changes. It was felt that such a meeting might be of more interest to groups outside the Naval Reactor Division. Therefore, the matter was referred to the Reactor Development Division of the AEC.