THE $^{252}$Cf $\bar{\nu}$ DISCREPANCY AND THE SULFUR DISCREPANCY

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The cantankerous discrepancy among measured values of $\bar{\nu}$ for $^{252}$Cf appears at last to be nearing a final resolution. A recent review has summarized the progress that has been achieved through revaluation upward by 0.5% of two manganese bath values ν and the performance of a new liquid scintillator measurement. A new manganese bath measurement at INEL is in reasonably good agreement with previous manganese bath values of $^{252}$Cf $\bar{\nu}$. It now appears that the manganese bath values could still be systematically low by as much as 0.4% because the BNL-325 thermal absorption cross section for sulfur may be as much as 10% low. There is a bona fide discrepancy between measurements of the sulfur cross section by pile oscillators and the values derived from transmission measurements. The resolution of this discrepancy is a prerequisite to the final resolution of the $^{252}$Cf $\bar{\nu}$ discrepancy.

[$^{252}$Cf $\bar{\nu}$, manganese bath, pile oscillator, sulfur, absorption cross section, small-angle scattering]

## Introduction

The discrepancy among measured values of $\bar{\nu}$, the total number of neutrons produced per fission event, for $^{252}$Cf, has long perplexed both the measurers and the users of this important nuclear datum. In recent years considerable progress has been achieved in understanding this important quantity, which is the standard against which $\bar{\nu}$ values for the fissile nuclei are compared.

The status of measurements of $\bar{\nu}$ for $^{252}$Cf was reviewed by the present author at the Symposium on Nuclear Data Problems for Thermal Reactor Applications in May, 1978.[1] A summary table representing the current values for the various experimental results is reproduced in Table I. Two significant developments are represented in the current values shown in Table I: The renormalization upward by approximately 0.5% of the manganese bath values of Axton[2] and Devolpi[3] and the addition of a preliminary result from a new liquid scintillator measurement of $\bar{\nu}_p$ by Spencer. The Spencer value may be revised in Spencer's paper prepared for this meeting.

A striking feature of Table I is the remarkable agreement among the manganese bath values, with the exception of the White-Axton value.[10] The latter measurement has been assigned a relatively large error by its authors, so it carries only a modest weight into a weighted average. An impressive feature of the general manganese bath agreement is the variety of fission assay techniques represented. Axton[2] used a unique fission-fission coincidence technique, DeVolpi[3] used neutron-fission coincidence counting, and Bozorgmanesh[9] and Aleksandrov[11] both used defined solid angle counting. The agreement between the latter two measurements would appear to accentuate the isolation of the White-Axton value.

The close general agreement of the manganese bath results, clustered as they are about 0.5% lower than the weighted average of all measurements, suggests that if this average is near the true $^{252}$Cf value, there may be a systematic bias of 0.5% somewhere in the manganese bath method itself. It is therefore appropriate to review the manganese bath method to ascertain where such a systematic problem might lie. Some characteristics might be postulated. The effect should be capable of producing the bulk of the 0.5% correction that is sought. It should preferably raise the value of $\bar{\nu}$ without simultaneously raising the values of $n_p$, since these are already in agreement with the higher $\bar{\nu}$ values. Finally, one should not overlook the possibility that the effect lies in something that has been done the same way for so long that we have

come to confuse familiarity with accuracy.

## TABLE I
### $^{252}$Cf $\bar{\nu}$ Summary

| MEASUREMENT | $\bar{\nu}$ (Total) |
|---|---|
| **Liquid Scintillator** | |
| Spencer[4] | 3.792+0.011 |
| Boldeman[6] | 3.755+0.016 |
| Asplund-Nilsson[7] | 3.792+0.040 |
| Hopkins-Diven[8] | 3.777+0.031 |
| **Manganese Bath** | |
| Axton[2] | 3.743+0.019 |
| DeVolpi[3] | 3.747+0.019 |
| Bozorgmanesh[9] | 3.744+0.023 |
| White, Axton[10] | 3.815+0.040 |
| Aleksandrov[11] | 3.747+0.036 |
| **Boron Pile** | |
| Colvin[12] | 3.739+0.021 |
| Wtd. Ave: | 3.766+0.007 |
| Ave: | 3.765+0.008 |

## The Absorption Cross Section of Sulfur

The sulfur thermal absorption cross section has rather abruptly emerged as perhaps the prime candidate for the dubious distinction of contributing the principal part of a half percent systematic error. This possibility came to light in a review of the sulfur file submitted by Howerton for inclusion in ENDF/B-V. The evaluation is a very old one, with little documentation.[13] The most unsettling part of this review was to find that the thermal capture had been normalized to 0.49 barn at 0.025 eV. For some 20 years the accepted value has been the BNL-325 value[14] of 0.52 barns, with the error variously assigned as 0.02 or 0.03 barn. The difference between 0.49 and 0.52 barn may seem inconsequential to some, but it represents about a quarter percent difference in a manganese bath measurement. An inquiry to Howerton revealed that he has no record of how he arrived at this thermal value. It is presumed that he followed his often-declared preference for selecting

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cross sections from the single experiment he judged to be best, rather than resorting to a weighted average. The implied disdain for weighted averages is a fairly common and well-founded attitude. Indeed, it is the use of weighted averages, and an attendant underestimate of the error, that has served to create an unwarranted attitude of contentment towards the 0.52-barn value. Similar problems have plagued the $^{252}$Cf $\bar{\nu}$ system, also. The weighted average could well be called the opiate of the evaluator. It is useful as a guide, but should not be used as a crutch.

The cross section chosen by Howerton as best was evidently the Pomerance value,[15] normalized to the current value of the cross section of gold, which was the standard for the measurement. A reexamination of possible reasons for favoring the Pomerance measurement led to the compilation of the comparison shown in Table II. This is a collection of sulfur absorption values, from three sets of pile oscillator measurements,[15-17] that would be derived if the sulfur result were renormalized to current values of cross sections for other materials measured in the same experiment. This table is intended as a test to see which series of measurements has the best internal consistency, and has stood the test of time best, as viewed from the point of view of the sulfur cross section. Included in the comparison are the cross section standards boron, lithium, and gold, plus some low cross section materials. Manganese has been included because it is the ratio to the manganese cross section that is important in evaluating manganese bath corrections.

TABLE II
Sulfur $\sigma_a$

| Normalization | Pomerance | Harris | Colmer |
|---|---|---|---|
| Original Value | 470 mb | 510 mb | 490 mb |
| B = 759 b | 489 | 545 | 532 |
| Li = 70.7 b | 496 | --- | 529 |
| Au = 98.8 b. | 489 | --- | --- |
| Mn = 13.35 b | 490 | 554 | 511 |
| Co = 37.2 b | 511 | 531 | 477 |
| K = 2.10 b | 481 | 508 | 544 |
| Ca = .43 b | 493 | 510 | 527 |
| Average | 493 mb | 530 mb | 520 mb |
| Std Dev | 9 | 21 | 24 |

*Harwell B = 771 b

The Pomerance set of measurements does appear to have a decided advantage in terms of internal consistency, and this might be considered support for the choice of the Pomerance values. A second glance, however, reveals that the internal consistency of each set is adequate to suggest that the difference in sulfur values may not be a matter of random measurement error alone. One is led to suspect the presence of extraneous effects in the sulfur samples themselves. It is well known that scattering from the sample complicates pile oscillator measurements. All three pile oscillator groups were aware of the scattering problem, and all three dealt with it in their experiments in one way or another. Since there does appear to be some sample-dependent differences, however, one is led to wonder whether the particulate nature of the powdered sulfur samples may have intro-

duced types of scattering that were not typified by the method of correction.

The sort of thing that might be suspected is illustrated by some studies of the $^{242}$Pu total cross section by Young.[13] Fig. 1 shows the original, uncorrected data, the same data corrected for moisture content of the oxide sample, and further correction for small-angle scattering from the particles of which the oxide was composed. Measurements on the metallic sample, which were made later, were found to be in good agreement with the corrected data from the oxide sample. Also evident is the structure that is often observed in the vicinity of the cutoff of Bragg scattering from sets of crystalline planes of the sample. The Bragg scattering is a matter of sample composition, and can be seen to be different for the two samples. Both the small-angle scattering and the Bragg cutoff phenomenon can be expected to produce scattering that is both energy-dependent and non-isotropic. Whether these effects would disturb a pile oscillator measurement is not clear, but the comparison in Table II suggests that some sample-dependent complication is present.
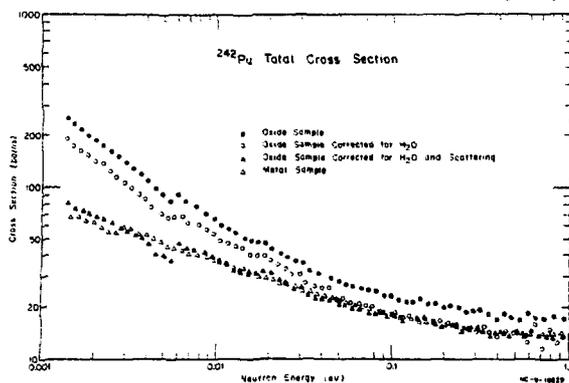


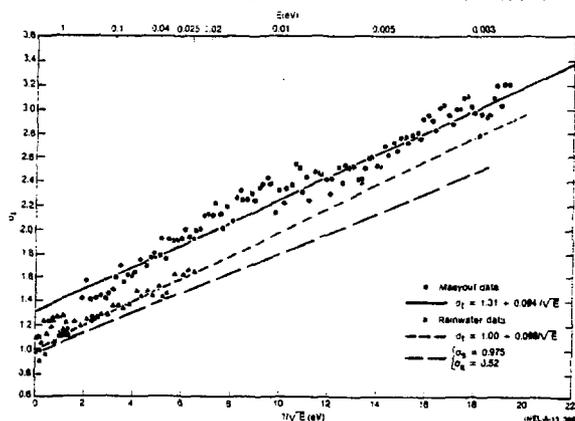Fig. 1  Neutron total cross section of $^{242}$Pu from 0.0015 to 1.0 eV.[18]



Fig. 2  The total cross section of natural sulfur. Measurements of Rainwater and Maayouf[20] are compared with the behavior indicated by the constants evaluated in BNL 325.[14]

The indication of possible sample-dependent effects in the pile oscillator measurements leads naturally to the question of whether the transmission measurements have been fairly treated. When there are no nearby resonances, the total cross section can be fitted to a simple curve representing the sum of a constant cross section and a 1/V term. The constant term is identified as the scattering cross section, while the 1/V term represents the absorption. There are two published values for the sulfur absorption cross section determined by this method. Both are shown in Fig. 2. Rainwater[19] derived a value of 0.62 barn from his measurements on a powdered sample of sulfur. There is some apparent structure near 0.04 eV, which could be due to a Bragg cutoff that would complicate the interpretation of the data. Maayouf[22] deduced an absorption cross section of 0.59 b. from his measurements on a cast sulfur sample. His data also show structure, but the double-humped nature of the structure suggests that it is an artifact of the rotor transmission, rather than Bragg structure. Maayouf used two different settings of his velocity selector rotor to cover the energy region of the measurement, and the dividing line between the two settings was between the two humps. Although there is considerable separation between the Rainwater and Maayouf data, the slopes are in reasonable agreement, considering the scatter in the data points. Neither measurement appears compatible with the 0.52-barn cross section, which is represented by the lower curve.

A third transmission measurement was made by the present author on the MTR cystal spectrometer, with a velocity selector to eliminate neutrons from higher order reflections in the Bragg beam. The data from the measurement appeared to be very linear on the $1/\sqrt{E}$ plot, with none of the structure evident in the Rainwater and Maayouf data. A fit to the data represented a thermal absorption cross section of . 0.57 barn, which is somewhat below the values from the other transmission measurements, but disturbingly above the values from the pile oscillator experiments. The lowest value with which the data could be deemed reasonably compatible was 0.55 b. The high value was tentatively ascribed to uncertainties in the characterization of the cast sulfur block, which was then ten years old. The data were not published, and were subsequently destroyed.

All three total cross section measurements have shown evidence of experimental difficulties that justify their being down-weighted in an evaluation. However, they agree in two significant points: they all yield sulfur capture cross section values well above the results of the pile oscillator measurements, and a curve representing a cross section of 0.52 b. does not appear to be compatible with any of the energy-dependent data. The transmission measurements do indeed have severe experimental problems that are clearly visible. The data comparison of Table II, however, suggests that the pile oscillator measurements may have problems that are no less severe; they may be simply less visible.

An inquiry to Dr. Pomerance, as to whether the intervening 30 years had brought any reason to modify the treatment of his sulfur data, brought a negative reply.[21] However, the reply included some details of his experiment that are very interesting, in the light of the preceding discussion. He supplied the results of the measurements on the individual samples, which included two powdered sulfur samples and two samples of sulfur salts. These values are shown in Table III, both as originally normalized to a gold cross section of 95 barns and as renormalized to the current gold value of 98.8 barns.[14] The salt samples yielded sulfur

cross sections near or above the results from the transmission measurements. Pomerance felt that the most likely error would come from the presence of impuritiies in the low cross section samples. Therefore it was the high values that were considered suspect and discarded. An alternative explanation might be that the differences among Pomerance's samples supports the above-postulated existence of sample-dependent scattering differences.

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TABLE III

POMERANCE SULFUR MEASUREMENTS

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| SAMPLE (wt) | ORIGINAL $\sigma_a$ (barns) | | RENORMALIZED $\sigma_a$ (barns) | |
|---|---|---|---|---|
| | Molecule | Sulfur | Molecule | Sulfur |
| Sulfur #1 (3.139 g) | 0.48 | 0.48 | 0.50 | 0.50 |
| | 0.475 | 0.475 | 0.494 | 0.494 |
| Sulfur #2 (1.936 g) | 0.51 | 0.51 | 0.53 | 0.53 |
| | 0.40 | 0.40 | 0.42 | 0.42 |
| PbSO$_4$ (5.327 g) | 0.76 | 0.57 | 0.79 | 0.62 |
| Na$_2$S$_2$O$_5$ (2.218 g) | 2.24 | 0.74 | 2.52 | 0.73 |

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AVERAGE VALUES

| All Sulfur | .47 | .49 |
|---|---|---|
| First 3 Sulfur | .49 | .51 |
| All Measurements | .53 | .55 |

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*CROSS SECTIONS ASSUMED

| | Au | Pb | Na |
|---|---|---|---|
| Original | 95 | 0.19 | 0.47 |
| Renormalized | 98.8 | 0.17 | 0.53 |

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Another curious factor in the Pomerance data set is the fact that the second measurement on the second sulfur sample was about 20% below the first measurement. This puts it nearly as far out of line on the low side as the salt samples are on the high side. Whether the presumption is that something happened to the sample between measurements or that the difference reflects the true reproducibility of the measurements, one wonders whether the high and the low values ought to be accepted or rejected on an equal basis. Rejection of the low measurement would leave three values whose average is 0.51 barn. This would have the rather unsettling consequence that fully 50% of the Pomerance sulfur data would thereby have been rejected. Inclusion of all data would bring the Pomerance average to 0.55 barn.

The puzzle posed by the Pomerance data is the same as that posed by the attempt to reconcile the pile oscillator and transmission measurements. Is it valid to reject the higher values because they "look" too high? Not enough information is available concerning the various samples to allow solution of the problem by evaluative measures alone. New and better measurements are required.

## Effects of Sulfur Absorption in the Manganese Bath

To show why a wretched half-barn cross section should be so important to manganese bath measurements and to the $^{252}$Cf $\bar{\nu}$ situation, it is necessary to examine briefly the equation describing the response of a manganese bath. When a source of strength Q is irradiated, the observed saturated activity $A_s$ is given by

$$A_s = Q \, f \, \varepsilon \, (1-S)(1-L)(1-P), \qquad (1)$$

where $\varepsilon$ represents the efficiency of the counter for detection of a $^{56}$Mn disintegration anywhere in the system, S is the loss to neutron absorption in structural material, L is the fraction leaking from the bath, and P is the loss due to (n,p) and (n,$\alpha$) reactions in oxygen and sulfur. The effect of the sulfur thermal absorption cross section is contained in f, the fraction of neutrons absorbed in manganese in the bath, where f is given by

$$\frac{1}{f} = 1 + \frac{\sigma_S}{\sigma_{Mn}\,(1+Grs)} + \frac{N_H}{M_{Mn}} \, \frac{\sigma_H}{\sigma_{Mn}\,(1+Grs)} . \qquad (2)$$

An uncertainty in the sulfur cross section has an impact not only upon the ratio $\sigma_S/\sigma_{Mn}$, but also upon the ratio $\sigma_H/\sigma_{Mn}$. This follows from the way the latter ratio is determined. To obtain better manganese bath accuracy than can be obtained through the use of individual hydrogen and manganese cross sections, their ratio is determined by making bath activation measurements as a function of $MnSO_4$ concentration. By rearranging equations (1) and (3), we obtain an equation of a straight line in which the independent variable is the modified concentration parameter $N_H/N_{Mn}(1+Grs)$:

$$\frac{(1-S)(1-L)(1-P)}{A_s} =$$

$$\frac{1}{Q} \, \frac{\sigma_H}{\sigma_{Mn}} \, \frac{N_H}{N_{Mn}\,(1+Grs)} + \frac{1}{Q}\left[1 + \frac{\sigma_S}{\sigma_{Mn}\,(\overline{1+Grs})}\right]. \qquad (3)$$

The slope, m, is divided by the intercept, b, of the fitted line, yielding

$$\frac{m}{b} = \frac{\sigma_H/\sigma_{Mn}}{1 + \sigma_S/\sigma_{Mn}\,(\overline{1+Grs})} \approx R_H , \qquad (4)$$

where $R_H$ is introduced to represent the observed ratio, before correction for the sulfur absorption. The hydrogen-to-manganese cross-section ratio is then simply

$$\sigma_H/\sigma_{Mn} \approx R_H \, [1 + \sigma_S/\sigma_{Mn} \, (\overline{1+Grs})] ,$$

The cross-section ratio thus derived has improved precision as compared to that derived from individual measurements on manganese and hydrogen, but both its value and its error estimate are correlated with those of the sulfur cross section. To avoid overlooking this important point, it may be instructive to insert the factor $R_H$ directly into Eq. (3), which may then be written

$$Q = \frac{A_s}{(1-S)(1-L)(1-P)}$$

$$X \left[1 + \frac{\sigma_S}{\sigma_{Mn}\,(\overline{1+Grs})}\right]\left[1 + \frac{N_H R_H}{N_{Mn}\,(1+G\bar{r}s)}\right] . \qquad (5)$$

This form of the equation separates the sulfur effect from the other corrections, and makes it clear that the sulfur correction is independent of concentration. The current BNL-325 value of $0.52 \pm 0.03$ barns represents a correction of nearly 4%, and implies an uncertainty of about 0.22% in the derived source strength. That is close to the overall error that is hoped for in the whole manganese bath measurement, but it represents the error from only one correction in one leg of the $\bar{\nu}$ measurement. If the sulfur cross section should really turn out to be 0.57 b or above, as suggested by the transmission measurements and the data from the Pomerance sulfur salt samples, the corresponding shift of $\bar{\nu}$ values from the manganese bath could approach half a per cent.

The sulfur cross section thus has the potential for representing an error of as much as half a per cent in measurements of neutron source strenth by the manganese bath method. A rise in the sulfur absorption cross section would raise manganese bath $\bar{\nu}$ values without imposing a corresponding rise in the n values, since these are determined from ratio measurements only. Finally, the cross section measurements date back 30 years. The acceptability of the 0.52 b value may be more a matter of familiarity than of accuracy. Thus the sulfur absorption cross section satisfies all of the criteria earlier postulated as characteristics of a possible source of a half per cent systematic effect in manganese bath measurements.

### Possible Impact on $^{252}$Cf $\bar{\nu}$

Further complicating the picture is the fact that the $^{252}$Cf $\bar{\nu}$ measurement by the present author, in its current state of analysis, appears to be headed for the vicinity of 3.77, roughly half a per cent above the other manganese bath values. This is with the traditional value of 0.52 b for sulfur used in the analysis. Since the analysis is still incomplete, it is perhaps premature to conclude that a half per cent systematic effect other than the sulfur absorption cross section has been overcome. In any event, it is essential to the final resolution of the $^{252}$Cf $\bar{\nu}$ discrepancy that the current ambiguity in the sulfur absorption cross section be resolved.

As an illustration of the effect of the sulfur cross section, let us suppose that the true value should be found to be 0.57 b. Then the array of $\bar{\nu}$ measurements would take on the aspect shown in Table IV, in which it is assumed that the present author's measurement should continue at its present preliminary value. In the weighted average, it is presumed that the sulfur cross section would be established with sufficient accuracy that the errors assigned in Table I would be maintained, and that the INEL measurement will prove to be at least as accurate as any other manganese bath measurement. The weighted average of Table IV is not a recommended value, because the adoption of a value of 0.57 b for sulfur is certainly not warranted at this point. However, it is useful for putting into perspective the effect of the current unsettled nature of the sulfur cross section.

### Conclusion

It is essential that the sulfur absorption cross section be measured with an accuracy approaching 1%. This will improve the accuracy with which neutron source strengths can be measured in the manganese bath and contribute mightily to the final resolution of the ancient $^{252}$Cf $\bar{\nu}$ discrepancy.

TABLE IV
Cf $\bar{\nu}$ Status if Sulfur $\sigma_a$ = 0.57 b

| MEASUREMENT | PROJECTED $\bar{\nu}$ (Total) |
|---|---|
| **Liquid Scintillator** | |
| Spencer | 3.792+0.011 |
| Boldeman | 3.755+0.016 |
| Asplund-Nilsson | 3.792+0.040 |
| Hopkins-Diven | 3.777+0.031 |
| **Manganese Bath** | |
| Axton | 3.757+0.019 |
| DeVolpi | 3.761+0.019 |
| Bozorgmanesh | 3.758+0.023 |
| White, Axton | 3.829+0.040 |
| Aleksandrov | 3.761+0.036 |
| Smith | 3.78 +0.02 |
| **Boron Pile** | |
| Colvin | 3.739+0.021 |
| Wtd. Ave: | 3.771+0.006 |
| Ave: | 3.773+0.007 |

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