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Measurements using the manganese bath technique are central to the discrepancy that has existed between measured values of $\bar{\nu}$ for ²⁵²Cf. Manganese bath measurements of $\bar{\nu}$ belong to the lower group of values, while the η measurements are consistent with the higher $\bar{\nu}$ values. A three-part study was performed to see if the discrepancy could be explained by differences in manganese bath techniques: (1) A ²⁵²Cf source previously calibrated by De Volpi was calibrated in the MTR manganese bath; (2) The recommendations made by De Volpi for altering the MTR eta values were carefully considered; and (3) The results of the Monte Carlo calculation of the MTR experiment, carried out at Bettis Atomic Power Laboratory, were examined in detail. The study produced insignificant changes in the η values.

(Fissile nuclei, eta, ²⁵²Cf, nu-bar, manganese bath)

1. Introduction

For several years there has persisted a discrepancy in measured values of $\bar{\nu}$ for ²⁵²Cf. This discrepancy has from time to time been declared resolved,^{1,2} but continues to cause concern. One way to view this discrepancy is to examine the results of pertinent measurements made using the manganese bath. De Volpi et al³ and Axton et al⁴ have both measured $\bar{\nu}$, the average number of neutrons per fission, for ²⁵²Cf. Both have obtained the value 3.725, and both estimate errors of slightly less than 0.5%. Macklin and deSaussure and Smith et al^{7,8} measured η , the average number of neutrons per absorption, for the fissile nuclei. Their results agree well, and again accuracies of 0.5% are claimed. Values of $\bar{\nu}$ for the fissile nuclei are derived from measurements of their ratios to $\bar{\nu}$ for ²⁵²Cf. Then α , the absorption-to-fission ratio, can be used to connect $\bar{\nu}$ and η through the well-known relation

$$\bar{\nu} = \eta(1 + \alpha). \quad (1)$$

By way of Eq. (1), using the values for α and the $\bar{\nu}$ which were input to the 1968 IAEA evaluation of the fission constants,⁹ we can derive the values for $\bar{\nu}$ for ²⁵²Cf which are consistent with the η measurements. Table I shows the $\bar{\nu}$ values derived in this manner from the η measurements made at the Materials Testing Reactor (MTR) in 1964.

TABLE I
²⁵²Cf $\bar{\nu}$ VALUES CONSISTENT WITH η VALUES

Nucleus	η	α	$\bar{\nu}/\bar{\nu}_{Cf}$	$\bar{\nu}_{Cf}$ Derived
²³³ U	2.298	.09001	.6635	3.775
²³⁵ U	2.079	.17011	.6417	3.791
²³⁹ Pu	2.108	.35975	.7618	3.763
Average				3.776

The average value indicated for ²⁵²Cf is 3.776. This is about 1.4% higher than the value measured directly by De Volpi and Axton. Thus, the "hard core" of the discrepancy is of the order of 1%.

The problem of reducing the residual 1% spread is a difficult one. It may be that no single source of error is present in any experiment, but a composite of several small differences in several experiments. In view of efforts to attribute the problem primarily to

errors in the measurements,^{1,4} it seemed appropriate to reexamine the systematics of the experiments. The investigation has the following three principal lines of inquiry.

1. The data from a measurements in the MTR manganese bath of the strength of a ²⁵²Cf source previously calibrated in the ANL bath were carefully examined. The purpose was to look for any indication that the MTR bath had an intrinsically greater sensitivity to neutrons, real or spurious, which might explain the difference between the η and $\bar{\nu}$ results.

2. Careful consideration was given to the recommendations of De Volpi¹ for modifying the MTR η values using revisions of three manganese bath corrections.

3. A detailed comparison was made of the corrections to the MTR η experiments, as originally applied,⁷ with those indicated by the Monte Carlo calculation performed by Mitchell and Emert.¹⁰ This calculation agreed very well with the original analysis overall, but there remained the question of whether this agreement was really good throughout the experiment or merely resulted from chance compensation of differences in individual corrections.

II. Source Strength Comparison

The ²⁵²Cf source which was measured in the MTR manganese bath was designated MB-4. De Volpi had calibrated this source as having an emission rate of 7.467×10 neutrons per second as of May 1, 1968.¹¹ The disintegration constant he recommended was 7.212×10^{-4} day⁻¹.

The source was used to irradiate the manganese bath seven times during a period of four weeks in August 1969. The average saturated activity observed, corrected for system efficiency and reduced to De Volpi's calibration date, was 2.553×10^6 sec⁻¹. From this activity, the source strength was derived by means of two different sets of corrections. One was consistent with the corrections used in the η measurements, while the other used corrections consistent with De Volpi's systematics.¹² Since the measurement was made at only one concentration, it was necessary to assume a value for σ_H/σ_{Mn} , the ratio of the thermal absorption cross sections of hydrogen and manganese. Axton's value, 0.024955,¹³ was used with the MTR corrections, while De Volpi's value, 0.02531 was used with the second set. The results are summarized in Table II.

[†]Work supported by the Energy Research and Development Administration

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TABLE II

²⁵²Cf SOURCE COMPARISON

⁵⁶Mn saturated activity: 2.553 X 10⁶ sec⁻¹
(total bath, corrected to 1 May 1968)

Correction Factors

Effect	Original MTR	De Volpi
Leakage	.9938	.9946
High Energy Abs	.9941	.99941 [†]
Fraction Abs in Mn	.34849*	.34475 [†]
Other	.99740	.99740
Total (Product)	.34512	.34180
Source Strength	7.399 X 10 ⁶ sec ⁻¹	7.471 X 10 ⁶ sec ⁻¹
Difference from De Volpi	- .92%	+ .05%
De Volpi calibration	7.467 X 10 ⁶ sec ⁻¹	

* Assume Axton Value $\sigma_H/\sigma_{Mn} = 0.024965$

† Assume De Volpi Value $\sigma_H/\sigma_{Mn} = 0.02531$

While the agreement with De Volpi's calibration appears excellent, it is evident that the difference in the hydrogen-to-manganese cross section ratio dominates the observed differences. Moreover, the MTR counter calibration turns out to have been accurate to only about 1%. Thus, the agreement seems somewhat fortuitous. This comparison appears to preclude the existence of any large systematic sensitivity to neutrons between the two manganese baths, but is otherwise not particularly revealing as to the validity of systematic corrections employed.

III. The De Volpi Recommendations

Alex De Volpi¹ has recommended certain changes in three corrections applied to the MTR η experiments. If appropriate, these would lower the values by about 0.4%, and go far toward eliminating the discrepancy between ν and η . Specifically, De Volpi would change the MTR η values by +0.3%, -0.5%, and -0.2% through use of modified values for corrections for neutron escape, high energy parasitic absorption, and manganese resonance absorption, respectively.

The author continues to prefer the leakage calculation of Goldstein¹⁴ to De Volpi's recommended leakage correction. For the present purpose, however, it seems unnecessary to discuss the leakage correction separately. This is because De Volpi's leakage and parasitic absorption corrections are inseparably entwined. His determination of parasitic absorption directly depends upon the value he uses for leakage of neutrons from the manganese bath. Therefore, the appropriate correction to discuss is -0.2% for the combination of leakage and parasitic absorption. An adjustment in this direction may be appropriate, as is discussed below.

De Volpi's third recommendation, a decrease in η of 0.2% for manganese resonance absorption, is not appropriate. It apparently resulted from overlooking the fact that the resonance absorption correction for the η experiments enters in a form slightly different from that appropriate for source strength measurements. The intent was to apply Axton's method,¹⁵ which differs from the original correction only in applying a self-shielding correction to the manganese resonance absorption. When correctly applied, this method results in a rise in η of 0.2%, rather than a drop of

the same magnitude. Thus De Volpi's "corrected" recommendations would result in no net change in the MTR η values.

IV. Monte Carlo Calculation of the MTR Experiment

Mitchell and Emert¹¹ have made a thorough Monte Carlo (RECAP) analysis of the MTR measurement of η for ²³³U. The results agreed extremely well with the original analysis, returning a value of 2.296, compared with the original value of 2.298. Nevertheless, such an agreement could have resulted from a fortuitous cancellation of individually discrepant effects. It was felt that a review of the manganese bath corrections should include a detailed comparison of the correction values implied by the Monte Carlo calculation with those evaluated separately. Mitchell and Emert very kindly made the RECAP output listings available for such a study.

The results of this comparison are summarized in Table III.

TABLE III

COMPARISON OF CORRECTIONS TO Mn BATH EXPERIMENT

	Neutron Fraction	
	Original	RECAP
<u>Incident Beam Effects</u>		
Abs. in sample	0.99302 (E, C)	0.99321
Scatter	0.00430 (C)	0.00485
Abs. in Cd backup	0.00069 (E)	0.00067
Abs. in Cd sleeves	0.00160 (C)	0.00129
Abs. in Al cladding	0.00044 (C)	-----
Abs. in Al snout	0.0114 (E)	0.0078
<u>Fission Neutrons</u>		
Leakage	0.012 (C)	0.00246
Absorbed in Al	0.0008 (E)	0.0002
Abs. in Cd	0.0136 (E)	0.0123
Return to sample	0.00286 (C)	0.00519
Mn resonances abs.	0.0098 (C)	0.0099
Oxygen & sulfur abs.	0.0059 (C)	0.0037
Fast mult.	1.0307 (C)	-----

E = Experimental
C = Calculated

Regrettably RECAP did not record the tallies necessary for a direct comparison of the fast multiplication factor for the sample foils. Among those corrections that could be derived, the agreement is on the whole very good. However, there were three effects where differences seemed significant enough to warrant more detailed inquiry.

The RECAP calculation showed about 0.3% less absorption of open beam neutrons in the structural aluminum of the sample "snout" than measurements indicated. In this case the measured value is preferred. The calculation assumed the snout extension was made of pure aluminum, whereas it was ⁶¹Al aluminum. The material was unfortunately incompletely specified to the Bettis group when the RECAP model was set up.

The RECAP results indicated a little higher absorption of thermal neutrons in the fissile samples than the original corrections allowed. Upon closer examination, it appeared that the difference was principally due to absorption in the unprotected edges of the sample foils. The original corrections considered such absorptions in the face of the first foil, where the beam struck the sample, but neglected the edges. The RECAP results suggested that these edges

should be considered.

The third significant difference is in the high energy parasitic absorption in oxygen. RECAP yielded 0.285% for this absorption, compared to 0.48% resulting from the calculation by Goldstein.¹⁴ Since this result agreed both in direction and magnitude with De Volpi's net correction, revision of this correction seemed in order.

V. Revisions of the Corrections

Oxygen Absorption

The RECAP analysis and De Volpi's results indicated that the loss to $^{16}\text{O}(n,\alpha)^{13}\text{C}$ was not as great as given by Goldstein's calculation. Further confirmation came from a 46-group diffusion theory calculation of the MTR η experiment made with the MONA¹⁵ code. MONA agreed very well with the RECAP results. Since both these calculations used ENDF/B cross section, it was appropriate to investigate the cross section file used by Goldstein.

Goldstein used $^{16}\text{O}(n,\alpha)^{13}\text{C}$ cross sections from the evaluation by Kalos et al.¹⁷ The ENDF/B file is lower in the 6-8 Mev region, due principally to differences between the preliminary and final values for the measurements of Davis et al.¹⁸ A repetition of the Goldstein calculation is appropriate, but not at hand. Therefore, a renormalization of his result for the oxygen correction was attempted in terms of the average over the manganese bath spectrum of the oxygen absorption cross section. As shown in Table IV, the renormalized Goldstein calculation is in good agreement with the RECAP and MONA results, yielding 0.32% absorption compared to 0.28%. On the other hand, Version IV of ENDF/B is now available, and shows slightly increased cross sections for $^{16}\text{O}(n,\alpha)^{13}\text{C}$. As long as renormalization was in order, it was decided to renormalize to Version IV. The average oxygen absorption for the three renormalized calculations is 0.35%. Retaining Goldstein's value of 0.11% for the loss to $^{32}\text{S}(n,p)^{32}\text{P}$, we have a total parasitic absorption correction of 0.46%.

Other Adjustments

To the adjustments in corrections already mentioned were added recalculation of all fast multiplication corrections, using our original Monte Carlo program with ENDF/B-III cross sections, and recalculation of scattering by the sample cladding material, which was neglected in the original analysis. The revised manganese resonance absorption correction is based on Axton's method¹⁵ to include self-shielding

TABLE IV

NEUTRON LOSS BY $^{16}\text{O}(n,\alpha)^{13}\text{C}$			
Calculation	Calc. Loss (%)	Norm to ENDF/B-III	Norm to ENDF/B-IV
GOLDSTEIN	.48	.323	.381
RECAP	.285	.285	.336
MONA	.283	.283	.334
		Ave. .297	Ave. .350
	Loss to $^{32}\text{S}(n,p)^{32}\text{P}$.11	.11
	Total loss to charged particle interactions	.407	.407
Adopted correction factor: $1 - .0046 = .9954 \pm .0010$			
Cross Section Set		σ^*	
	Kalos et al	.0842	
	ENDF/B-III	.0566	
	ENDF/B-IV	.0668	

* $^{16}\text{O}(n,\alpha)^{13}\text{C}$ average cross section, weighted over spectrum of MTR Manganese Bath

effects, but is normalized to an above - $1/\nu$ resonance integral of 8.4 barns²⁰ instead of the 8.0 barns used by Axton. The correction changes are summarized in Table V. The changes are of fairly random sign and mainly small, except for the scattering correction for the nickel-clad ^{239}Pu sample. The latter change had only a modest effect on the ^{239}Pu η value, as only a part of the data taken involved nickel-clad samples.

VI. Readjustment by Least Squares Analysis

With the revised correction outlined above, it is appropriate to reanalyze the whole system of MTR η measurements by the method of least squares. The values to be shown here must for the present be considered interim values, pending consideration of the results of new Bettis Monte Carlo calculations¹⁹ and of experimental studies now being made of fast multiplication and indirect multiplication effects. Preliminary indications are that any changes forthcoming from either of these sources will be in the neighborhood of one or two tenths of a percent. Such results will not appreciably affect the picture presented here.

TABLE V

REVISIONS OF CORRECTIONS TO THE η EXPERIMENTS

Effect	Original	Revised	Effect on η
$^{16}\text{O}(n,\alpha)^{13}\text{C}$ & $^{32}\text{S}(n,p)^{32}\text{P}$	$0.9941 \pm .002$	$0.9954 \pm .002$	- .17%
Mn Res. Abs.	$1.0098 \pm .002$	$1.0083 \pm .002$	+ .15%
*Indirect Mult., foil edges	sample - dependent	sample - dependent	- .15%
*Scatter (Al clad)	sample - dependent	sample - dependent	+ .1%
*Scatter (Ni clad Pu)	$0.9988 \pm .0012$	$0.9899 \pm .0013$	+ .9%
*Fast Mult., ^{233}U	no change	no change	0.0%
*Fast Mult., ^{235}U	$1.0149 \pm .002$	$1.0146 \pm .002$	+ .03%
*Fast Mult., ^{239}Pu	$1.0237 \pm .002$	$1.0240 \pm .002$	- .03%
*Fast Mult., ^{241}Pu	$1.0149 \pm .002$	$1.0130 \pm .002$	+ .19%

*Representative examples

Table VI summarizes the results of the reanalysis for the data at 0.0253 eV. The 0.060 eV data have not yet been reanalyzed. Shown in separate columns are the results for the original 0.0253 eV data, the data taken in 1967 when the ^{241}Pu measurements were made, and a composite analysis including both sets of data simultaneously. Since the third set of values comes from a separate least-squares run, it does not necessarily represent averages from the first two columns.

For the second two columns the errors include not only the values yielded by the least-squares analysis, but an enlarged error including a contribution of 0.2% representing an ambiguity in the effect of solution concentration changes. The ^{241}Pu result includes also a 0.1% contribution convoluted to the ^{241}Pu error for uncertainties in the decay corrections.

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TABLE VI
REANALYZED η VALUES

Nucleus	η (1964 data)	η (1967 data)	η (All data)
^{233}U	2.298 \pm .009	2.291 \pm .010	2.295 \pm .009
^{235}U	2.080 \pm .010	2.082 \pm .009	2.081 \pm .009
^{239}Pu	2.110 \pm .008	2.106 \pm .009	2.110 \pm .008
^{241}Pu	not measured	2.166 \pm .010	2.165 \pm .010

After all the additional analysis that has gone into this revision, it appears that the errors in the corrections are reasonably random in sign. They almost balance out, leaving the η values essentially unchanged. The differences between the revised values and the originally published values are not statistically significant. This study revealed no justification for either lowering the η values or for expanding their errors to allow better agreement with the manganese bath measurements of $\bar{\nu}$ for ^{252}Cf . The discrepancy of 1% remains.

VII. Acknowledgments

The author would like to express his appreciation to A. De Volpi for discussions concerning manganese bath techniques, to J. A. Mitchell and C. J. Emert for providing the RECAP output and its interpretation, and to F. W. Sprakes for counsel in the least-squares reevaluation.

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