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REACTIVITY EFFECTS OF LOW NEUTRON ABSORBER SAMPLES IN
THE ADVANCED REACTIVITY MEASUREMENT FACILITIES

A. E. Wilson, E. Fast, J. W. Rogers, and D. A. Millsap

IDAHO NUCLEAR CORPORATION
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A B S T R A C T

The absorption statistical weights in the Advanced Reactivity Measurement Facilities are shown to be dependent upon the slowing down power of the sample medium containing the absorber. Measurements were made for light and heavy water solutions of boric acid and ^3He in the range of 0.0 to 2.0 cm^2 of total sample absorption cross section with 46 ml cylindrical samples. The observed increase in absorption statistical weight with slowing down power of the sample is attributed to the coupling between the neutron scattering and absorption. A simple mathematical model is presented that accounts qualitatively for the results. Reactor calculations are in general agreement with experimental observations.

Some data are also given of samples in which the absorption was maintained constant while the slowing down power was changed, and of samples of reduced density. Further measurements are suggested to improve the interpretation of these results.

This report sets forth in detail the work by the same authors presented in summary in Trans. Am. Nucl. Soc. 10, 224-5 (1967).

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REACTIVITY EFFECTS OF LOW NEUTRON ABSORBER SAMPLES IN THE
ADVANCED REACTIVITY MEASUREMENT FACILITIES

A. E. Wilson[†], E. Fast, J. W. Rogers, and D. A. Millsap

I. INTRODUCTION

Advanced Reactivity Measurement Facility (ARMF) reactors are generally used for reactivity measurements due to neutron absorption. Relatively small quantities of moderately strong absorbers can be measured with good precision with no appreciable interference. However, when materials of very low absorption cross section are measured reactivity effects usually include a significant component due to scattering. Work done previously on scatter reactivity effects has been reported⁽¹⁾. Some results obtained in this preliminary investigation were not well understood and therefore a further and more detailed study was considered desirable.

Results are reported here of experiments designed to show how neutron scatter reactivity effects might relate to those due to neutron absorption. An analysis based on a rather simple model indicates that coupling between the two effects would be expected. The purpose, then, was to show experimentally that indeed such coupling was observed. Calculations based on a one-dimensional diffusion code were also made and are in reasonable agreement with the experimental results.

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II. EXPERIMENTAL

A. The Reactors

The experimental work reported here was performed on the Advanced Reactivity Measurement Facilities, ARMF-I and ARMF-II. These reactors have been described in detail elsewhere⁽²⁾ and, therefore, only some of the principal features important to this study will be reviewed. Both of these reactors are pool-type facilities at opposite ends of the same tank in the ARMF building in the Test Reactor Area. They operate at low power, about 40 watts, in servo control and with a readout capability of the regulating rod positions that makes them unique in sensitivity and reproducibility. Measurements of reactivity change as low as one μk ($10^{-6} \Delta k/k$) are routinely made and reproducibilities of the order of 0.1 μk ($10^{-7} \Delta k/k$) are obtained. Data are recorded on IBM punch cards which are handled by a computer program⁽³⁾ to give the sample reactivity relative to the unperturbed reactor or a standard sample. A high degree of symmetry is retained in the core loading as may be seen in the diagrams of Figures 1 and 2. The vertical flux symmetry is further preserved by use of full length rotating control elements and by complete withdrawal of the safety rods during operation.

Each experimental sample is placed in the center of the reactor for measurement. The larger water hole of ARMF-I has a rather soft spectrum at the center in which both neutron scatter and absorption produce a negative reactivity change. The smaller sample hole of ARMF-II, 1.25 inch square, has a harder spectrum and shows a positive reactivity change due to scatter in addition to an increased

NOTE: Numbers on elements are grams of U^{235} .
Total 5085.1 g U^{235} .

Fig. 1. ARMF-I Core Loading

NOTE: Numbers on elements are grams of U^{235} .
Total 2654.9 g U^{235} .

Fig. 2. ARMF-II Core Loading

sensitivity for both scatter and absorption relative to ARMF-I. This provides the capability for studying the effect of scattering on reactivity under two considerably different conditions. Previous work⁽¹⁾ separated the reactivity change into neutron scattering and absorption components where the absorption was fairly small.

B. Samples

Three types of samples were considered in this study. The first type consisted of pure H_2O with varying amounts of boron added in the form of H_3BO_3 (boric acid). Since only very small amounts of boron were required to cover the range of absorption desired, it can be assumed that the scattering cross section remained constant while the absorption cross section changed. The second type sample was similar except that pure (99.8%) D_2O was used instead of H_2O . The boron dissolved in D_2O was also in the form of H_3BO_3 and the effect of the small amount of hydrogen added as a result was included in both absorption and scattering cross-section calculations. The third type of sample consisted of high purity (99.6%) 3He gas. This gas has a very large absorption cross section compared to the scattering cross section, which is the reverse of samples of types one and two. The gas samples were assumed to be pure neutron absorbers and were used as a reference for reactivity change due to absorption with no scattering. An attempt was made to use BF_3 gas also, but the reaction of the gas with the sample holder made it impossible to determine accurately by pressure measurements the amount of boron present. The various sample compositions and calculated absorption cross sections are given in Table I.

TABLE I. DATA FOR EXPERIMENTAL SAMPLES

A. H₂O Sample Data

Capsule Number	Wt. of Standard Solution (gm)*	Wt. of Pure H ₂ O (gm)	Σa (cm ²)	Measured Reactivity of Capsule in ARMF-II (μ k)			Measured Reactivity of Capsule in ARMF-I (μ k)		
				Empty	Filled	Net	Empty	Filled	Net
08-194	0	45.9365	1.0216	-391.72	-320.37	+ 71.35	331.12	134.82	-196.30
19-194	0.9733	44.8396	1.0400	-392.25	-333.72	+ 58.53	331.20	133.60	-197.60
16-209	1.9604	44.0471	1.0658	-380.65	-349.17	+ 40.48	330.72	130.85	-199.87
21-204	5.9667	40.0031	1.1520	-389.51	-403.77	- 14.26	329.59	124.83	-204.76
05-220	16.0151	29.8561	1.3680	-390.18	-544.82	-154.64	329.71	113.05	-216.66
06-193	30.0041	16.0846	1.6767	-390.02	-734.17	-344.15	330.42	99.84	-234.58
07-205	45.8896	0	2.0173	-385.74	**	----	329.94	77.87	-252.07

* Standard solution contains 0.5148 mg of standard boron per gram of solution in the form of H₃BO₃.
Remainder is pure H₂O.

** Reactivity change exceeded range of calibrated control rod.

TABLE I. (Cont)

B. D₂O Sample Data

Capsule Number	Wt. of Standard Solution (gm)*	Wt. of Pure D ₂ O (gm)	Σ_a (cm ⁻²)	Measured Reactivity of Capsule in ARMF-II (μ k)			Measured Reactivity of Capsule in ARMF-I (μ k)		
				Empty	Filled	Net	Empty	Filled	Net
01-202	0	50.8001	0.0036	-387.80	+ 49.25	+437.05	330.37	278.18	- 52.19
14-185	11.1004	39.7881	0.4312	-388.69	-187.86	+200.83	330.46	248.74	- 81.72
03-187	22.1401	28.8085	0.8563	-386.25	-407.35	- 21.10	329.14	221.66	-107.48
13-195	27.7494	23.1214	1.0723	-387.25	-517.58	-130.33	330.80	210.19	-120.61
09-199	33.9371	17.5790	1.2316	-390.22	-623.93	-233.71	330.52	198.41	-132.11
04-186	41.9371	8.8550	1.6185	-339.94	**	----	330.30	180.76	-149.54
22-219	50.9059	0.0	1.9639	-392.62	**	----	330.28	162.85	-167.43

* Standard solution contains 0.9101×10^{-3} gm of standard boron per gram of solution in the form of H₃BO₃. Remainder is 99.8% D₂O.

** Reactivity change exceeded range of calibrated control rod.

TABLE I. (Cont)

C. ^3He Sample Data

<u>Sample Number</u>	<u>Gas Pressure (mm of Hg)</u>	<u>Σ_a (cm²)</u>	<u>Net Reactivity in ARMF-II (μk)</u>	<u>Net Reactivity[*] in ARMF-I (μk)</u>
1	177	1.418	-680.95	-85.3
2	176.5	1.416	-682.73	
3	173	1.385	-666.29	
4	158	1.266	-612.60	
5	156	1.253	-613.66	-77.33
6	148	1.188	-579.67	
7	138	1.105	-538.89	
8	128	1.026	-506.25	-63.88
9	120	.963	-483.97	
10	118	.948	-471.63	-60.08
11	110	.882	-440.76	-56.03
12	102	.817	-408.47	
13	88.5	.711	-360.63	-46.27
14	88	.706	-359.97	
15	77	.616	-315.10	
16	68	.545	-276.03	-35.50
17	50	.402	-210.19	-27.24
18	42.5	.341	-181.37	
19	34	.274	-149.99	-19.52
20	28	.225	-121.38	
21	24	.192	-107.20	
22	15	.122	- 69.97	- 9.10
23	10	.080	- 47.41	

* Note: Not all samples were run in ARMF-I.

Compositions of boric acid solutions were prepared based on weight while the amount of gas in the type three samples was determined from pressure, temperature and volume measurements. The liquid samples were contained in capsules shown in Figure 3. The sample holders consist of an outer aluminum jacket designed for accurate location in the experimental positions of the ARMF reactors. A marlex plastic tube which fit snugly into the aluminum jacket held the liquid samples. An "O" ring seal on the aluminum jacket closure insured against any leakage into or out of the sample. Sample reactivity change measurements were made relative to the identical aluminum jacket and empty marlex capsule for each sample. Approximately 46 ml of solution was used in each of the liquid samples studied.

Sample holders for the gas samples, shown in Figure 4, were made as nearly identical as possible to the liquid sample holders. The assembled unit appeared identical for both gas and liquid sample capsules as shown in the lower item of Figure 4. The same type of aluminum jacket and of marlex tube was used and the sample volume was again 46 ml. The major difference was the inclusion of a small vacuum-tight valve, in the top of the aluminum jacket, of the type developed by R. J. Richards⁽⁴⁾ at the NBS Cryogenic Laboratory. By means of this valve, the gas could be encapsulated to a known pressure and the capsule sealed. After filling, an "O" ring sealed cap was placed over the valve parts to exclude water. All gas sample reactivity measurements were made relative to the evacuated gas sample holder. ^3He gas pressures ranged from zero up to about 180 mm of Hg for the data reported here. Pressure measurements were made of the filling system

Fig. 3. Capsules for Liquid Samples

Fig. 4. Capsule for Gas Sample

after equilibrium pressure and temperature were reached. Pressure readings were accurate to about ± 0.5 mm of Hg, and temperature was maintained at 21.0 ± 0.5 C.

C. Measurement Procedure

Reactivity measurements in the ARMF-I and -II followed the established procedure.^(2,3) A normal sequence included measurement of the empty or unperturbed reactor and/or of a comparison standard before and after several sample measurements. The samples encapsulated as indicated above were allowed to come to temperature equilibrium with the canal water at 18.000 ± 0.005 C before being placed into the center of a reactor for the measurement.

III. RESULTS OF REACTIVITY MEASUREMENTS

Total reactivity changes observed in ARMF-I for the samples of the three types are shown in Figure 5 as the sample reactivity in μk versus the total absorption cross section. The total absorption cross section of the light water sample could not be reduced below about 1 cm^2 since this represents the absorption of the hydrogen in 46 ml of pure H_2O . The absorption cross sections of the other two sample types could be adjusted below this value by adjusting the boron content or the helium gas pressure.

Since the reactivity data are plotted as a function of the absorption cross section, extrapolation of the curves to zero cross section provides a means of determining the effect of all mechanisms other than absorption on the reactivity (primarily scattering in these samples). The curve for the gas data passes through zero within the

Fig. 5. Total Reactivity in Samples of Various Absorber Concentrations, ARMF-I

experimental error. In the case of the D₂O data, there is an effect due to the low cross section n,2n reaction with fast neutrons, but it was calculated to be negligibly small compared to the effect due to scattering. A scattered neutron generally experiences a change in both energy and direction. Since the small samples were placed at the reactor center where the flux is isotropic, change in direction on the average should not perturb the system. It may be assumed that scattering of thermal neutrons has negligible effect on reactivity. The reactivity change at zero absorption cross section will, therefore, be interpreted as due only to energy degradation of the epithermal neutrons.

Fitting a second order polynomial to the data of Figure 5 resulted in the following analytical expression:

$$\text{Boron in H}_2\text{O: } \rho = (5.24 \Sigma_a^2 - 71.65 \Sigma_a - 128.9) \pm 0.4$$

$$\text{Boron in D}_2\text{O: } \rho = (5.71 \Sigma_a^2 - 69.71 \Sigma_a - 52.2) \pm 0.4$$

$$\text{He: } \rho = (6.57 \Sigma_a^2 - 69.35 \Sigma_a - 0.2) \pm 0.4.$$

The reactivity is in μk and the absorption is the total sample cross section (cm^2). The error limit given is the standard deviation of the measured values from the analytical expression. The three terms in each equation can be given the following physical significance. The constant term is the reactivity change due to neutron scatter for each type of sample. It can be seen that this effect is much larger for H₂O than for D₂O. The coefficient of the first order term is the slope of the curve at the zero absorption cross-section intercept and is, therefore, the absorption statistical weight at that point in Σ_a . The coefficient of the second order term is related to the self-

shielding of the 46 ml samples used in these experiments and would, in general, be a function of sample geometry.

The results of measurements of the same samples in ARMF-II are presented graphically in Figure 6. The corresponding fitted analytical expressions are:

$$\text{Boron in H}_2\text{O: } \rho = (52.8 \Sigma_a^2 - 776.1 \Sigma_a + 808.8) \pm 0.9$$

$$\text{Boron in D}_2\text{O: } \rho = (30.9 \Sigma_a^2 - 564.3 \Sigma_a + 439.0) \pm 0.6$$

$$\text{He: } \rho = (38.6 \Sigma_a^2 - 533.7 \Sigma_a - 1.3) \pm 2.7.$$

The range of absorption cross sections used in the ARMF-II is not as large as for the ARMF-I since the worth of the regulating rod was exceeded by the samples of higher absorption cross section. The relatively high standard deviation for the gas data ($\pm 2.7 \mu\text{k}$) is a result of inaccuracies in the gas pressure measurement and should not be attributed to uncertainties in the reactivity measurement.

Two important facts are apparent from the above data: First, the reactivity effect due to scattering is positive in ARMF-II as contrasted with the negative effect in ARMF-I. Thus, in the latter case, the partial reactivities add directly to give the overall sample reactivity. In ARMF-II, however, one effect tends to cancel the other, so that the net sample reactivity may be positive or negative, depending on the relative contributions of scattering and absorption. The fractional error of this measurement will be amplified since the net reactivity actually represents the difference between two relatively large effects. Second, the absorption statistical weight for a sample at any designated value of Σ_a depends on the slowing down power of the sample. This is most readily seen at $\Sigma_a = 0$ where the slope is given

Fig. 6. Total Reactivity in Samples of Various Absorber Concentrations, ARMF-II

simply by the coefficients of the first order terms of the above analytical expressions. In general, the absorption statistical weight is given by $\frac{\Delta\rho}{\Delta\Sigma_a}$ which is a function of Σ_a . From these observations it is apparent that a strong coupling exists between neutron scattering and absorption. A simple analysis presented later will show qualitatively that this coupling would be expected.

In order better to illustrate the coupling between absorption and scattering in effecting reactivity change, the data have been plotted in Figures 7 and 8 with the constant term obtained in the least-square fitting program subtracted from each data point. These data then represent the sample absorption reactivity as a function of Σ_a . Solid lines representing the best fit have been drawn for the gas and the H₂O data only in Figure 7. The data points for the D₂O samples lie between the two curves. While there is some difference in the slopes of the several curves of Figure 7, and qualitatively in the expected direction, the effect is not very pronounced. The neutron spectrum in the center of the ARMF-I experimental area is relatively soft and further scattering has but little effect on absorption probabilities. The coupling effect is much more pronounced in the data from ARMF-II shown in Figure 8. In this case, the neutron spectrum is relatively hard and any scattering of epithermal neutrons in the sample leads directly to an increase in the probability that the neutron will be absorbed in the sample, thus further reducing the reactivity.

A set of representative absorption statistical weights were calculated from the above analytical expression and are given in Table II. For an absorption statistical weight to be uniquely

Fig. 7. Moderator Effect on Absorption Reactivity, ARMF-I

Fig. 8. Moderator Effect on Absorption Reactivity, ARMF-II

TABLE II

REPRESENTATIVE ABSORPTION STATISTICAL WEIGHTS

<u>Sample</u>	<u>$\mu k/cm^2$ at Σ_a (cm^2)</u>		
	<u>0</u>	<u>1.0</u>	<u>2.0</u>
ARMF-I			
H ₂ O + B	- 71.65	- 61.17	- 50.69
D ₂ O + B	- 69.71	- 58.29	- 46.87
³ He	- 69.35	- 56.21	- 43.07
ARMF-II			
H ₂ O + B	-776.1	-670.5	-564.9
D ₂ O + B	-564.3	-522.5	-440.7
³ He	-533.7	-456.5	-379.3

determined requires specification of both the matrix material and the magnitude of absorption cross section for samples of a definite geometry. The predicted uncertainties in these values are largest (about 8%) for H₂O at zero cross section as a result of the large extrapolation required. This case is only of academic interest since the least absorption attainable with constant scattering cross section is that of pure water ($\Sigma_a \approx 1.0$ cm²). All other uncertainties are less than 4% and are as low as 0.25% (D₂O at $\Sigma_a = 1.0$ cm²).

The data shown in Table II make it apparent that a single value for absorption statistical weight cannot be assigned to either reactor for the purpose of evaluating absorption cross sections in samples for which there is a large reactivity effect due to scattering within the sample. However, by measuring or calculating the effect of

scattering on reactivity and using this value as a parameter, it should be possible to select an appropriate absorption statistical weight.

IV. MATHEMATICAL MODEL

The following mathematical model was used in the analysis of the foregoing experiments to explain the various interactions in the sample. Let $P(E)$ be the total event probability for a neutron of energy E entering the sample volume. Then

$$P(E) = p_x(E) + p_a(E) + p_s(E \rightarrow E', \Omega \rightarrow \Omega'), \quad (1)$$

where p_x is the probability that the neutron escapes the sample volume without interaction, p_a is the probability the neutron is absorbed and p_s , that it is scattered with a change of energy and direction. Assuming other possible interactions to be negligible, it is obvious the $P(E)$ equals unity. For simplicity it will be assumed that the small sample at the center of the reactor does not effect the reactivity because of a change in neutron direction, thus eliminating the functional dependence of $\Omega \rightarrow \Omega'$. The scattered neutron has the further possibility of undergoing an interaction the same as before, i.e., $p_x(E') + p_a(E') + p_s(E' \rightarrow E'')$. This line of reasoning may be continued for each successive scattering event until the neutron escapes or is absorbed. The process may be represented symbolically as follows:

$$\begin{aligned} P(E) = & p_x(E) + p_a(E) + p_s(E \rightarrow E')\{p_x(E') + p_a(E') \\ & + p_s(E' \rightarrow E'')[p_x(E'') + p_a(E'') + \dots]\} = 1, \end{aligned} \quad (2)$$

where $E \geq E' \geq E'' \geq \dots$

In an attempt to define an absorption reactivity, $\Delta\rho_a$, it has been assumed that $\Delta\rho_a$ is a function of all terms in the model which contain an absorption probability. Each such term is multiplied by the function $\phi^*(E)\phi(E)$ and integrated over all incident neutron energies. The result is proportional to the reactivity change due to absorption of neutrons in the sample. Thus, from Equation (2)

$$\begin{aligned}\Delta\rho_a &\propto \int_E p_a(E) \phi^*(E) \phi(E) dE \\ &+ \int_E \int_{E'} p_s(E \rightarrow E') p_a(E') \phi^*(E) \phi(E) dE' dE \\ &+ \int_E \int_{E'} \int_{E''} p_s(E \rightarrow E') p_s(E' \rightarrow E'') p_a(E'') \phi^*(E) \phi(E) dE'' dE' dE \\ &+ \dots \end{aligned} \quad (3)$$

This expression may be simplified by means of several assumptions based on the system used in the experimental program. These assumptions are:

1. The probability of scattering from energy E to E' is a constant for all samples of the same type and related to the macroscopic differential scattering cross section of the sample.
2. A scattering frequency function of the form

$$\Sigma_s(E \rightarrow E') = \frac{\bar{\Sigma}_s}{E(1-\alpha)}$$

may be used, where $\bar{\Sigma}_s$ is an average macroscopic scattering cross section in the epithermal region. This expression implies that any energy from E to αE is equally probable following a scattering event.

3. The probability of absorption at energy E is proportional to the change in macroscopic absorption cross section, $\Delta\Sigma_a(E)$, introduced by the sample.

4. The absorption cross section varies as $\frac{1}{\sqrt{E}}$ so that

$$\Delta\Sigma_a(E) = \Delta\Sigma_{a_0} \left(\frac{E_0}{E} \right)^{\frac{1}{2}}.$$

Substituting these assumptions in Equation (3) gives:

$$\begin{aligned} -\Delta\rho_a = & C \Delta\Sigma_{a_0} \sqrt{E_0} \int_E \frac{\phi^*(E)\phi(E)}{\sqrt{E}} dE \\ & + \frac{2C' \Delta\Sigma_{a_0} \sqrt{E_0} \bar{\Sigma}_s}{1 + \sqrt{\alpha}} \int_E \frac{\phi^*(E)\phi(E)}{\sqrt{E}} dE \\ & + \frac{4C'' \Delta\Sigma_{a_0} \sqrt{E_0} (\bar{\Sigma}_s)^2}{(1 + \sqrt{\alpha})^2} \int_E \frac{\phi^*(E)\phi(E)}{\sqrt{E}} dE \\ & + \dots \end{aligned} \quad (4)$$

From this the absorption statistical weight may be written as:

$$\frac{\Delta\rho_a}{\Delta\Sigma_{a_0}} = - \left[C_0 + \frac{C_1 \bar{\Sigma}_s}{(1+\sqrt{\alpha})^2} + \frac{C_2 (\bar{\Sigma}_s)^2}{(1+\sqrt{\alpha})^2} + \dots \right] \int_E \frac{\phi^*(E)\phi(E)}{\sqrt{E}} dE \quad (5)$$

Since this equation does not attempt to consider the effect of self-shielding, the absorption statistical weights correspond to those at low or zero absorption cross section. The first constant C_0 , must then correspond to the absorption statistical weight at zero absorption cross section for the gas sample in the data shown earlier. The other terms correspond to the differences observed between the gas data, i.e.,

zero scatter, and those for the H₂O and D₂O samples where scattering is important.

An expression for the scatter reactivity change may be obtained by relating it to the other terms in the model proposed in Equation (2). The neutron which enters the sample at energy E has a worth of $\phi^*(E)$ while the one which leaves at energy E' has a worth of $\phi^*(E')$. The change in worth is thus $\phi^*(E') - \phi^*(E)$. By reasoning similar to that used to obtain Equation (3), the scatter reactivity may be written as

$$\begin{aligned} \Delta\rho_s = & \int_E \int_{E'} p_s(E \rightarrow E') p_x(E') [\phi^*(E') - \phi^*(E)] \phi(E) dE' dE \\ & + \int_E \int_{E'} \int_{E''} p_s(E \rightarrow E') p_s(E' \rightarrow E'') p_x(E'') [\phi^*(E'') \\ & - \phi^*(E)] \phi(E) dE'' dE' dE \\ & + \dots \end{aligned} \quad (6)$$

Assume that the scattering probability is given by

$$p_s(E \rightarrow E') \propto \Delta N \sigma_s(E \rightarrow E') = \frac{\overline{\Delta \Sigma}_s}{E(1-\alpha)} \quad (7)$$

In Equation (7), the number of atoms in the sample is indicated by ΔN to emphasize the fact that the scatter reactivity change is measured relative to the empty sample holder. The scatter reactivity, Equation (6), then becomes

$$\Delta\rho_s = \frac{K\overline{\Delta \Sigma}_s}{1-\alpha} \int_E \int_{\alpha E}^E \frac{p_x(E')}{E} [\phi^*(E') - \phi^*(E)] \phi(E) dE' dE$$

$$\begin{aligned}
& + \frac{K'(\overline{\Delta\Sigma}_s)^2}{(1-\alpha)^2} \int_E^E \int_{\alpha E}^E \int_{\alpha E'}^{E'} \frac{p_x(E'')}{E E'} [\phi^*(E'') - \phi^*(E)] \phi(E) dE'' dE' dE \\
& + \dots
\end{aligned} \tag{8}$$

In the case of small samples, $p_x(E)$ is nearly equal to unity. If it can be assumed that $p_x(E)$ is a constant, a fairly good approximation over the epithermal energy range, then the terms containing the product $\phi^*(E) \phi(E)$ may be integrated and the result rearranged to obtain

$$\begin{aligned}
\frac{\Delta\rho_s}{\Delta\Sigma_s} = & - [K + K' \overline{\Delta\Sigma}_s + \dots] \int_E \phi^*(E) \phi(E) dE \\
& + \frac{K}{1-\alpha} \int_E^E \int_{\alpha E}^E \frac{1}{E} \phi^*(E') \phi(E) dE' dE \\
& + \frac{K' \overline{\Delta\Sigma}_s}{(1-\alpha)^2} \int_E^E \int_{\alpha E}^E \int_{\alpha E'}^{E'} \frac{1}{E E'} \phi^*(E'') \phi(E) dE'' dE' dE \\
& + \dots
\end{aligned} \tag{9}$$

It is difficult to interpret Equation (9) as a statistical weight due to scattering since the scattering cross section appears on the right hand side of the equation. Also, there is no common term for neutron worth as there was in the absorption statistical weight expression given in Equation (5). It is interesting to note, however, the similarity in form of Equation (5) and the first portion of Equation (9).

V. COMPARATIVE REACTOR CALCULATIONS

Calculations were made using an 18 group one dimensional multi-region diffusion code, AIM-6⁽⁵⁾. The appropriate cross sections were obtained from GAM-I⁽⁶⁾ and INCITE⁽⁷⁾, a locally developed code. Reactivity definitions used were based on theoretical work of Putnam⁽⁸⁾. Since a detailed report of calculations on scattering samples and related work is available⁽⁹⁾, only the principal features in the calculations will be given here along with pertinent results for comparison.

The group structure is that of equal lethargy groups of one lethargy unit width except for the last two. Group 17 is $3/4$ unit and group 18 includes the whole energy region below 0.532 eV. Cylindrical models of the ARMF reactors are assumed which are based upon the actual dimensions and fuel loadings. Results obtained from the program include real and adjoint fluxes averaged over each group. Scattering cross sections and reactivity contributions were calculated for scattering from any group to each group of lower energy. Tables III and IV are representative of the detailed information available. Note that the reactivity contribution from scatter involves primarily the higher energy neutrons whereas absorption occurs largely in the thermal group.

Comparison of the experimental results and the corresponding calculated values are shown in Table V for samples with absorption cross section of 1.02 cm^2 , the approximate absorption cross section of 46 ml of pure H_2O . It is apparent that there is fairly good agreement in most cases considered. The largest difference is in the

TABLE III. CALCULATED GROUP REACTIVITIES FOR H₂O IN ARMF-II

Lethargy Group, g	$\Delta\rho_a$ (μk)	Scatter Reactivity, ρ_s (μk) for Group				$^{17}\Sigma$ (g $\rightarrow$ g + k) k=5
		g $\rightarrow$ g + 1	g $\rightarrow$ g + 2	g $\rightarrow$ g + 3	g $\rightarrow$ g + 4	
1	- 5.06	27.16	14.92	7.16	2.92	1.82
2		117.98	63.07	27.70	11.10	6.98
3		99.64	51.06	22.19	9.00	5.86
4		50.10	26.61	12.06	5.18	3.56
5		26.04	15.31	7.64	3.43	2.48
6		17.28	11.66	5.97	2.73	2.08
7		15.36	10.01	5.15	2.44	1.87
8		12.45	8.47	4.58	2.19	1.70
9		11.36	8.03	4.32	2.07	1.60
10		10.83	7.55	4.06	1.89	1.55
11		10.18	7.10	3.68	1.85	1.50
12		9.58	6.30	3.66	1.92	1.34
13	- 0.36	7.89	6.42	3.90	1.55	1.33
14	- 2.14	9.86	7.65	3.28	2.80	
15	- 3.52	11.35	5.90	4.90		
16	- 5.55	6.94	5.02			
17	- 6.02	- 1.81				
18	-666.54					
				Total $\Delta\rho_a$	-689.19 μk	
				$\Delta\rho_s$	902.30	
				Net $\Delta\rho_s$	213.11	calculated
				Net $\Delta\rho$	72.11	experimental

TABLE IV. CALCULATED GROUP REACTIVITIES FOR D₂O IN ARMF-II

Lethargy Group, g	$\Delta\rho_a(\mu k)$	Scatter Reactivity, $\rho_s(\mu k)$, for Group				$\Delta\rho_{n,2n}(\mu k)$
		$g \rightarrow g + 1$	$g \rightarrow g + 2$	$g \rightarrow g + 3$	$g \rightarrow g + 4$	
1	-4.99	21.47	16.71	1.24	0.02	Total All Group 7.55
2		103.36	76.95	2.06		
3		82.22	41.75	0.76		
4		29.09	11.54	0.24		
5		10.20	4.24	0.11		
6		5.64	2.42	0.07		
7		3.80	1.77	0.05		
8		2.84	1.38	0.04		
9		2.52	1.28	0.04		
10		2.38	1.19	0.04		
11		2.22	1.11	0.03		
12		2.08	0.98	0.03		
13		1.70	0.99	0.04		
14	-0.01	2.10	1.17	0.03		
15	-0.01	2.40	0.98	0.11		
16	-0.01	1.48	0.59			
17	-0.01	-0.31				
18	-0.90					
		Total $\Delta\rho_e$				- 5.93
		$\Delta\rho_s$				444.55
		$\Delta\rho_{n,2n}$				7.55
		Net $\Delta\rho$				446.17 calculated
						437.0 experimental

TABLE V

COMPARISON OF CALCULATED AND EXPERIMENTAL REACTIVITY
CHANGES FOR $\Sigma_a = 1.02 \text{ cm}^2$

	Total Reactivity (μk)			
	<u>ARMF-I</u>		<u>ARMF-II</u>	
	Calculated	Experimental*	Calculated	Experimental*
H ₂ O	-226.2	-196.5	213.1	72.1
D ₂ O + H ₃ BO ₃	-121	-117.4	-101	-104.4
³ He Gas	- 74.2	- 64.1	-513	-505
	Absorption Reactivity (μk)			
	Calculated	Experimental*	Calculated	Experimental*
	Calculated	Experimental*	Calculated	Experimental*
H ₂ O	- 82.4	- 67.6	-723.5	-736.7
D ₂ O + H ₃ BO ₃	- 74.8	- 65.2	-535	-543.4
³ He Gas	- 74.2	- 63.9	-513	-504

* From analytical expressions representing experimental data, Section III.

total reactivity change for the H₂O sample in ARMF-II and appears to be the result of an over estimation of the positive reactivity change caused by scattering. The relative error in such cases should be based on the total reactivity change involved rather than the net reactivity observed which could be positive, negative or even zero. This error is less than 10%. Further comparison may be seen in Figures 9, 10 and 11 in which the experimental and calculated results are compared graphically.

Fig. 9. Reactivity of Water-Boric Acid Solutions in ARMF-I and ARMF-II

Fig. 10. Reactivity of Heavy Water-Boric Acid Solutions in ARMF-I and ARMF-II

Fig. 11. Reactivity of a $1/v$ Absorber Gas in ARMF-I and ARMF-II

It is also worthwhile noting that the trend in coupling between absorption and scattering is properly predicted by the calculations. Qualitatively, the absorption reactivity from Table V is greater for samples of higher moderating power. Thus, the higher order terms of Equations 4 or 5 are significant.

VI. RELATED EXPERIMENTS AND FURTHER WORK

The results of several related experiments, which chronologically preceded those reported above, need to be included. These relate specifically to measurement of samples in which the scattering cross sections are varied while absorption is held constant, and to samples of different densities, referred to as the "sponge" experiment. In brief, it is not readily apparent how the results of these experiments fit into the simple framework of the above mathematical model. Further experiments are suggested to help understand the phenomenon more completely.

A. Mixture Experiments

A series of samples were prepared consisting of mixtures of H_2O with a dilute solution of H_3BO_3 in D_2O . The boric acid concentration in D_2O was intended to be such that the absorption cross section would equal that of pure water. Thus, mixtures of the two in any proportions would maintain a constant absorption cross section, while the scattering cross section would vary between that of pure H_2O and D_2O .

Sample data and results of reactivity measurements in ARMF-I and ARMF-II are summarized in Table VI. These are compared with reactivities obtained by interpolation between the analytical expressions

TABLE VI. DATA FOR H₂O-D₂O MIXTURE SAMPLES

Number	H ₂ O gm	D ₂ O gm ^(a)	Σ_a cm ²	$\xi \Sigma_s$ cm ²	Reactivity of Sample (μk)			
					Experimental		Interpolated ^(b)	
					ARMF-I	ARMF-II	ARMF-I	ARMF-II
17702	45.8536	0.0	1.012	62.5	-194.78	+ 76.14	-196.04	+ 77.46
18013	34.9605	12.0443	1.038	49.8	-179.80	+ 28.76	-178.88	+ 19.02
18122	22.9035	25.1706	1.030	35.9	-160.39	- 29.56	-160.71	- 50.65
18223	10.9425	38.3820	1.091	22.1	-140.52	- 91.15	-140.55	-100.22
17909	0.2331	50.4352	1.118	9.5	-121.07	-149.87	-123.40	-152.45

(a) All samples contained 0.5136×10^{-3} gm standard boron as boric acid per gram of solution.

(b) These values obtained by linear interpolation, based on mole fractions, between the analytical expressions for experimental H₂O and D₂O curves of Section III.

TABLE VI. (Cont)

Sample Number	Reactivity (μk) Corrected to $\Sigma_a = 1.012 \text{ cm}^2$			
	Experimental		Interpolated ^(b)	
	ARMF-I	ARMF-II	ARMF-I	ARMF-II
17702	-194.78	+ 76.14	-196.04	+ 77.46
18013	-178.23	+ 45.10	-177.31	+ 35.36
18122	-156.36	+ 10.09	-156.68	- 11.00
18223	-135.91	- 48.56	-135.94	- 57.63
17909	-114.97	- 96.94	-117.30	- 99.52

presented in Section III. The linear interpolation between the H_2O and D_2O expressions was based on the mole fractions of each in these samples, and resulted in analytical expressions for Σ_a for the respective proportions of the two moderators. Since the goal of a constant Σ_a was not achieved exactly, these analytical expressions were used to correct the reactivity to the Σ_a for the pure water sample, i.e., to 1.012 cm^2 . The corrections so determined were then also applied to the experimental values. The final values of reactivities listed in Table VI are plotted versus the total slowing down power of the sample, $\xi\Sigma_s$, in Figure 12.

The adjusted experimental reactivities are in excellent agreement with the interpolated values for ARMF-I. Some significant differences appear in the results for ARMF-II for the intermediate mixtures. Since linear interpolation was assumed, the calculated values lie in a straight line. Hence, at $\xi\Sigma_s = 0$, the reactivity for $\Sigma_a = 1.012 \text{ cm}^2$ would be -103.2 and $-131.1 \text{ } \mu\text{k}$, respectively, for the two reactors. This condition cannot be attained experimentally with a D_2O - H_2O system while maintaining a full sample volume. Comparison with the corresponding values of -63.65 and $-501.9 \text{ } \mu\text{k}$ for ^3He in which $\xi\Sigma_s$ is essentially zero would indicate that a linear extrapolation is not valid.

B. "Sponge" Experiments

Reactivity measurements were made on a series of samples of pure H_2O and D_2O of various densities. The reduced density samples were obtained by use of cellulose sponges in the capsules, impregnated to varying degrees with the liquid up to complete saturation. Hence, the

Fig. 12. $\text{H}_2\text{O}-\text{D}_2\text{O}$ -Boron Mixtures in ARMF-I and ARMF-II

title of "sponge" was used. Details of the composition of samples and results of reactivity measurements are summarized in Table VII. The reactivity effect of the sponge was, of course, subtracted from the total along with that of the capsule to get the net reactivity for the sample. The results are also presented in Figures 13 and 14 as plots of reactivity as a function of $\xi\Sigma_s$. The values of Σ_a are proportional to $\xi\Sigma_s$, but is significant only for the H₂O samples since for D₂O, Σ_a is very nearly zero.

For comparison, reactivity values were also determined by interpolation between the analytical expressions of Section III for the values of $\xi\Sigma_s$ and Σ_a of the "sponge" experiment. The agreement was reasonably good when the interpolated values were obtained between the H₂O and ³He curves, although the section of the H₂O curve used was necessarily that extrapolated beyond possible experimental data of a full sample. However, linear interpolation between H₂O and D₂O curves for values of $\xi\Sigma_s$ and Σ_a identical to the pure H₂O samples of the sponge experiment disagreed systematically with the difference in sample densities. The point at which experimental data are available for direct comparison of the two experiments is at $\xi\Sigma_s = 9.5$ and $\Sigma_a = 0.1538 \text{ cm}^2$, where the D₂O-boron curve and the H₂O-sponge data curve each have these values of the parameters. The respective reactivities are -62.8 and -30.3 μk for ARMF-I and +352.9 and +35.0 μk for ARMF-11. A corresponding comparison cannot be made in the case of the D₂O sponge experiment because of the low value of Σ_a .

C. Discussion

The primary physical difference in the mixture and sponge experiments is that the first contains a full volume of sample liquid

TABLE VII. SPONGE EXPERIMENT DATA

A. ARMF-I

Sample Number	Weight Grams		Net Reactivity μ k	
	H ₂ O	D ₂ O	Experimental	Interpolated
10009	0.0349	10.9146	- 11.44	- 11.46
20009	0.0705	22.0179	- 22.84	- 22.92
31009	0.1082	33.8155	- 35.05	- 35.11
17807*	0.2337	50.5608	- 50.37	- 52.69
2090	1.9703		- 8.24	- 8.74
10090	9.9538		- 43.98	- 43.12
20090	19.9296		- 86.92	- 85.81
29791	29.6764		-129.93	-127.32
33890	32.8342		-142.32	-140.66
2014*	2.0095		- 9.11	- 8.89
10014*	9.9912		- 42.15	- 43.23
20014*	19.9819		- 85.66	- 85.94
33014*	32.9626		-142.53	-141.05
46014*	45.9496		-197.46	-195.98

* No sponge in capsule.

TABLE VII. (Cont)

B. ARMF-II

Sample Number	Weight in Grams		Net Reactivity (μ k)	
	H ₂ O	D ₂ O	Total	Interpolated
1723	0.0054	1.6942	15.25	13.45
3323	0.0106	3.3147	30.30	27.62
7323	0.0233	7.2565	67.04	62.04
18323	0.0580	18.2582	164.73	158.13
35923	0.1148	35.7580	316.37	310.95
17909*	0.1652	51.4590	448.02	448.02
1013	1.0326		5.77	4.51
2013	2.0180		10.63	10.36
4013	4.0022		21.96	20.67
11813	11.9204		63.10	55.24
17113	17.1486		73.81	71.87
25213	25.2496		87.94	87.61
33313	33.2445		94.65	92.08
17807*	46.0711		82.55	77.46

* Sponge absent from these samples. Pure liquid only.

Fig. 13. H_2O and D_2O "Sponge" Experiment, ARMF-I

Fig. 14. H_2O and D_2O "Sponge" Experiment, ARMF-II

and the second does not. Where a full sample volume is maintained, a reasonably consistent picture is presented, and a continuous and single-valued reactivity should be obtained for the $\xi\Sigma_s - \Sigma_a$ surface. Whether the same would be true for all variations of the sponge experiments is open to question. Available results indicate the actual sample volume, or the "void" volume, may need to be included as a variable. Some further experiments will be suggested here that would help complete the picture. However, results would probably be more of academic than practical interest.

Mixture samples of Σ_a , both greater and less than 1.012 cm^2 would fill in important areas where data are now lacking. As a complement to that series, one or two series of samples having definite proportions of H_2O and D_2O in which the H_3BO_3 content was varied would aid in establishing the interpolation form between the cases of pure solvents of these types.

Sponge type experiments with samples of selected proportions of H_2O , D_2O and H_3BO_3 would help to understand these systems and the effect of sample voids on reactivity measurements. Addition of this type of data would help complete the picture of the reactivity surface.

Comparative reactor calculations using actual sample compositions are desired for checking such calculations with experiment. Calculational analyses might also lead to a better understanding of the mechanism involved.

The agreement of the sponge experiment reactivities with those interpolated from the absorption curves of H_2O and ^3He is in some respects remarkable. The entire portion of the H_2O curve used was extrapolated beyond experimental data. This would indicate the

adequacy of the second-order power series expression in Σ_a for the extrapolation to $\Sigma_a = 0$ and lend confidence to its accuracy.

Considerable further theoretical interpretation is desirable. The assumed mechanisms are generalized from empirical and intuitive concepts. These could be put on a much better and more sophisticated basis using reactor theory and molecular-physical concepts. The effects of molecular structure and conglomerates on the reactivity effects of the near-thermal neutrons should be of interest. Other phenomenon such as upscatter likewise would improve the interpretation.

Comparison of the above results with measurements in a cadmium-shielded facility should be interesting and informative. The response due to neutron absorption in the sample would be largely negated and the scattering reactivity component emphasized. A boron-shielded facility, however, would vary the spectrum more uniformly and farther into the resonance region rather than providing a relatively sharp cutoff. Results of this type could assist in the theoretical interpretation, particularly in the scattering of neutrons in the low eV region where inter-molecular forces are of comparable energy. Inclusion of simple organics⁽¹⁰⁾ would add to the interesting possibilities of an expanded study.

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