Investigation of Deuterium Cross Section Data by Integral Testing: ZED-2 Measurements of High-Enriched Uranium Fuel Substituted into a Natural Uranium Core

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Abstract

Historical ZED-2 measurements of an HEU fuel rod substituted into a lattice of NU rods were analysed to determine their reactivity sensitivity to differences between the neutron elastic scattering cross-sections of deuterium from different evaluated nuclear data libraries. The differences in the deuterium nuclear data concern the angular probability distribution at neutron energies below 3.2 MeV. These ZED-2 experiments were selected due to the presence of HEU fuel in D_2O, since analyses of other critical experiments involving solutions of HEU fluoride in D_2O show substantial sensitivity (~10 mk) to these differences in the deuterium nuclear data. This analysis shows that the existing ZED-2 HEU experiments are insufficiently sensitive to resolve the discrepancy between the different deuterium data libraries. Further analysis of hypothetical configurations with high sensitivity shows that the sensitivity to the angular probability distribution of deuterium is strongly correlated with the leakage of fast neutrons, and it is recommended that further experiments to address this deuterium nuclear data issue be designed/evaluated to maximize this quantity.

1. Introduction

The accuracy of reactor simulations relies on the quality of information about the reactor, as well as modelling of fundamental nuclear processes as expressed in the neutron interaction cross sections of evaluated nuclear data libraries. Of particular importance for simulating heavy water reactors are neutron-deuterium reactions, since about 65% of the interactions that a typical neutron experiences involve deuterium [1], particularly through elastic neutron scattering.

The elastic scattering cross section for deuterium is represented as the product of an energy-dependent cross section and a probability distribution for a neutron of that energy to scatter in a particular direction, as indicated by the cosine of the scattering angle. Modern nuclear data evaluations for elastic scattering on deuterium agree reasonably well on the value of the energy-dependent cross section, with estimates of uncertainty at thermal energy (0.0253 eV) ranging from about ±0.35% [2] to ±2% [3]. However, large differences (>20%) exist in the angular probability distributions from different data evaluations at energies <3.2 MeV.

The differences in the angular probability distributions were found [4] to cause a large change of ~10 mk (1000 pcm) in the calculated neutron multiplication constant for a series of old critical experiments involving solutions of High-Enriched Uranium (HEU) fluoride in D_2O that were performed at the Los Alamos National Laboratory (LANL) [5]. While subsequent simulations of ZED-2 (Zero Energy Deuterium) experiments involving Natural Uranium (NU) fuel showed little sensitivity (<1 mk), they did show a small change in the D_2O coolant void reactivity (CVR) simulation bias (~0.6 mk). Accordingly, improvements to the deuterium data were sought via new nuclear-theory calculations [5] and new neutron-scattering measurements [6].

Since the new scattering measurements are proving to be quite challenging and the LANL HEU measurements had large experimental uncertainties (±3.3 to ±11.6 mk), the authors decided to see if new ZED-2 experiments using existing fuel materials could be defined with enhanced reactivity sensitivity to deuterium data by better matching the neutronic conditions of the LANL HEU experiments. Preliminary assessments [1] using a hypothetical compact critical core arrangement, consisting of 46 annular Zr-alloy fuel elements containing HEU UO_2 powder on a hexagonal lattice pitch of 5 cm, suggested that good reactivity sensitivity might be obtained if HEU material were used in ZED-2.

It was later realized that a series of ZED-2 experiments had already been conducted in 1980-81, wherein an experimental HEU fuel assembly was inserted into the centre of a lattice of NU fuel rods[1]. This paper describes the reanalysis of these experiments to see whether they show any sensitivity to the deuterium data, even though the HEU assembly occupies only a small region of the core and the HEU is configured with substantial other structural material. Insight gained from studying these measurements can be used to plan additional experiments to maximize sensitivity to the elastic scattering data of deuterium, with the goal of improving the nuclear data libraries used for safety analyses and licensing of heavy-water moderated critical facilities.

2. ZED-2 Reactor

ZED-2 is a low power research reactor. The reactor is made critical by pumping heavy water moderator into a 336 cm diameter calandria vessel containing a vertical lattice of fuel assemblies, and power is controlled by adjusting the moderator level. Typical moderator critical levels range from 150 to 250 cm above the calandria floor. The maximum power is about 200 watts (nominal), corresponding to an average neutron flux of $\sim10^9$ n/cm^2/s. Fuel type and core layout are both highly variable, but the main fuel charge typically consists of natural or low-enriched uranium oxide pellets packaged in fuel bundles, and loaded within CANDU-type fuel channels. A thorough description of the ZED-2 reactor is given in References [7] and [8].

Historically, use of ZED-2 has focused on validation of the CANDU reactor physics code suite, but is also available for public research as well as non-CANDU specific commercial work. Recent efforts have focused on expanding the availability for academic and fundamental research. Further details are given in Reference [7].

3. Description of Experiments

The experiments analysed consisted of critical height measurements for an experimental HEU fuel assembly inserted in the centre of a lattice of ZEEP rods. Both fuels are described below. Flux distribution measurements were also conducted but are not studied here.

[1] This work is documented in an AECL Report.

3.1 Experimental HEU Fuel Assembly

The HEU assembly contained sixteen fuel pins. Each pin comprised an HEU-Al alloy fuel, clad in a finned aluminum sheath (see Figure 1). A vertical string of four pins was inserted into each of four holes bored into the aluminum bar stock which makes up the rod. These holes are evenly spaced on a pitch circle diameter of 3.81 cm. Excess aluminum is machined off the rod around the holes containing fuel (see Figure 2). Details of the rod bottom were omitted from the experimental report, save that the four holes containing fuel pins were open at the bottom of the rod to allow moderator ingress.

3.2 ZEEP Fuel Rods

Each ZEEP fuel rod is composed of flat ended cylinders ("slugs") of natural uranium metal (18.835 g/cm^3) loaded into an aluminum tube ("sheath"). The tubes are nominally 0.1 cm thick with a 3.5 cm outer diameter, loaded with a stack of 19 fuel slugs (each nominally 6 in. (15.24 cm) long × 3.25 cm diameter) per rod. Each rod is sealed at the bottom with a welded cap, and has a top closure assembly sealed by a neoprene gasket. All rods were suspended with the bottom of the fuel nominally 15.0±0.3 cm above the calandria floor.

3.3 Lattice Geometry

The reference lattice employed for these measurements was a hexagonal lattice of 54 ZEEP rods at a 19.68 cm pitch, as shown in Figure 3 (with the central position, K0, empty). The other measurements employ the HEU assembly inserted in the centre of the lattice, with 0, 3, or 4 of its nearest neighbours removed.

The results of the critical measurements for all four lattice configurations are listed in Table 1.

4. Analysis Methodology

Sensitivity of k_{eff} to deuterium elastic scattering data is expected to increase with increasing ^{235}U enrichment. However, the bulk of the lattice is still NU, and therefore it is desired to isolate the effect of the HEU-D_2O interaction. To this end, these experiments were analysed using the MCNP-based substitution method, described in Reference [9]. This method attempts to remove calculation bias (i.e. $k_{eff} \neq 1$) by adjusting ν (and therefore k_∞) by an appropriate value (termed the Neutron Production Correction Factor, *NPCF*), specific to each fuel type in the simulation (if only a single fuel type is present, then the *NPCF* is equal to the inverse of the simulation k_{eff} value). The *NPCF* derived for a fuel from a substitution experiment will quantify the bias associated with that fuel type, separate from the influence of the reference fuel.

The MCNP-based substitution method was chosen in order to distinguish between the effects of the ZEEP rods (NU) and the HEU pins. The *NPCF* for the HEU fuel alone is used to construct a critical bare lattice simulation of only the experimental HEU assemblies. This simulation has an increased sensitivity to the interaction of the HEU fuel and D_2O moderator (at the cost of an increased uncertainty in k_{eff}).

Simulations were conducted using MCNP version 5 Release 1.40 [10], with a patch developed at AECL's Chalk River Laboratories (CRL) implemented to allow application of *NPCF*s. The continuous energy cross section libraries used were based on ENDF/B-VII.0 [11], created at CRL [12]. Sensitivities (Δk_{eff}) were calculated by changing all deuterium cross section libraries (including $S(\alpha,\beta)$) between the ENDF/B-VII.0 and ROSFOND-2008 evaluations [13]; these evaluations utilize different angular probability distributions of neutron elastic scattering below 3.2 MeV (peak difference >20%). ROSFOND-2008 ^{2}H employs the angular probability distribution of neutron elastic scattering from ENDF/B-VI.4, but is otherwise very similar to ENDF/B-VII.0.

5. Analysis

5.1 Reference Core Simulations

The initial reference lattice measurement was modelled (See Table 1) to derive an *NPCF* value for the ZEEP rods (=1/k_{eff} of simulation without the *NPCF* applied). This *NPCF* was then verified against a simulation of the final reference lattice measurement. The results are listed in Table 2 (rows 1 and 5). The uncertainty in *NPCF* assumes the only uncertainty in k_{eff} is the statistical uncertainty reported by MCNP ($\sigma k_{eff\text{-}MCNP}$), and is equal to it because the only fuel in the simulation is ZEEP rod fuel. This approach largely underestimates the NPCF uncertainty (0.1 mk compared to ~0.3 - 0.5 mk for similar experiments), but is sufficient to support the conclusions of this analysis.

5.2 Experimental HEU Fuel Assembly Substitution Simulations

The ZEEP rod *NPCF* was applied to a simulation of the HEU Assembly substitution measurement employing the 54 ZEEP rod lattice (See Table 1) in order to derive an *NPCF* for the HEU Assembly. This *NPCF* was then verified against simulations of the remaining HEU Assembly substitution measurements. The results are listed in Table 2 (rows 2-4). Again only considering the MCNP statistical uncertainties in k_{eff}, the uncertainty in the HEU Assembly *NPCF* ($\sigma NPCF_{HEU\ Assembly}$) was calculated as

$$\sigma NPCF_{HEU\ Assembly} = \left(\frac{dk_{eff}}{dNPCF_{HEU\ Assembly}} \right)^{-1} \sqrt{\sigma k_{eff-MCNP}{}^{2} + (dk_{eff} / dNPCF_{ZEEP})^{2} \sigma NPCF_{ZEEP}{}^{2}} \qquad (1)$$

where $dk_{eff}/dNPCF$ is the sensitivity of the substitution simulation k_{eff} to changes in *NPCF* of either the ZEEP or HEU Assembly.

Because only a single experimental HEU Assembly was substituted into the ZEEP rod reference lattice, the uncertainty in the HEU Assembly *NPCF* is ~100× that of the ZEEP rod *NPCF* (or ~10 mk from considering the MCNP statistical uncertainties alone). This results in a k_{eff} uncertainty from the HEU Assembly bare lattice simulation that is too large to distinguish between the results of simulations employing ENDF/B-VII.0 and ROSFOND data for deuterium elastic scattering (i.e. k_{eff} uncertainty > Δk_{eff}, despite not including experimental uncertainties). Therefore, the MCNP-based substitution analysis of the experiments listed in Table 1 was not pursued further.

5.3 Hypothetical HEU Assembly Experiments

Simulations of some additional, hypothetical, HEU Assembly substitution measurements were made to determine if additional HEU in D_2O measurements (particularly using a pin type of fuel assembly) should be pursued in ZED-2 to study deuterium scattering data.

Substitution measurements in ZED-2 were simulated with the lattice size reduced from 55 to 37 sites total, where HEU Assemblies were substituted into 1, 3, 5 or 7 of the centremost sites (where ZEEP rods occupy all of the remaining sites). The simulation of the 7-rod-substitution was repeated with the lattice pitch reduced to 18.0 cm (this pitch is still readily achievable within ZED-2). Sensitivities were calculated from these simulations to determine if these hypothetical experiments exhibited significant sensitivity to angular probability distribution, without employing the MCNP-based substitution method. The results are listed in Table 3 (with the initial substitution measurement from Table 1 included).

Additionally, bare lattice results were produced by filling a cylindrical region with a lattice of HEU Assemblies, at different lattice pitches. These results were used to determine sensitivity for HEU Assemblies under idealized conditions, and are listed in Table 4. All of the simulations with the experimental HEU assemblies show little increase in the sensitivity of k_{eff} to deuterium scattering data, with a maximum sensitivity (Δk_{eff}), attained from the 10.0 cm pitch bare lattice simulations, of only ~ −3 mk.

6. Discussion

The analyses of substitution experiments and hypothetical bare lattices of the experimental HEU Assemblies show that enrichment alone is insufficient to produce the large sensitivities observed in previous critical experiments. Experiments with uncertainties small enough to resolve the reduced sensitivity associated with this fuel would be unrealistic, requiring many of the experimental assemblies with extremely well defined geometries and compositions.

Previous experiments exhibiting high sensitivity were characterized by high neutron leakage and comparatively low deuterium to ^{235}U ratios. These were homogeneous solutions of HEU fluoride (UO_2F_2) in D_2O, as described in HEU-SOL-THERM-020 of Reference [14]. Case 5 of HEU-SOL-THERM-020 has a leakage of ~47% (i.e., 0.47 estimated MCNP particle weight loss to escape per source particle), and a D:^{235}U concentration ratio of ~2000:1. The neutron spectrum is thermal, and

~62% of all neutron collisions are on deuterium (compared to ~63% for the 10.0 cm pitch experimental HEU assembly bare lattice simulations). MCNP simulations of this benchmark measurement show a Δk_{eff} of ~7.7 mk from changing the angular probability distribution for elastic scattering of neutrons from deuterium.

6.1 Moderator/Fuel Ratio

The effect of changing the D:^{235}U ratio of the solution was studied using an MCNP model of a bare unreflected cylinder of the same solution as in case 5 of the HEU-SOL-THERM-020 benchmark. Simulations were produced by changing the atom density of uranium without altering the density of the solution, while adjusting the radius of the solution to maintain criticality. The Δk_{eff} values (from changing deuterium angular probability distribution data) are shown in Figure 4 for different D:^{235}U ratios. A strong correlation is seen from the results of the unreflected cylinder calculations. However, with the inclusion of data from the HEU Assembly bare lattice calculations, the results indicate that this correlation is highly dependent on reactor geometry (such as using separated or homogenized fuel and moderator). Therefore, consideration of the D:^{235}U ratio may be useful for maximizing sensitivity for experiments once fuel type is selected, but cannot be used as a primary metric for predicting the sensitivity of a measurement to deuterium elastic scattering data.

6.2 Leakage

The large leakage of the HEU-SOL-THERM-020 measurements was also investigated. MCNP simulations showed no significant correlation between the neutron weight lost to escape and Δk_{eff}, unless the neutrons are weighted according to their energy (i.e. effectively ignoring thermal neutrons). MCNP simulations of the experimental HEU Assembly bare lattices and HEU/D_2O solutions described above were redone to calculate the importance of energy of neutrons lost to leakage ($E_{Leakage,i}$), calculated as

$$E_{Leakage,i} = \int_{E_i} dE \sum_j w_j(E) \cdot E \tag{2}$$

where $w_j(E)$ is the weight of a neutron j, having energy E, which has leaked from the simulation boundary. All leaked neutrons with energies falling within the range of energy bin i are included in the integral. Δk_{eff} is plotted against the energy lost to leakage for several energy bins (including the sum over all energies) in Figure 5[2]. The results show a strong relationship with higher energy neutrons (which dominate the energy of neutrons lost to leakage). The hypothesized explanation for this behaviour is that fast neutrons escaping the simulation geometry will have had relatively few interactions with the moderator, and therefore the number of such neutrons has a higher sensitivity to the cross section and angular probability distribution of deuterium than does the much larger

[2] These plots include results from simulations of an infinite lattice of the experimental HEU Assemblies at 19.68 cm pitch, and the 28-element NU benchmark models from Reference [8] (where this last case includes the presence of a large D_2O and graphite reflector).

number of thermal neutrons escaping the core. Additionally, the greatest difference in angular probability distribution data between ENDF/B-VII.0 and ROSFOND-2008 appears to be for non-thermal neutron energies (see Reference [1]), thereby also weighting the sensitivity to higher energies. The fast flux leakage is therefore sensitive to the changing deuterium data, and the greater the number of fast neutrons escaping the core, the higher the sensitivity of k_{eff} to them. Leakage of fast neutrons therefore provides an excellent metric for predicting the sensitivity of an experiment to deuterium elastic scattering data. However, this relation was found for unreflected bare lattices. It is expected that this correlation will change with the addition of a reflector, due to the reduction in fast leakage produced. This has been confirmed from some preliminary analyses of the experiments listed in HEU-SOL-THERM-004 (Reflected spheres of HEU/D_2O solutions, [14]), as well as from simulations of experimental HEU assembly lattices.

7. Conclusions

Analysis of the 1980-1981 ZED-2 experiments utilizing experimental HEU assemblies showed no discernable sensitivity to the differences in deuterium scattering data between ENDF/B-VII.0 and ROSFOND, whose differences are mainly in the energy-dependent angular scattering probability distribution rather than in the elastic scattering cross section for thermal neutrons. Simulations of other possible configurations indicate that experiments in ZED-2 with these experimental assemblies will not result in a useful measurement for studying these differences in scattering data (a relatively small Δk_{eff} with large uncertainty could be expected). However, further study of previous experiments which exhibit either high or low sensitivity to the changes in deuterium elastic scattering data showed that this sensitivity is strongly correlated with fast leakage. Therefore, it is recommended that further experiments/analyses to investigate the angular probability distribution of neutron elastic scattering from deuterium be designed/selected based on maximizing fast flux leakage from a bare critical core (the strong correlation does not hold for reflected assemblies).

8. Acknowledgements

The authors would like to thank Michael Atfield for helpful discussions and a thoughtful review, Bruce Wilkin for developing the NPCF patch in MCNP5, and Peter Pfeiffer for implementing the patch on the parallelized computer cluster used in the analysis.

9. References

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Table 1: Results of Critical Height Measurements

Date	Time	Core Configuration	Critical Moderator Height (cm)	T (°C)	D_2O Purity (wt%)
Nov. 20, 1980	9:03	54 ZEEP Rod Reference Lattice (K0 Empty)	177.093	22.16	99.67
	9:55	54 ZEEP Rod Reference Lattice with HEU Assembly at K0	173.856	22.21	
	10:48	51 ZEEP Rod Lattice with HEU Assembly at K0 (K2W, J1E, L1E Removed)	189.392	22.25	
	13:50	50 ZEEP Rod Lattice with HEU Assembly at K0 (J1E, L1E, J1W, L1W Removed)	196.450	22.36	
	14:46	Repeat of 54 ZEEP Rod Reference Lattice	177.173	22.36	

Table 2: *NPCF* Values Calculated From Substitution Analysis

Core Configuration	ZEEP *NPCF*	$\sigma NPCF_{ZEEP}$	HEU Assembly *NPCF*	$\sigma NPCF_{HEU\ Assembly}$	k_{eff}	$\sigma k_{eff\text{-}MCNP}$
54 ZEEP Rod Reference Lattice (K0 empty)	0.99878	~0.0001	-	-	1.00007	0.00008
54 ZEEP Rod Reference Lattice with HEU Assembly at K0	0.99878	~0.0001	0.94000	~0.01	1.00003	0.00008
51 ZEEP Rod Lattice with HEU Assembly at K0 (rods at K2W, J1E, L1E removed)	0.99878	~0.0001	0.94000	~0.01	0.99996	0.00007
50 ZEEP Rod Lattice with HEU Assembly at K0 (rods at J1E, L1E, J1W, L1W removed)	0.99878	~0.0001	0.94000	~0.01	0.99987	0.00008
Repeat of 54 ZEEP Rod Reference Lattice	0.99878	~0.0001	-	-	1.00005	0.00007

Table 3: Sensitivity of Hypothetical ZED-2 Experiments to Change in Deuterium Elastic Scattering Data

Core Configuration	Moderator Height	ENDF/B VII.0		ROSFOND		Δk_{eff} (mk)	σ_{MCNP} (mk)
		k_{eff}	σk_{eff_MCNP}	k_{eff}	σk_{eff_MCNP}		
54 ZEEP Rod Reference Lattice with HEU Assembly at K0[3]	173.856	1.00195	0.00008	1.00183	0.00008	0.12	0.11
36 ZEEP Rod Reference Lattice with HEU Assembly at K0	224	1.00007	0.00008	1.00018	0.00008	-0.11	0.11
34 ZEEP Rod Lattice with 3 HEU Assemblies at K0, L1E and L1W	233	0.99952	0.00008	0.99906	0.00008	0.46	0.11
32 ZEEP Rod Lattice with 5 HEU Assemblies at K0, J1E, L1E, J1W, and L1W	248	0.99978	0.00008	0.99942	0.00007	0.36	0.11
30 ZEEP Rod Lattice with 7 HEU Assemblies in Centre	273	0.99956	0.00008	0.99910	0.00008	0.46	0.11
Repeat of 30 ZEEP Rod Lattice with 7 HEU Assemblies in Centre at 18.0 cm Pitch	256	0.99928	0.00008	0.99926	0.00008	0.02	0.11

[3] This measurement was conducted as part of the 1980 experiments with the experimental HEU fuel assembly (See Table 1).

Table 4: Sensitivity of HEU Assembly Bare Lattices to Change in Deuterium Elastic Scattering Data

Hexagonal Lattice Pitch (cm)	Height (cm)	Diameter (cm)	ENDF/B VII.0		ROSFOND		Δk_{eff} (mk)	σ_{MCNP} (mk)
			k_{eff}	σk_{eff_MCNP}	k_{eff}	σk_{eff_MCNP}		
18.0	114.75 (6 pins)	314	1.00010	0.00011	1.00124	0.00012	-1.14	0.16
10.0	76.50 (4 pins)	380	0.99914	0.00012	1.00181	0.00012	-2.67	0.17
10.0	191.25 (10 pins)	140	1.07414	0.00011	1.07647	0.00012	-2.33	0.16

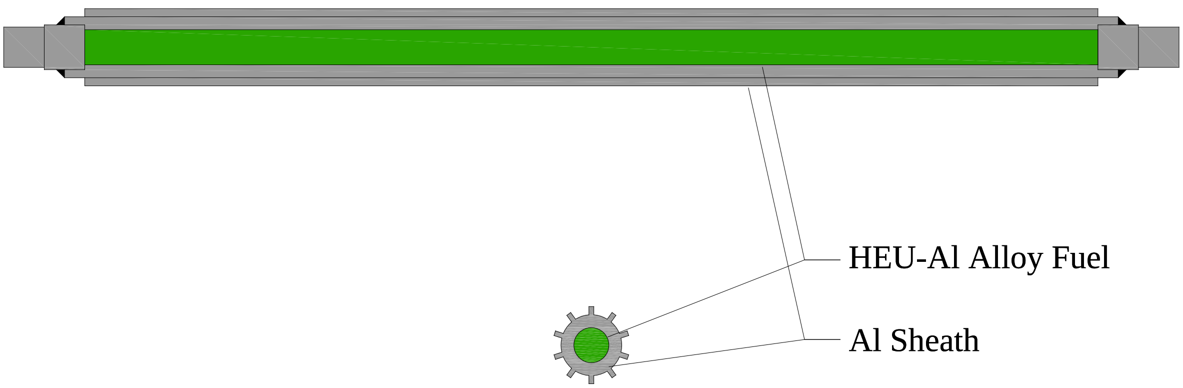

Figure 1 Experimental HEU-Al Fuel Pin

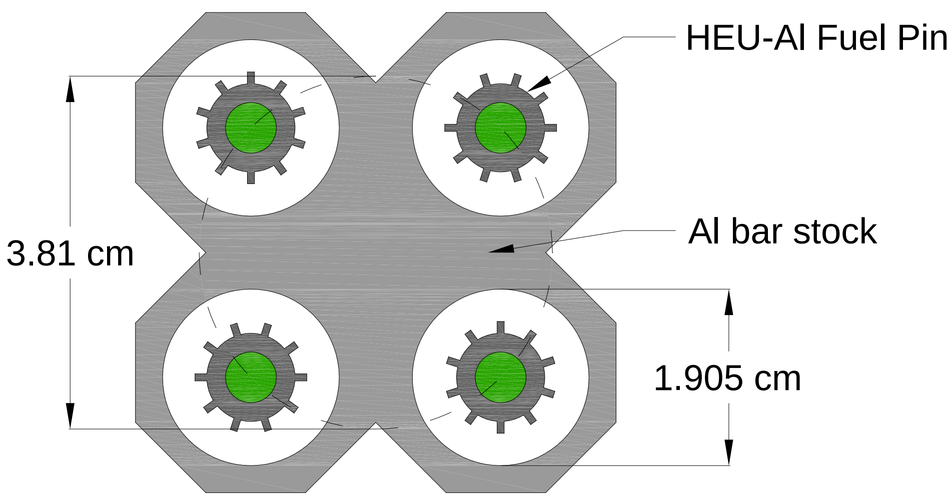

Figure 2 Cross section of Experimental HEU Assembly Used for ZED-2 Experiments

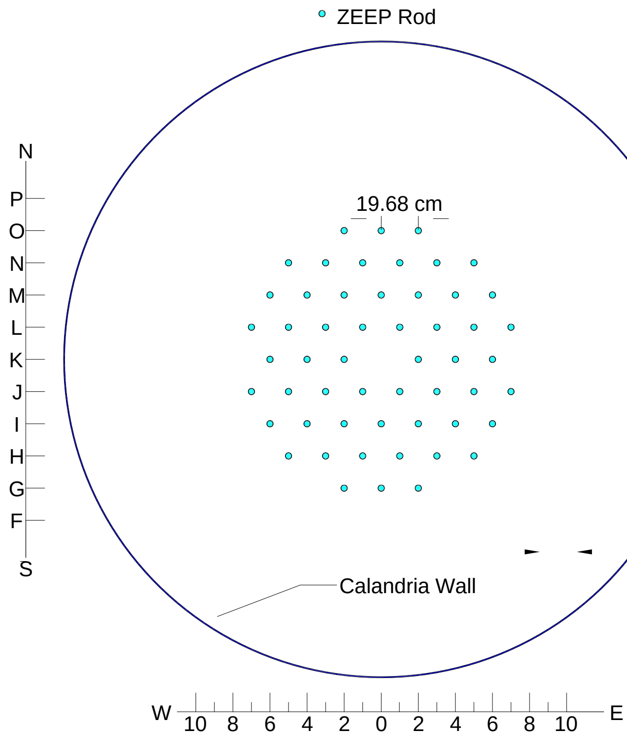

Figure 3 Plan View of 54 ZEEP Rod Reference Lattice (K0 Empty)

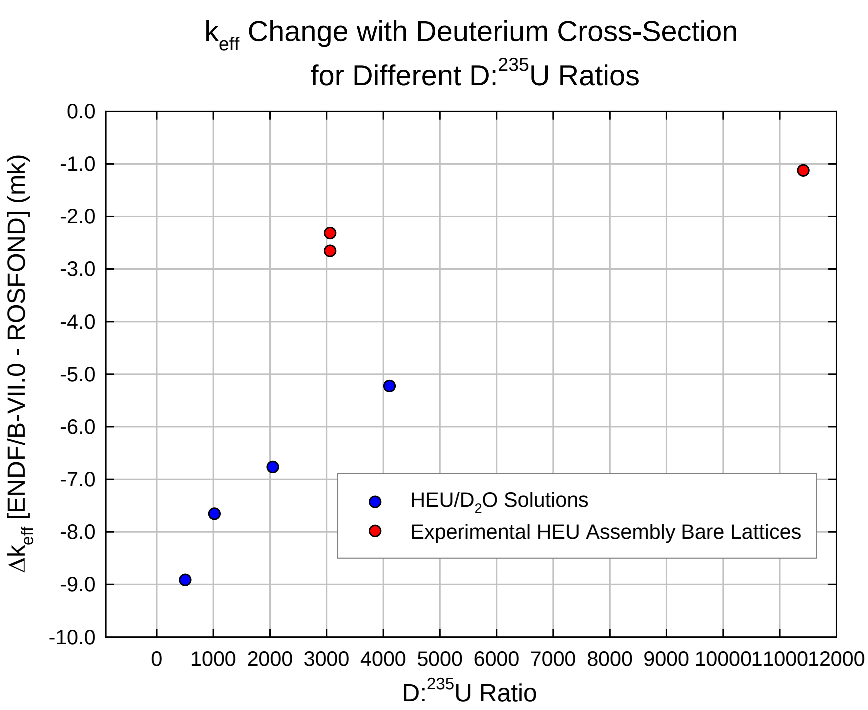

Figure 4 Sensitivity of Core to Deuterium Scattering Data Evaluation as a Function of D:^{235}U Ratio

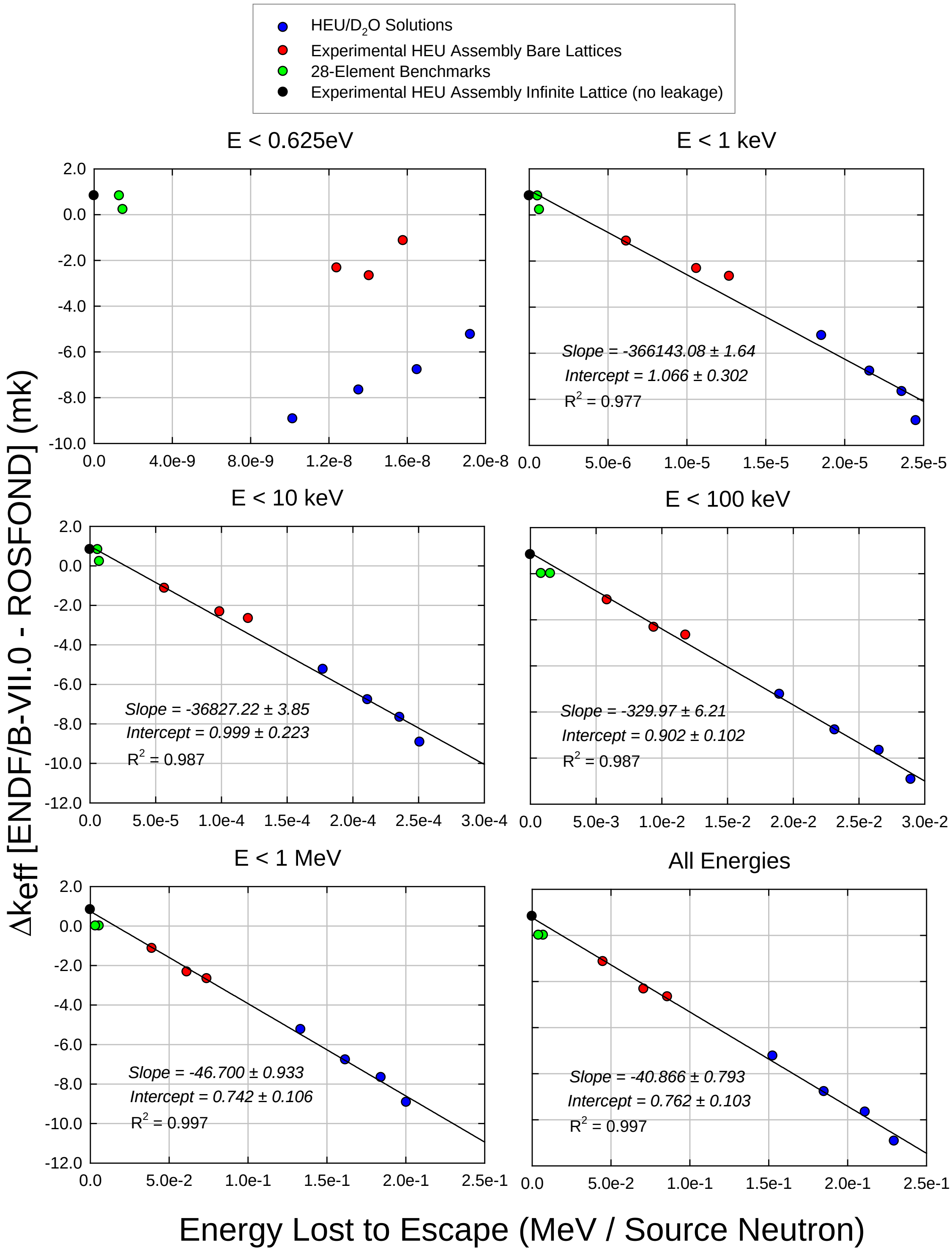

Figure 5 Sensitivity of Core to Deuterium Scattering Data Evaluation as a Function of Energy lost to Leakage for Several Neutron Energy Ranges