Thermal Neutron Scattering Law for Beryllium Hydride and Critical Mass Calculations

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ABSTRACT

The thermal neutron scattering law (TSL) for crystalline beryllium hydride (BeH_2) is developed from first-principles *ab initio* lattice dynamics calculations and the impact of neutron thermalization in this material on critical mass is estimated. BeH_2 has a body-centered orthorhombic crystal structure with 12 molecules per unit cell and a theoretical density of 0.755 g/cm^3. The vibrational (phonon) densities of states for H and Be bound in BeH_2 are determined using VASP density functional theory and PHONON lattice dynamics calculations. The TSLs for H bound in BeH_2, H(BeH_2), and Be bound in BeH_2, Be(BeH_2), are then evaluated in the incoherent approximation from the calculated H and Be partial phonon density of states using FLASSH. Finally, critical mass as a function of ^{235}U loading density for bare and reflected BeH_2 moderated spheres is predicted from MC21 Monte Carlo neutron transport calculations using ENDF/B-VIII.0 cross sections and the H(BeH_2) and Be(BeH_2) TSL evaluations. Comparisons are made to water (H_2O), polyethylene (CH_2), and beryllium oxide (BeO) as moderators. These critical mass predictions are a refinement upon the prior work by Rao and Srinivasan that neglected thermal neutron scattering effects. The minimum critical mass of a BeH_2 moderated assembly is estimated to be 0.207 kg ^{235}U for a 0.20 m thick BeO reflected sphere and 0.178 kg ^{235}U for a 0.40 m thick BeO reflected sphere.

Key Words: Beryllium Hydride, BeH_2, Thermal Neutron Scattering Law, Critical Mass

1 INTRODUCTION

Beryllium hydride (BeH_2), with a hydrogen number density 24% greater than water, is an alkaline earth metal hydride that received considerable attention as a potential high-performance neutron moderator and reflector early in the nuclear era [1,2]. Early work to synthesize pure crystalline BeH_2 in the 1950s-1960s proved to be challenging. However, researchers were able to produce a high-purity amorphous form of BeH_2 consisting of long chains having many repeating BeH_2 monomer units [1,3,4]. In 1978, Brendel [5] developed a process to synthesize high-purity crystalline BeH_2 based on high-pressure compaction-fusion of amorphous BeH_2. Two stable crystallographic phases were observed: a low-temperature phase below 523 K (250 ºC) and high-temperature phase. Brendel reported bulk density changes from 0.62-0.65 g/cm^3 for the amorphous BeH_2 feed material, to 0.73-0.74 g/cm^3 for the low-temperature phase, and 0.77-0.78 g/cm^3 for the high-temperature phase. Initial X-ray diffraction measurements by Brendel [5] incorrectly indexed the low-temperature phase of BeH_2 as hexagonal and were not able to accurately determine the structure of the high-temperature phase. The crystal structure of the low-temperature phase of BeH_2 was more precisely determined by Smith [6] using high-resolution X-ray diffraction data obtained at a synchrotron radiation source, where he determined it to have a body-centered orthorhombic crystal structure with 12 BeH_2 molecules per unit cell and a theoretical density of 0.755 g/cm^3.

Rao and Srinivasan [7] estimated the minimum critical mass of bare and reflected spherical assemblies fueled with ^{235}U, ^{233}U, and ^{239}Pu and moderated by BeH_2 based on the bulk density of 0.78 g/cm^3

reported by Brendel [5]. They found that the "minimum critical masses for thick BeO-reflected spherical systems having BeH_2 moderator of density ≥680 kg/m^3 are lower than with any other known moderator" [7]. These minimum critical mass estimates were based on one-dimensional discrete ordinates neutron transport (DTF IV) calculations using 16-group Hansen Roach cross sections and neglected the effect of the thermal neutron scattering in BeH_2.

In this paper we calculate the phonon density of states (DOS) for the low-temperature phase of BeH_2 using the *ab initio* lattice dynamics (AILD) calculation. These DOS are used to evaluate the thermal neutron scattering law (TSL) for hydrogen bound in BeH_2, H(BeH_2), and beryllium bound in BeH_2, Be(BeH_2), in the incoherent approximation. These TSLs are then used in MC21 [8] continuous-energy Monte Carlo calculations to estimate the minimum critical mass of bare and reflected HEU-fueled and BeH_2-moderated spherical configurations.

2 CALCULATION OF PHONON DOS

As illustrated in Figure 1, BeH_2 has body-centered orthorhombic (space-group Ibam) crystal structure with 12 molecules per unit cell [6]. The molecular structure is based on a network of corner-sharing BeH_4 tetrahedra with bond angles that range from 107º to 113º. The atom site locations are provided in Table I. The Be(1) molecules are located along the z = 0.0 and 0.5 planes while the Be(2) molecules are located along the z = 0.25 and 0.75 planes. The VASP code [9,10] was used to perform first-principles quantum mechanics simulations of the BeH_2 lattice structure using density functional theory (DFT). The BeH_2 lattice structure was optimized using a total electronic energy threshold of 10^{-7} eV, a 0.3 Å^{-1} k-point spacing (7×7×7 k-point mesh), a 500 eV planewave cutoff energy, and the GGA-PBE [11] exchange-energy correlation functional. As shown in Table II, the VASP optimized 0 K lattice constants are in good agreement with the room-temperature lattice constants measured by Smith [6], with a maximum difference of -1.5%.

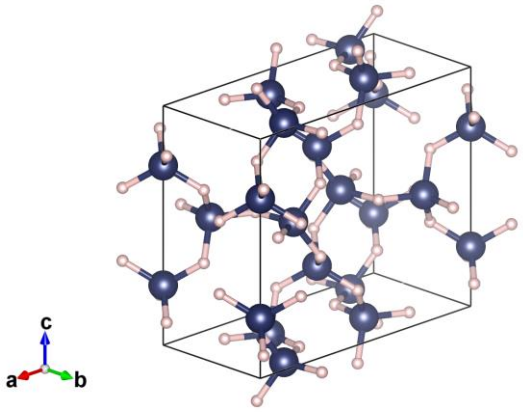


Figure 1. BeH_2 Unit Cell. Be atoms are colored blue and the H atoms rose. The Be-H bonds extend beyond the unit cell boundary to illustrate the corner-sharing BeH_4 tetrahedra.

Table I. Atom Locations for BeH_2

Atom	x	y	z
Be(1)	0.1677	0.1199	0.0
H(1)	0.3114	0.2750	0.0
Be(2)	0.0	0.0	0.25
H(2)	0.0884	0.2265	0.1524

Table II. Optimized BeH_2 Unit Cell Lattice Parameters

Lattice Parameter	Experimental, Room Temperature (Å)	Calculated, 0 K (Å)	Relative Change
a	9.082(4)	8.947337	-1.5 %
b	4.160(2)	4.127790	-0.8 %
c	7.707(3)	7.634728	-0.9 %

Lattice dynamics calculations were performed to determine the phonon DOS for crystalline BeH_2 using the PHONON [13] code. Interatomic Hellman-Feynman forces on a 1×2×1 supercell (76 atoms) with ± 0.02 Å asymmetric atom displacements were determined from a series of VASP calculations using the equivalent parameters to the structure relaxation. Our calculated phonon DOS are provided in Figure 2. There are four broad bands. The lowest energy band from 0.0-0.069 eV is due to low-frequency vibrations of the "rigid" BeH_4 units in the structure. The band between 0.069-0.103 eV, with a peak at 0.100 eV, is due to intramolecular H-Be-H deformation/bending modes (ν_2). The next broad band was between 0.127-0.174 eV, with a peak at ~0.159 eV, is due to symmetric Be-H stretching modes (ν_1). The high-energy band between 0.231-0.253 eV, with a peak at ~0.242 eV, is due to intramolecular Be-H antisymmetric stretching modes (ν_3). To our knowledge, no inelastic neutron scattering measurements on crystalline BeH_2 are available to validate our calculated phonon DOS. However, our calculated phonon DOS are in good agreement with inelastic neutron scattering measurements on amorphous BeH_2 by Sampath [13] which reported four broad band from 0.0-0.050 eV, 0.050-0.120 eV (peak at ~0.097 eV), 0.120-0.180 eV (peak at ~162 eV), and 0.200-0.260 eV (peak at ~0.224 eV).

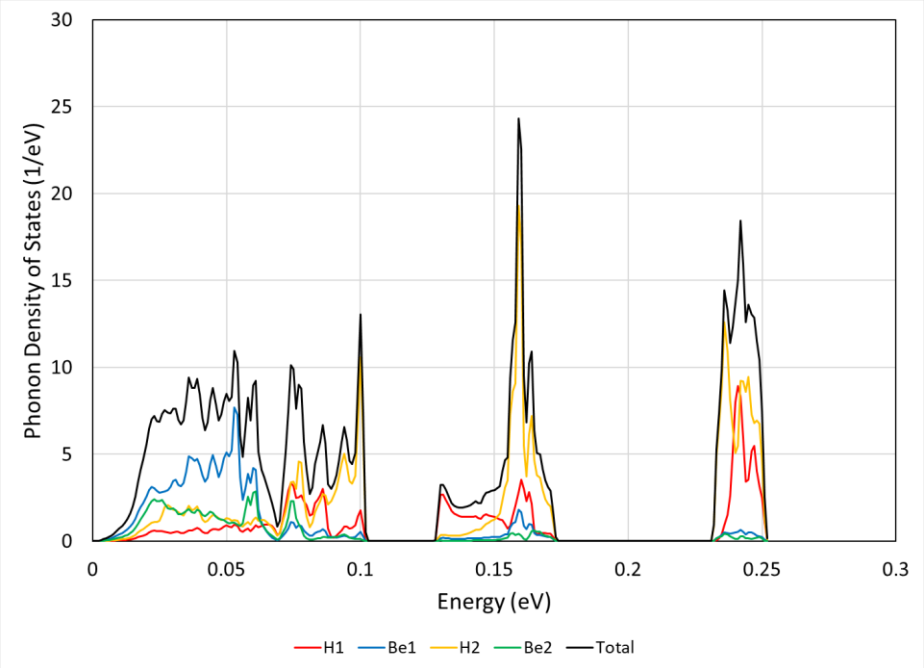


Figure 2. Calculated Phonon Density of States for BeH_2.

Normalized effective H and Be phonon DOS suitable for use in the FLASSH [14] or NJOY/LEAPR [15] TSL evaluation codes were prepared by summing the H(1) and H(2) contributions and weighting by 18/12 and summing the Be(1) and Be(2) contributions and weighting by 18/6. The effective H and Be phonon DOS are provided in Figure 3.

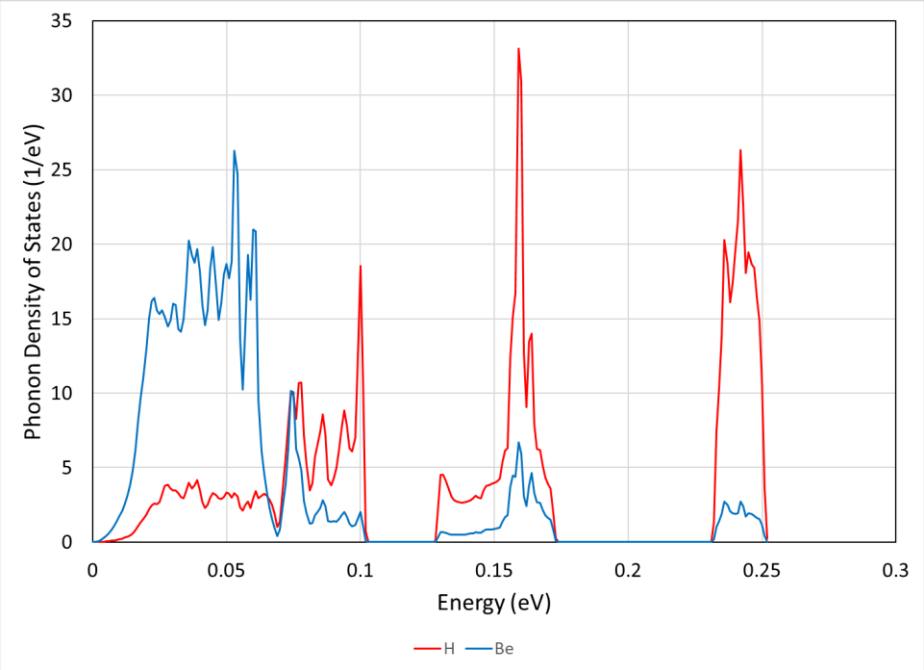
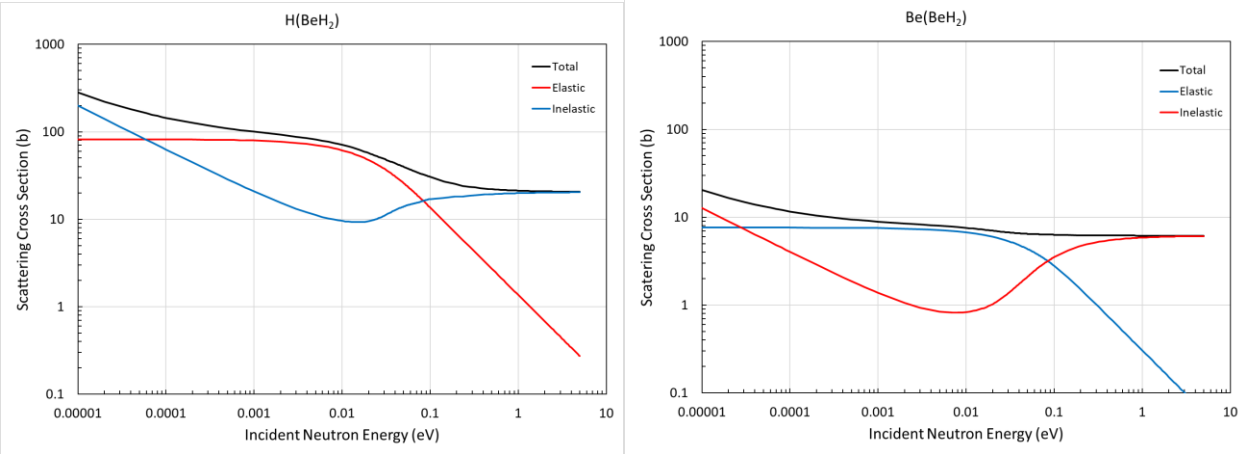


Figure 3. Effective H and Be Phonon DOS for BeH_2.

3 THERMAL NEUTRON SCATTERING LAW

The H(BeH_2) and Be(BeH_2) TSLs were evaluated in the incoherent approximation at four temperatures (293.6 K, 400 K, 500 K, and 523 K) using the FLASSH code [14]. Free-atom scattering cross sections of 20.43608 b for ^{1}H and 6.153875 b for ^{9}Be were used to ensure consistency with the ENDF/B-VIII.0 evaluations for these nuclides [18]. The normalized H and Be partial phonon DOS provided in Figure 3 and a phonon expansion order of 400 were used to eliminate the need to resort to the short collision time approximation. The total, elastic, inelastic scattering cross sections for H(BeH_2) and Be(BeH_2) at 293.6 K are provided in Figure 4. The thermal neutron scattering cross section for BeH_2 does not exhibit the oscillatory behavior found in metal hydrides like ZrH_x, ZrH_2, and YH_2 [16,17]. This is because the atomic masses of H and Be are similar and the three broad bands in the more complex BeH_2 crystal structure combine in a manner that suppresses the emergence of distinct multi-phonon peaks within the H(BeH_2) TSL.



(a) (b)

Figure 4. Total, Elastic, and Inelastic Scattering Cross Sections for the (a) $H(BeH_2)$ and (b) $Be(BeH_2)$ TSL Evaluations at 293.6 K.

4 CRITICAL MASS CALCULATIONS

Critical mass calculations for bare and reflected spherical configurations were performed using MC21 [8]. All MC21 calculations were performed with 10-50 inactive generations (the number of inactive generations was increased for the larger radius assemblies with a higher dominance ratio), 200 active generations, 10^6 neutrons/generation, for a total of 200×10^6 active neutron histories and Monte Carlo 1-σ statistical uncertainties on k_{eff} calculations on the order of 5-7 pcm. Cross sections were based on ENDF/B-VIII.0 [18] and were processed by the NDEX nuclear data processing code [19]. The core of the sphere was assumed to be a homogenized mixture of highly-enriched uranium (HEU) and a moderator at a variety of ^{235}U loading densities. Moderation by water (H_2O), polyethylene (CH_2), and BeH_2 were studied. Bare; 0.20 cm thick H_2O, CH_2, BeH_2, and beryllium oxide (BeO) reflected; and 0.40 m thick BeO-reflected configurations were considered. ENDF/B-VIII.0 TSL evaluations were used when modeling H_2O, CH_2, and BeO. The TSL evaluation described in Section 3 was used for BeH_2. Critical radius searches were performed by a Python script that managed the MC21 calculations and converged k_{eff} to within $\pm10^{-4}$ Δk. Table III provides the number densities for the HEU, moderator, and reflector materials used in these calculations. Moderator compositions were based on the density of light water at 293.6 K and 0.101325 MPa (1 atmosphere) from IAPWS-95 [20] and the typical mass density of high-density polyethylene. BeH_2 has a H density that is 24% higher than H_2O and 0.03% higher than CH_2. Homogenized HEU and moderator mixtures were prepared by appropriately volume weighting the base materials to achieve the desired ^{235}U loading density.

Figure 5 shows the estimated critical mass for bare H_2O, CH_2, and BeH_2-moderated spheres as a function of ^{235}U loading density. The minimum critical mass of the bare BeH_2-moderated sphere is 0.796 kg ^{235}U and is substantially lower than the H_2O (1.469 kg ^{235}U) and CH_2 (0.931 kg ^{235}U) moderated spheres. Figure 6 provides the estimated critical mass for BeH_2-moderated bare and reflected spheres as a function of fissile loading density. Reflectors studied include 0.2 m thick H_2O, CH_2, BeH_2, BeO, and 0.4 m thick BeO. BeO reflection produces consistently lower critical masses for the BeH_2-moderated spheres. The minimum critical mass of the 0.2 m and 0.4 m thick BeO-reflected spheres were 0.207 kg ^{235}U and 0.178 kg ^{235}U, respectively. These estimated critical masses are substantially lower than the 0.405 ± 0.005 kg ^{235}U critical mass achieved for the Proserpine experiment (HEU-SOL-THERM-046) [21], the HEU-fueled critical assembly with the lowest critical mass in the ICSBEP Handbook.

The calculated minimum critical mass for bare and reflected spheres moderated by H_2O, CH_2, and BeH_2 are compared in Table IV. The BeH_2 moderated spheres have the lowest minimum critical mass of these moderators. Moreover, thick BeO-reflected and BeH_2-moderated spherical assemblies have a critical mass lower than any other moderated system we have studied to date.

Table III. Composition of Fuel, Moderator and Reflector Materials

Density (kg/m^3)	**Material**				
	HEU	**H_2O**	**CH_2**	**BeH_2**	**BeO**
	17534	998.11	960	775	3010
Nuclide	**Number Densities (10^{30} nuclei/m^3)**				
^{1}H		6.67297E-2	8.24329E-2	8.24572E-2	
^{9}Be				4.12286E-2	7.24730E-2
^{12}C			4.07754E-2		
^{13}C			4.41016E-4		
^{16}O		3.32838E-2			7.24730E-2
^{17}O		1.26786E-5			
^{18}O		6.83980E-5			
^{234}U	5.08565E-4				
^{235}U	4.49245E-2				
^{236}U	1.92094E-4				
^{238}U	2.57142E-3				

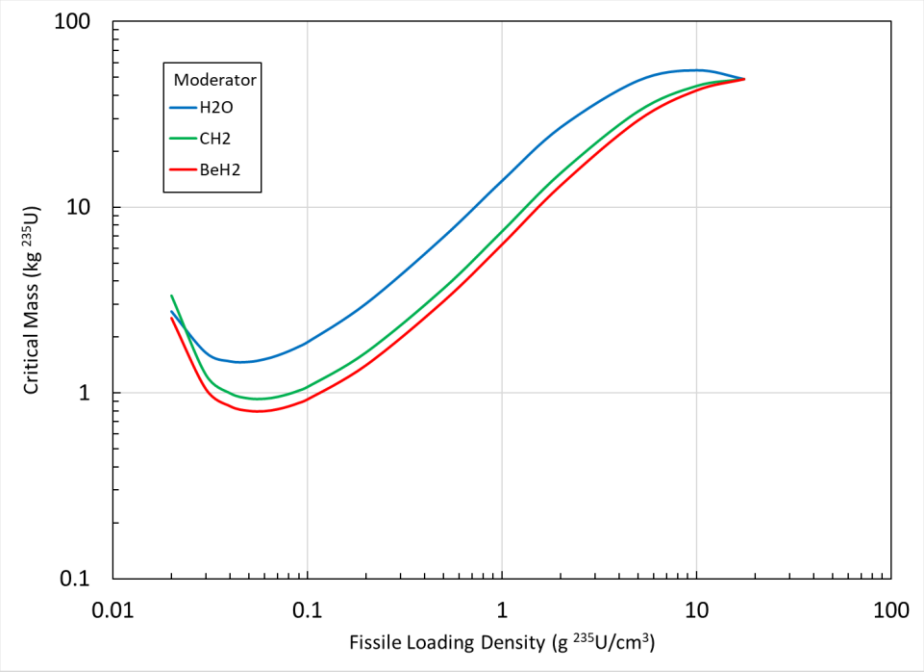


Figure 5. Calculated ^{235}U Critical Mass for H_2O, CH_2, and BeH_2-Moderated Bare Spheres.

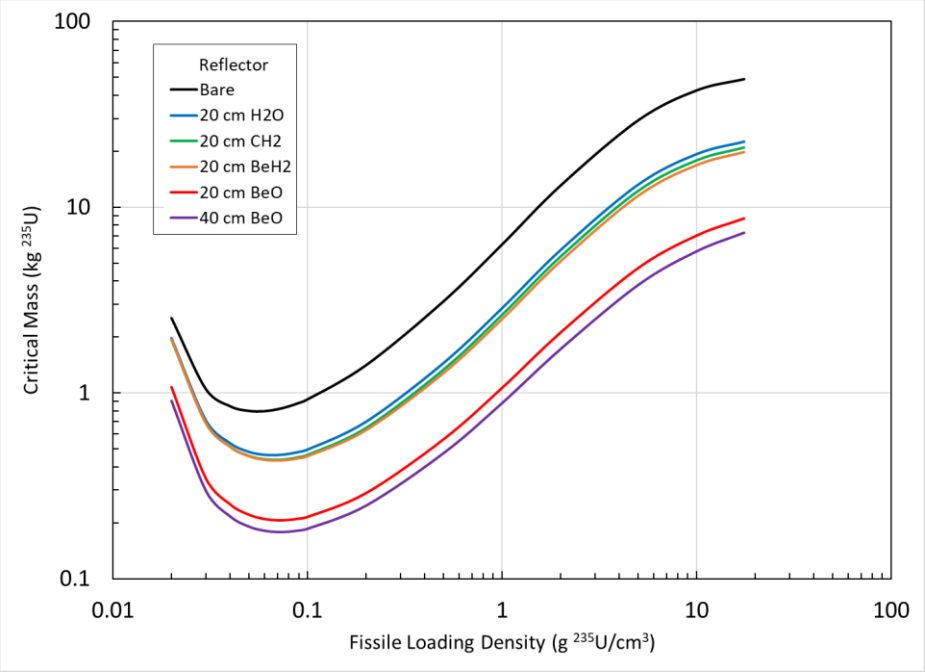


Figure 6. Critical Mass for BeH_2-Moderated Bare and Reflected Spheres. Reflectors include 0.2 m thick H_2O, CH_2, BeH_2, BeO and 0.4 m thick BeO.

Table IV. Comparison of Calculated Minimum Critical Mass (kg ^{235}U) for Bare and Reflected H_2O, CH_2, and BeH_2-Moderated Spheres

Core Moderator	Bare Critical Radius (m)	Bare Critical Mass (kg ^{235}U)	Reflector Materials				
			H_2O	CH_2	BeH_2	BeO	BeO
			0.20 m thick				0.40 m thick
H_2O	0.19144	1.469	0.810	0.766	0.754	0.296	0.244
CH_2	0.15475	0.931	0.551	0.523	0.518	0.235	0.201
BeH_2	0.14688	0.796	0.462	0.437	0.433	0.207	0.178

5 CONCLUSIONS

The TSL for BeH_2 has been evaluated in the incoherent approximation using FLASSH based on phonon DOS calculated using the *ab initio* lattice dynamics approach. The TSL for BeH_2 was then used to increase the fidelity of the thermal scattering physics in MC21 critical mass calculations of BeH_2 moderated spherical systems. The minimum critical mass of BeH_2 moderated sphere surrounded by 0.2 m and 0.4 m thick BeO reflectors was calculated to be 0.207 kg ^{235}U and 0.178 kg ^{235}U, respectively. Thick BeO-reflected and BeH_2-moderated spherical assemblies have a critical mass lower than any other moderated systems we have studied to date.

6 ACKNOWLEDGMENTS

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