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Nuclear Requirements for Control Materials

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ABSTRACT*

Nuclear factors that affect the effectiveness of control rod materials for power reactors with a thermal or near thermal neutron spectrum are described in this memorandum. The principal elements and isotopes which qualify as neutron absorbers are described and physical requirements, such as, rod thickness, surface density, and weight ratios are compared. The importance of epithermal absorption and the nuclear effects of burnup as a result of long neutron exposure in power reactors are shown. The characteristics of absorption isotope chains, such as, Europium are described, and finally, the deleterious effects of cladding and scattering cross section of poison diluents on control effectiveness are shown.

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*The general survey contained in this Memorandum served as the basis of a paper entitled "Nuclear Requirements for Control Rod Materials", presented by the writer at a symposium on reactor control materials. This symposium was part of the Second Winter Meeting of the American Nuclear Society held at New York City, October 28-31, 1957. The technical information was obtained from several Knolls Atomic Power Laboratory reports.

Nuclear Requirements for Control Materials

Neutron absorbing materials are used to control reactors primarily of a thermal or near thermal neutron spectrum. The first consideration in selecting a control material is its absorption cross section, because a rod must be adequately black to thermal neutrons without being unduly large. To determine whether a material has an adequate cross section, the criterion, $N\sigma t$, is used, where N is the number of atoms/cm^3 in the full density material, σ is its average microscopic thermal absorption cross section for a Maxwellian neutron energy distribution, and t is the thickness of the control rod blade. If a poison does not have a large $N\sigma$, either because its theoretical density is too low or because its concentration in a matrix material is metallurgically restricted, it may be made nuclearly acceptable either by using a thicker control blade or by increasing the control rod surface. Figure 1 shows the increase in control rod surface which would be required if $N\sigma t < \infty$. The curve in Figure 1 is actually dependent on L/λ of the medium external to the rod where L is the diffusion length and λ is the transport mean free path. However, this is not a critical parameter and a representative value of $L/\lambda = 2$ has been chosen. A value of $N\sigma t = 2$ is usually selected as a compromise between excess thickness of rod and excess control rod surface. Hereafter, for convenience, the criterion $N\sigma t = 2$ will be called the blackness criterion. On this basis, the amount of poison required for thermal blackness is given in column eight of Table I for several neutron poisons.

Another consideration in selecting control rod materials is the burnup or loss of poison atoms caused by neutron exposure in long-endurance, high-power reactors. During power operation, the control rods are normally inserted part way into the high neutron flux of the core, particularly if reactivity transients associated with the fission product, xenon, are provided. For example, during the lifetime of an enriched uranium core, about 200 moles of neutrons are produced in fission in operating 4000 hours at a power of 100 megawatts. It is estimated that about 10% or 20 moles of these neutrons would be absorbed in the control rods. Absorption of neutrons in the rods generally decreases their blackness, so that if the rods are to meet the blackness criterion after 4,000 hours, exta poison must be added at the start. Ideally, this extra poison would be added non-uniformly, because the neutron absorption in the rods is non-uniform. On this <u>ideal</u> basis, the <u>minimum</u> extra poison required for burnup is given in column nine of Table I.(1) The amount of extra poison required has been computed on the assumption that 20 moles of the absorbing nuclide are destroyed during burnup. The isotopic abundance of the absorbing nuclide has been considered in computing this extra poison, but the absorption cross section of all daughters is assumed to be zero. Europium, which has several important absorbing daughters, is discussed in some detail later in this general summary.

For ease of fabrication, it is usually desirable to have the poison material uniformly distributed in the rods. In this case, the minimum quantities of materials listed in Table I are no longer adequate since non-uniform burnup would destroy the effectiveness of certain portions of the rods. The maximum amount of extra poison required for burnup is about six times the average burnup. Rods designed with the maximum amount of poison would not lose any of their effectiveness during the life of the reactor core.

A number of the absorbers given in Table I have such low cross sections that the amount required for blackness is considerably greater than the minimum 20 moles required for burnup. In these cases non-uniform burnup is a minor problem. A few examples of such poisons would be rhodium, silver, indium, hafnium, rhenium, and gold. For these poisons, the total amount listed in Table I would be sufficient if spread uniformly throughout the rods. Note, however, that the silver rod would have to be about 1/4" thick to be black. Rhodium, on the other hand, could be clad or alloyed if necessary. Iridium presents the same possibilities as rhodium, though subject to more burnup.

The boron-10 isotope, with an averaged thermal cross section of 3,470 barns, is not required in large amounts for rod blackness. Therefore, with initially uniform poison the problem of non-uniform burnup is quite serious and the amount of boron required to prevent loss of control is about twice the minimum total amount listed in Table I. In the case of higher cross-section materials such as cadmium or gadolinium, the amount listed in Table I would have to be considerably more than doubled. For absorbing nuclides with cross sections between Hf^{177} and B^{10}, the amount listed in Table I would have to be increased by a factor between one and two.

Currently, europium and gadolinium are of considerable interest as possible control poisons. Europium is of special interest as it has five adjacent absorbing isotopes which prevent the loss of control caused by burnup. Only two of these isotopes, the first and third in the chain, occur in natural europium. Natural gadolinium has two absorbing isotopes of very high cross section and five other isotopes of small or unknown cross section. The two highly absorbing isotopes, Gd^{155} and Gd^{157}, are apparently not linked by a strong absorber and hence, do not form a chain such as is found in europium. Consequently, gadolinium loses its effectiveness as a poison much more rapidly than europium does, therefore, it is not as suitable for the high burnup regions of the rods. However, gadolinium is considerably cheaper than europium and can be used in reasonable concentrations in the low burnup portions of control rods.

Natural europium consists of 47.8% Eu^{151} and 52.2% Eu^{153}. The cross sections of these isotopes and the three additional highly absorbing isotopes are:

Eu^{151}	9000 ± 3000 barns
Eu^{152}(13 year)	5500 ± 1500 barns
Eu^{153}	420 ± 100 barns
Eu^{154}	1500 ± 400 barns
Eu^{155}	14,500 ± 4000 barns

An absorption in Eu^{151} may result in either of two isomers of Eu^{152}. About 16% of the time a fairly short-lived isomer is formed which decays to a low cross-section isotope of gadolinium. The remaining 84% of the absorptions result in the 13-year Eu^{152}. The differential equations linking the five isotopes of europium have been solved.(2) The isotopic composition of the natural europium is shown in Figures 2 and 3 as a function of the number of neutrons absorbed per initial europium atom. The relative cross section of natural europium as a function of burnup is shown in figure 4. Two conclusions can be noted from figure 4;

1. The plateau above one neutron absorbed per initial europium atom indicates that an indefinitely long burnup could be tolerated with seven times the amount of natural europium required for thermal neutron blackness;
2. The amount of europium required for 20-mole burnup shown in Table I is high by a factor of two.

However, the most optimistic feature shown in Table I is the amount of poison required for thermal neutron blackness. Even in a well thermalized reactor, the contribution to control effectiveness of epithermal absorptions is significant. To make a control rod black to epithermal neutrons requires considerably more poison. This is illustrated in figure 5 where a 1/16 in. thick cadmium slab is compared to a 0.200 in. thick hafnium slab. Also shown is a typical thermal neutron spectrum. The tail of the spectrum extends far beyond the cadmium cut off at 0.4 ev. The huge resonances of hafnium in the epithermal range capture many epithermal neutrons that cadmium fails to capture. Experimental data taken from 1/16 in. cadmium-covered hafnium slabs of various thicknesses are shown in figure 6.(3) For 0.200 in. thick hafnium, about 40% of the control effectiveness comes from epicadmium captures. Since most poison cross sections decrease rapidly with neutron energy, it is difficult to find a better material than hafnium. Hence, thicker control rods are required or the poison concentrations in the dispersions must be increased. The latter is easily illustrated with boron. In Table I, only 0.15 wt % B^{10} in stainless steel was required to make a 0.200 in. thick slab thermally black. This control slab is compared with hafnium in Figure 7. It is obvious that hafnium is superior. Higher weight per cent of boron increases the epithermal absorption. As shown in Figure 8, critical assembly data indicate that 1.2 wt. % B^{10} in stainless steel is necessary to match the effectiveness of hafnium.(4) This is about eight times the thermal blackness composition shown in Table I. Similar experiments with other poison materials confirm the epithermal effects. For example 18 wt % europium oxide in stainless steel is required to match the effectiveness of hafnium. This is about five times the amount required for thermal blackness described in Table I. Even indium which is thermally black when 0.124 in. thick is only 91% as effective as hafnium when 0.200 in. thick.

Thus, the epithermal absorption requirement dominates the selection of control rod poisons and is far more limiting than either burnup in most cases or the thermal blackness criteria. It is difficult to surpass hafnium in epithermal absorption and high endurance burnup.

As can be seen in Table I, indium appears to be a good control rod material. It has a high thermal blackness and small burnup. However, indium has a low absorption at 5 to 8 ev. as shown in Figure 9. Silver, which has a resonance at 5.5 ev, could be added to form a 50-50 mixture. This combination improves the high energy drop off of indium as shown on the middle graph of Figure 10 but also produces a weak absorption region around 0.3 ev. since the indium is now only one-half as thick. The net result is a loss of effectiveness. Since cadmium is good up to about 0.4 ev, cadmium may be added to form a 33-33-33% mixture. This improves the weak absorption region at 0.3 ev but now impairs the low point at 3.0 ev since the indium and silver are now only two-thirds as thick. A mixture of 71 wt % silver, 10 wt % cadmium, and 18 wt % indium was found to be 5% more effective than 0.200 in. thick hafnium.(4) Thus little improvement can be made by adding poison materials with complementary resonance absorptions. At the present time a mixture of hafnium and EuO_2 with a small addition of silver has been the optimum for 0.200 in. slabs and is about 18% more effective than pure hafnium.

Another practical consideration in the selection of control rod materials is the effect of neutron scattering within the control rod. Often the poison materials themselves have scattering, such as silver where one-tenth of its thermal cross section is scattering. In the dispersions, the diluent, such as, steel or titanium has a large scattering cross section. Further, a cladding is often necessary to avoid corrosion of the poison material. The cladding reflects neutrons before they can reach the poison.

The effect of scattering in selecting cladding materials is illustrated in Figure 11, which shows the fraction of neutrons reflected from a black absorber by a cladding of thickness, t, and cross section, $N\sigma_s$. A 0.100 in. thick steel cladding would reflect 17% of the neutrons whereas 0.040 in. of steel would reflect only 8% of the neutrons. Titanium would be better than steel but nickel would be much poorer.

The effect of scattering within the poison is demonstrated in Figure 12. For example, a thermal absorber such as B^{10} with $\sum_a t = 2$ and no scattering cross section would permit the transmission of 6.0% of the thermal neutrons through the absorber, a loss of effectiveness of 6% over a thick absorber. The addition of a scattering cross section of 20% of the absorption cross section permits a loss of 8.3% of the thermal neutrons, an increase of 2.3% caused by the scattering. This particular example corresponds to the dilution of B^{10} by steel to form a 0.200 in. thick control slab. Of the two forms of scattering, the loss caused by cladding is the more significant. Thus cladding must be kept as thin as corrosion requirements permit.

In summary, the previous discussion indicates that high performance control rod poisons must be selected for blackness over a large range of neutron energies, at least to 10 ev. Burnup is also a prime consideration when high cross-section poisons, above 2000 barns, are selected. Hafnium is difficult to surpass in both respects for 0.200 in. thick control slabs. Mixtures of europium and hafnium or of cadmium, indium, and silver are also promising. The B^{10} isotope would be excellent in high concentrations, about 2 wt % in steel, if high burnup were made feasible. By diluting high cross-section poisons with a scattering material is permissible, but claddings should be carefully selected with low scattering cross section and maintained as thin as corrosion permits.

REFERENCES

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(2) KAPL-M-PB-1, by A.P. Bray, May 6, 1957, KAPL (not released).

(3) KAPL-M-ATR-11, by R.G. Luce, May 4, 1956, KAPL (not released).

(4) SFR-Ph-269, by R.R. Eggleston, July 27, 1956, Westinghouse (not released).

TABLE 1

Element or Nuclide	Average Thermal Cross Section (Barns)	Abundance of Important Isotopes (%)	Surface Density for Blackness (gm/cm²)	Density (gm/cm³)	Thickness (cm)	Thickness (Inches)	Amount† for Blackness (Kg)	Amount for 20 Mols. Burnup (Kg)	Total Amount (Kg)	Total Thickness Inches	Useful as Single Material	Useful as Dispersion	Weight Ratio in 200" Stainless Steel Slab (%)
Li	[illegible]		.362	.53	.68	.270	13.7	1.6	15.3	.300			
Li-6	[illegible]	7.52	.024	.53	.045	.018	.9	.12	1.02	.020		x	.67
B	[illegible]		.054	2.3	.023	.009	2.05	1.06	3.11	.014		x	1.39
B-10	3,470	18.8	.0093	2.3	.004	.0016	.35	.20	.55	.0025		x	.23
Rh	[illegible]		2.56	12.5	.20	.080	97.1	2.0	99.1	.082		x	52.
Ag	[illegible]		6.47	10.5	.62	.240	245.	2.2	247.	.242			
Ag-107	[illegible]	51.4											
Ag-109	[illegible]	48.6											
Cd	[illegible]		.165	8.6	.0192	.0077	6.24	18.4	24.6	.030		x	4.4
Cd-113	[illegible]	12.3											
In	[illegible]		2.26	7.3	.310	.124	85.1	2.3	87.4	.128	x		
Sm	[illegible]		.100	7.2*	.0139	.0055	3.85	21.5	25.3	.036		x	3.0
Sm-149	57,200	13.8											
Eu	[illegible]		.128	7.3*	.0175	.0070	4.66	6.36	11.0	.017		x	3.4
Eu-151	[illegible]	47.8											
Gd	[illegible]		.0128	7.6*	.0017	.00068	.48	10.2	10.7	.0151		x	.35
Gd-155	[illegible]	14.7											
Gd-157	[illegible]	15.7											
Dy			.55	8.1*	.066	.027	20.8	11.6	32.4	.042		x	14.8
Dy-164	[illegible]	28.2											
Er	[illegible]		3.78	8.6*	.44	.176	143.	?	?	?	?		
Lu			6.05	?	~.67	.270	229.	135.	364.	.430			
Lu-176	[illegible]	2.6											
Hf	91		6.38	13.4	.471	.188	241.	11.4	252.	.197	x		
Hf-177	[illegible]	18.4											
Re	73		8.28	20.	.414	.166	314.	3.	317.	.167	x		
Ir	372		1.68	22.4	.075	.030	63.4	10.	73.4	.035		x	34.
Ir-191	820	38.5											
Ir-193	104	61.5											
Au	85		7.52	19.3	.392	.157	284.	4.	288.	.159	x		
Hg	330		1.97	14.22	.138	.0552	74.5	23.8	98.3	.073		x	14.
Hg-199	2,160	16.8											

* X-Ray density for oxide

† 6×10^5 in^2 of control slab surface (one side)

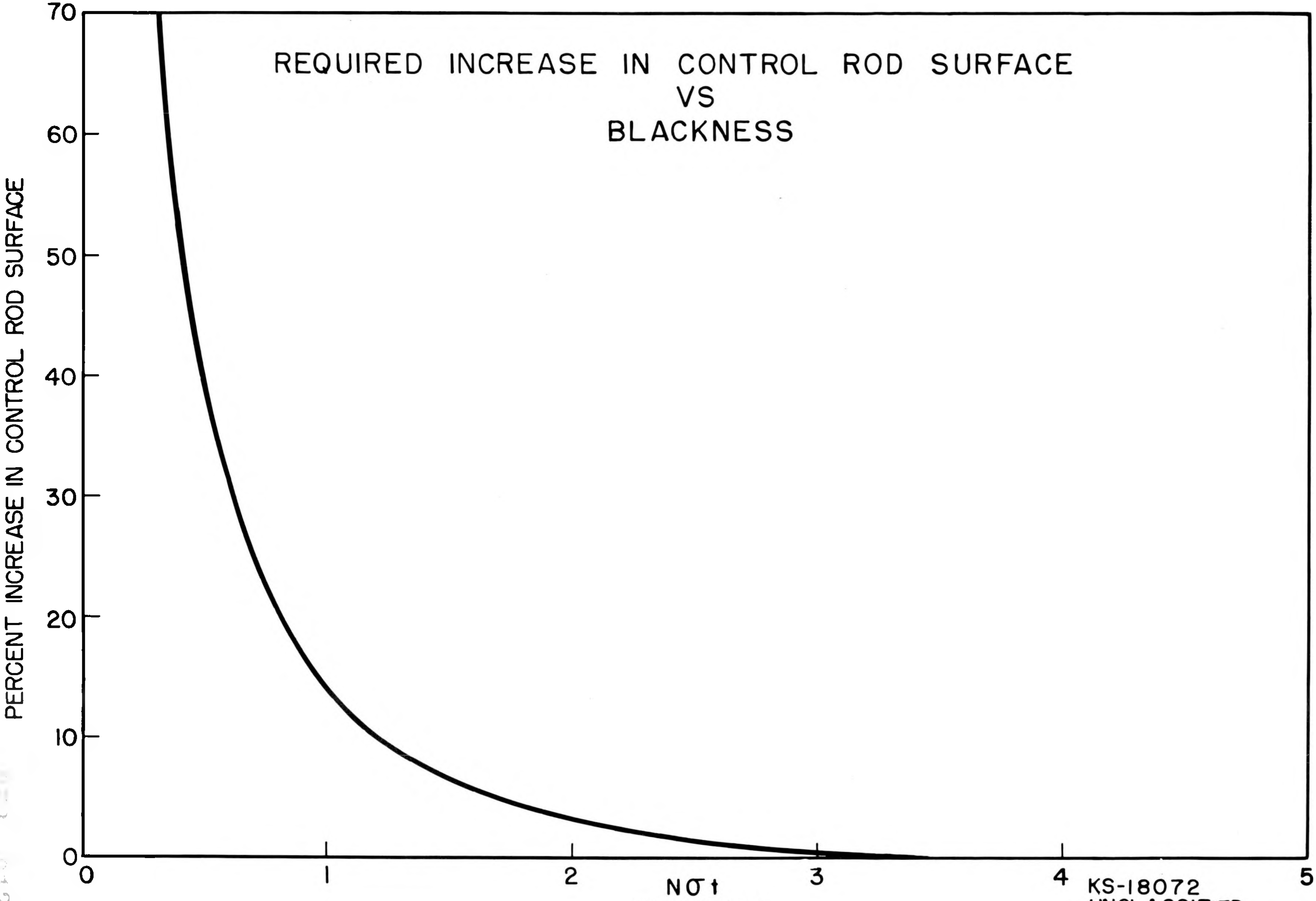
REQUIRED INCREASE IN CONTROL ROD SURFACE
VS
BLACKNESS

FIGURE I

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VARIATION OF ISOTOPES OF EUROPIUM CHAIN WITH TOTAL NEUTRON ABSORPTIONS
PART I

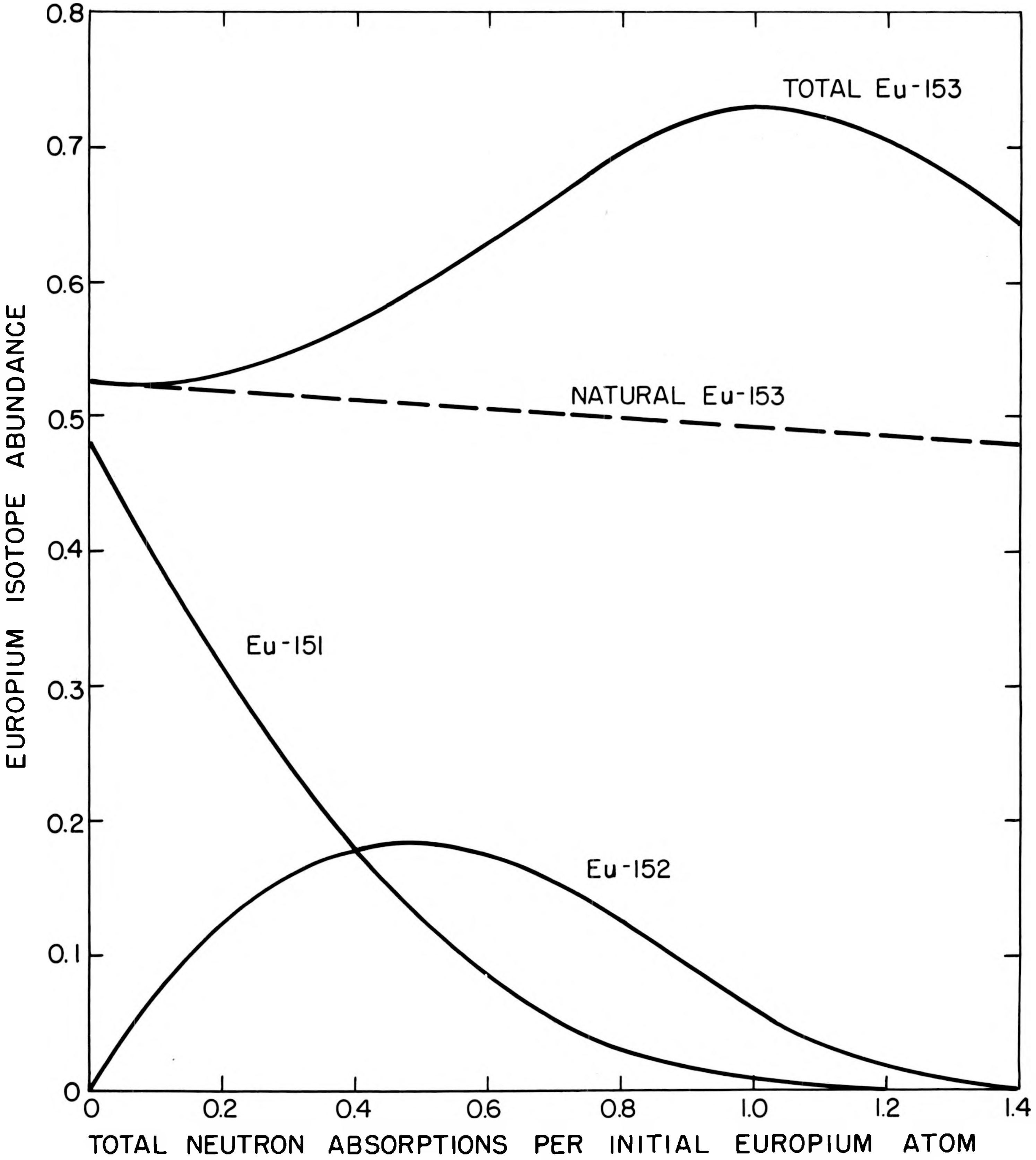

FIGURE 2

KS-18073
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VARIATION OF ISOTOPES OF EUROPIUM CHAIN
WITH TOTAL NEUTRON ABSORPTIONS
PART II

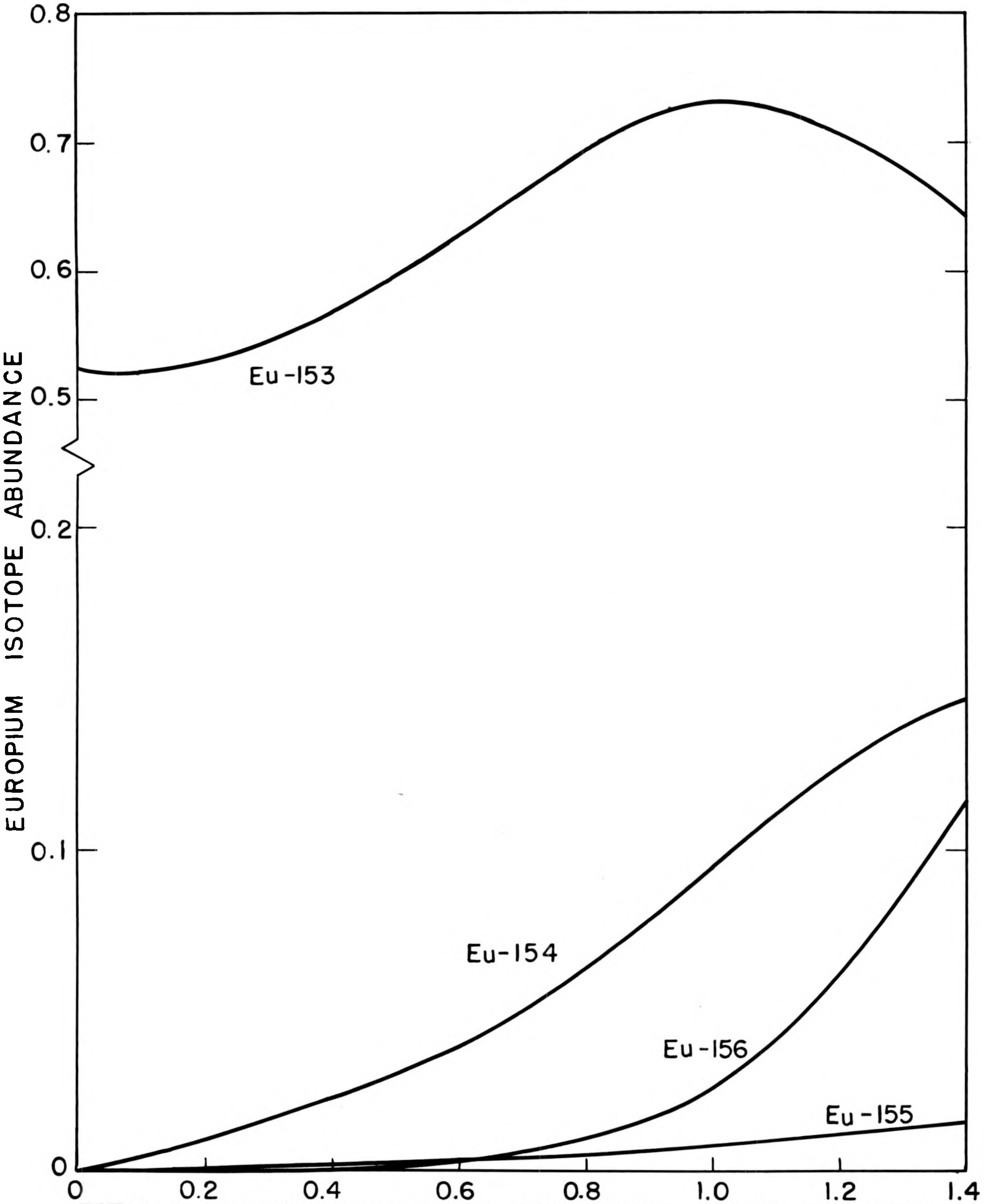

FIGURE 3

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VARIATION OF CROSS SECTION OF NATURAL EUROPIUM
WITH TOTAL NEUTRONS ABSORBED

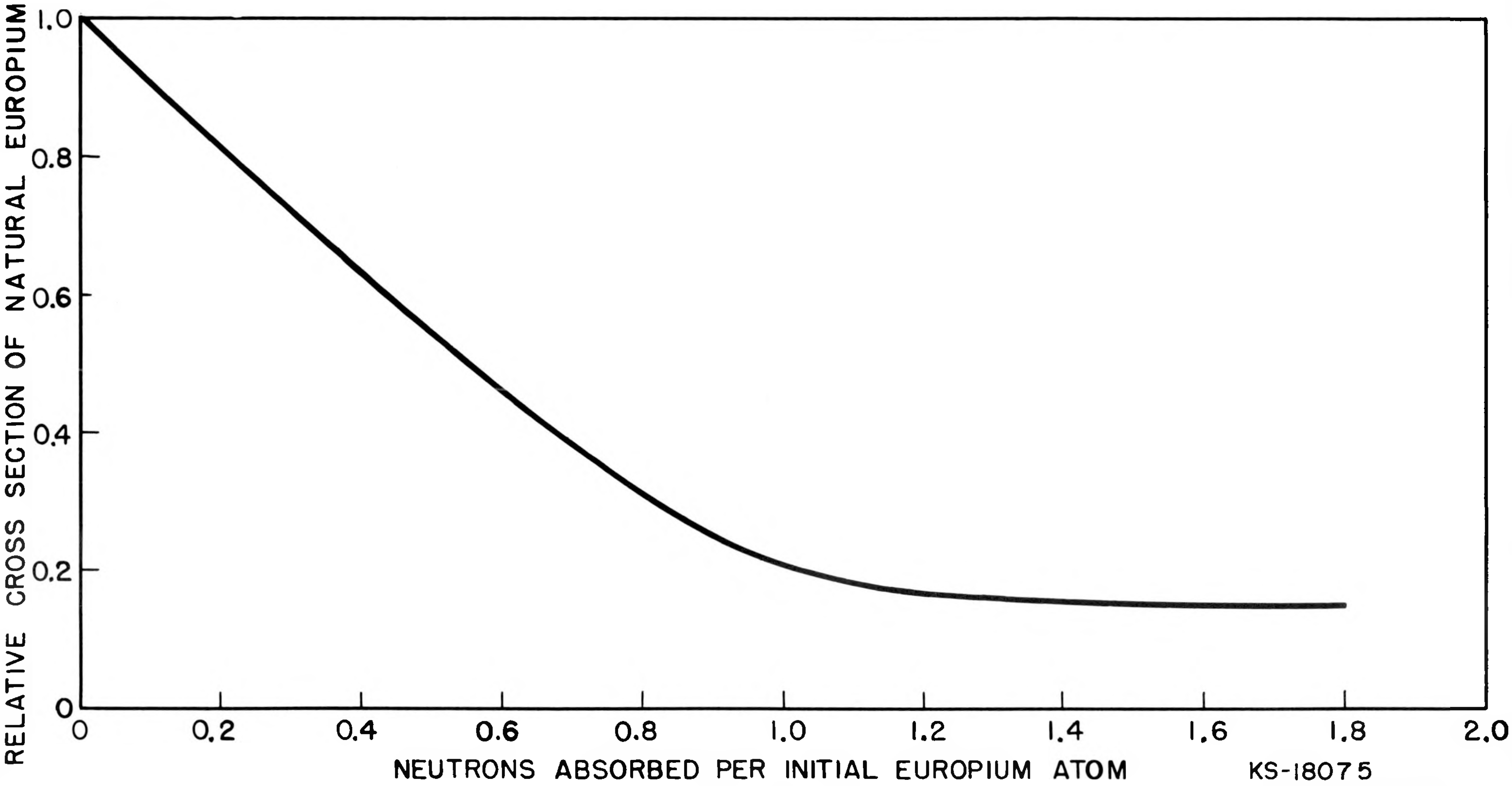

FIGURE 4

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COMPARISON OF HAFNIUM AND CADMIUM ABSORPTIONS

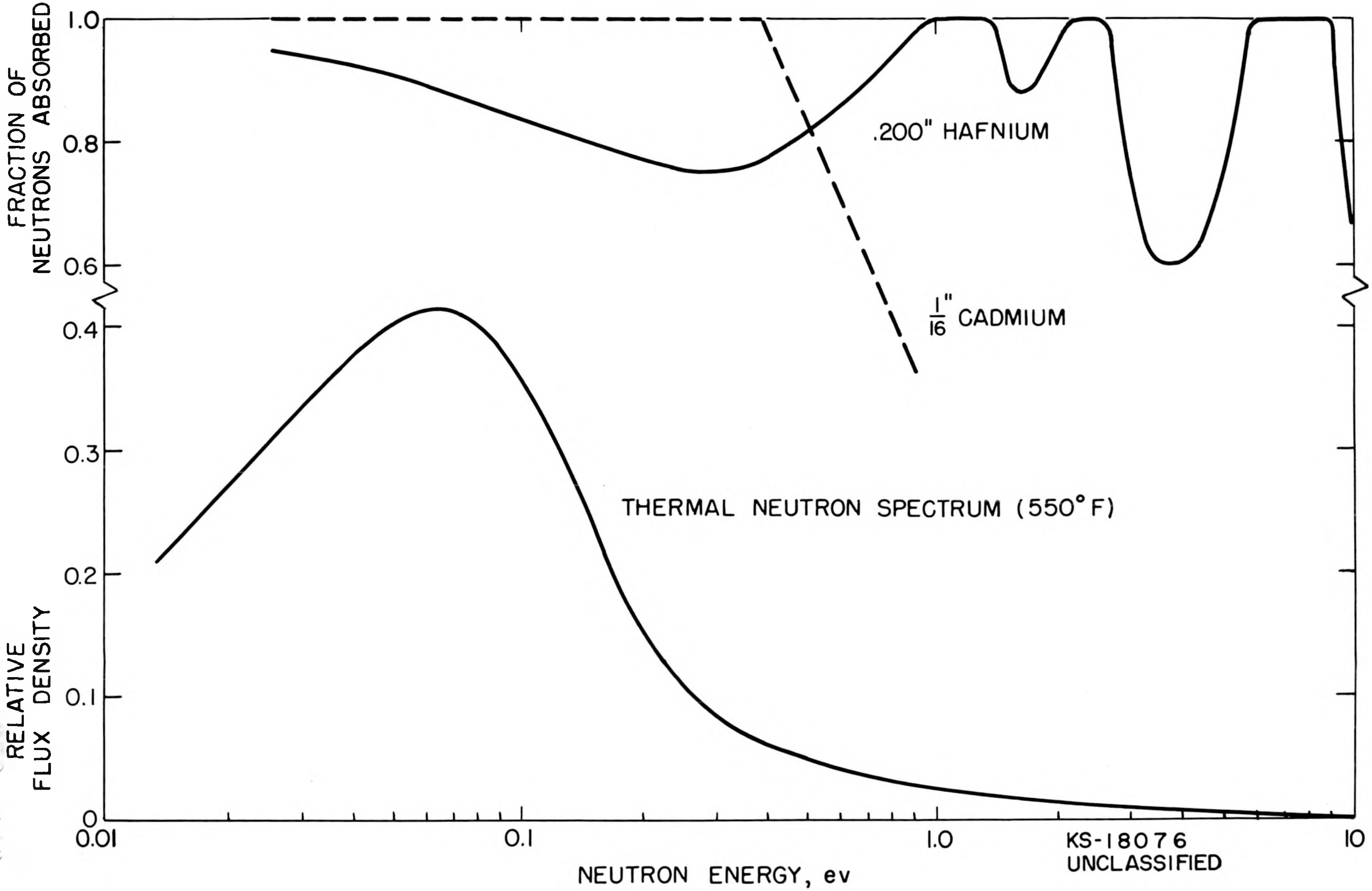

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FIGURE 5

COMPARISON OF SUB-CADMIUM AND EPI-CADMIUM EFFECTIVENESS OF HAFNIUM FROM CRITICAL ASSEMBLY EXPERIMENTS

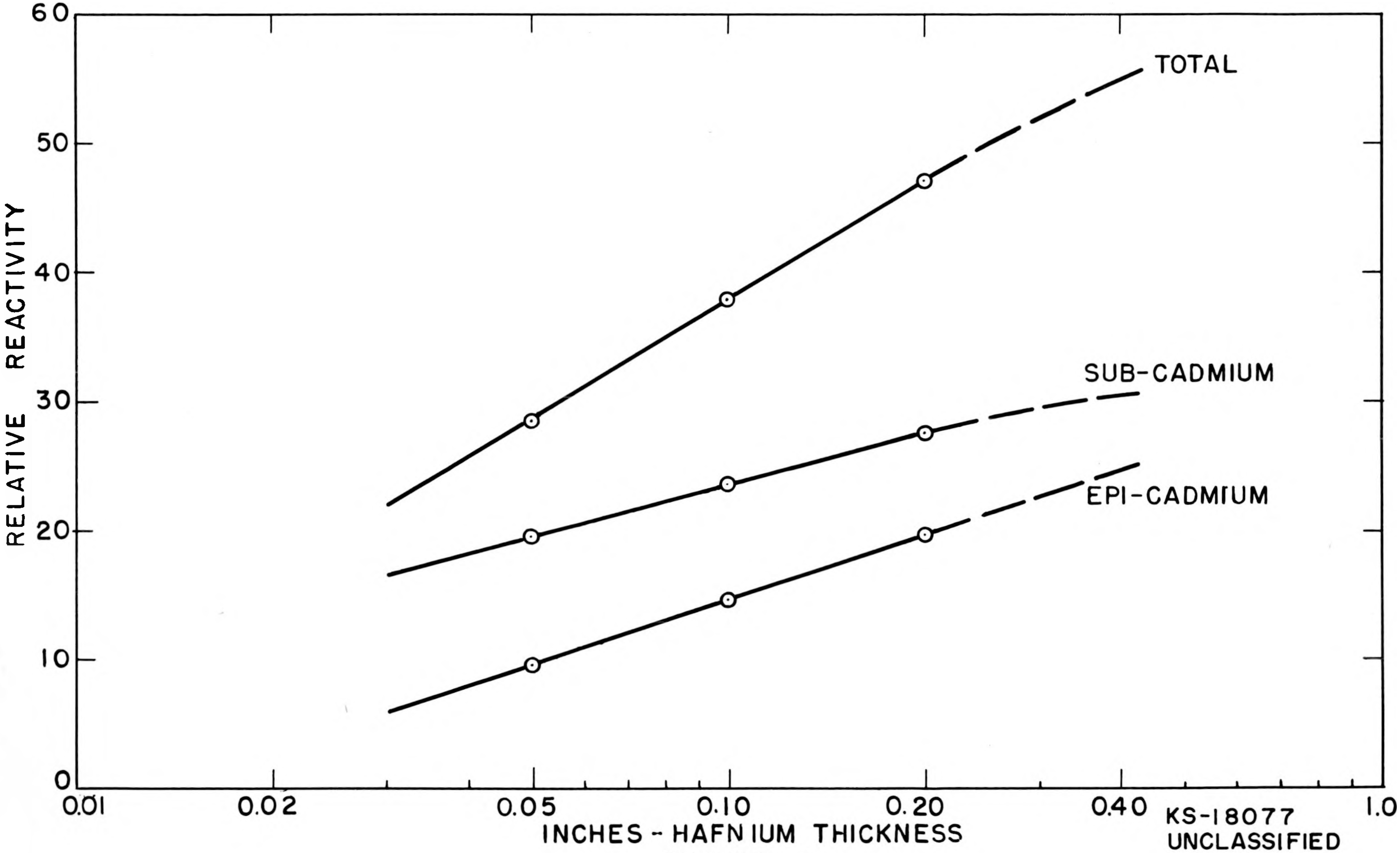

FIGURE 6

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COMPARISON OF HAFNIUM AND BORON-10 ABSORPTIONS

.200" THICK SLABS

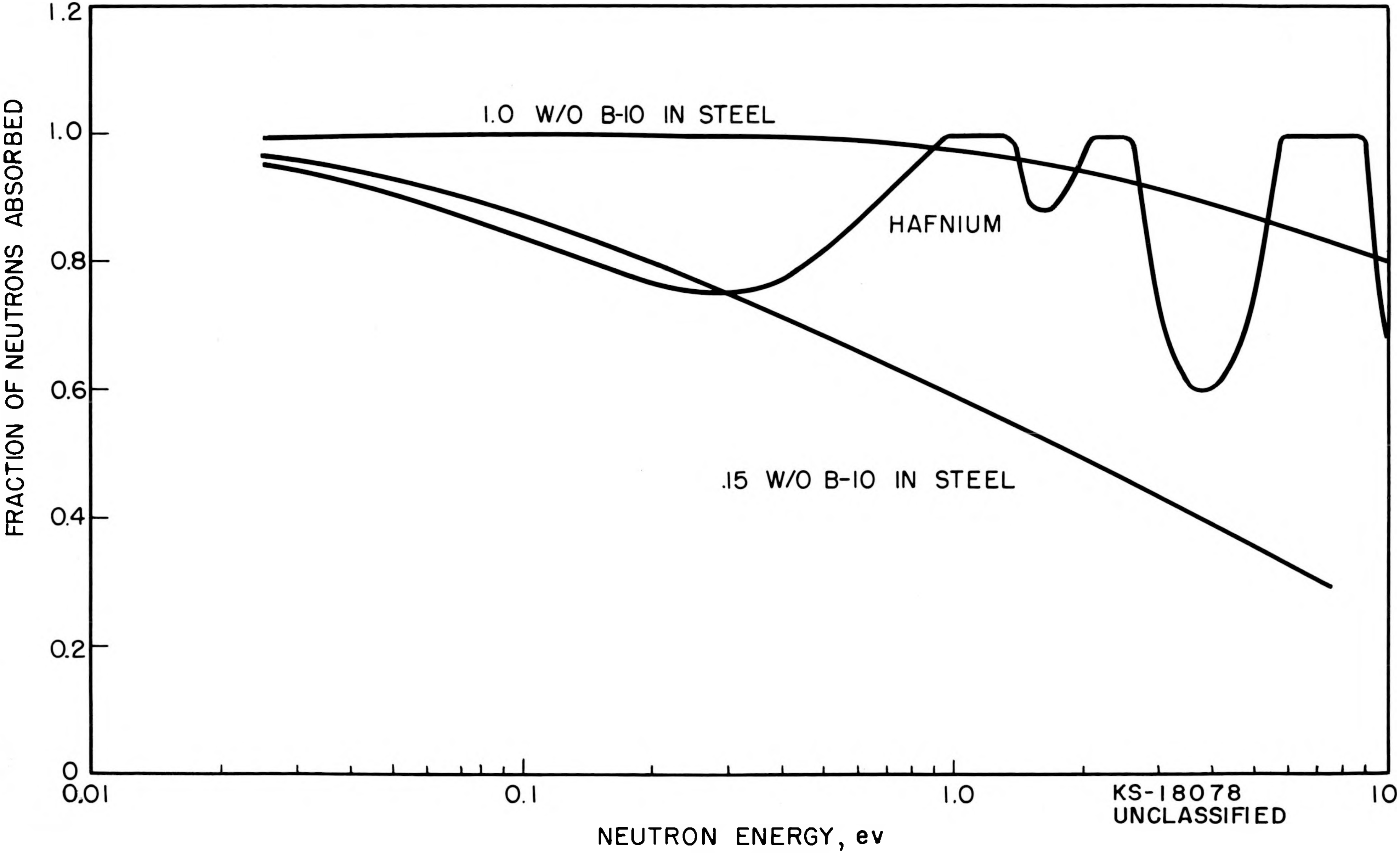

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FIGURE 7

COMPARISON OF HAFNIUM AND BORON-10 EFFECTIVENESS FROM CRITICAL ASSEMBLY EXPERIMENTS

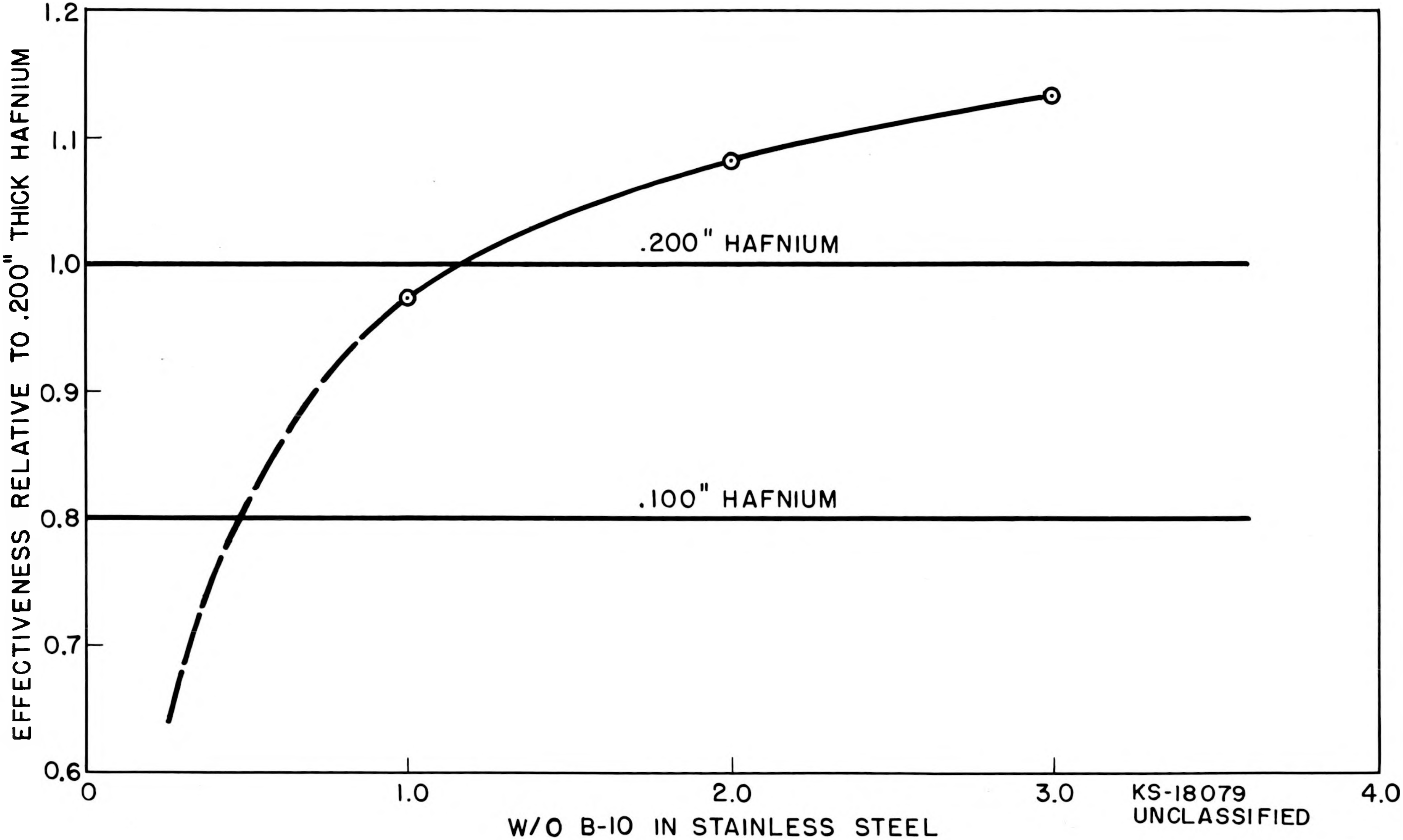

FIGURE 8

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COMPARISON OF CADMIUM, INDIUM, SILVER ABSORPTIONS

.200 THICK SLABS

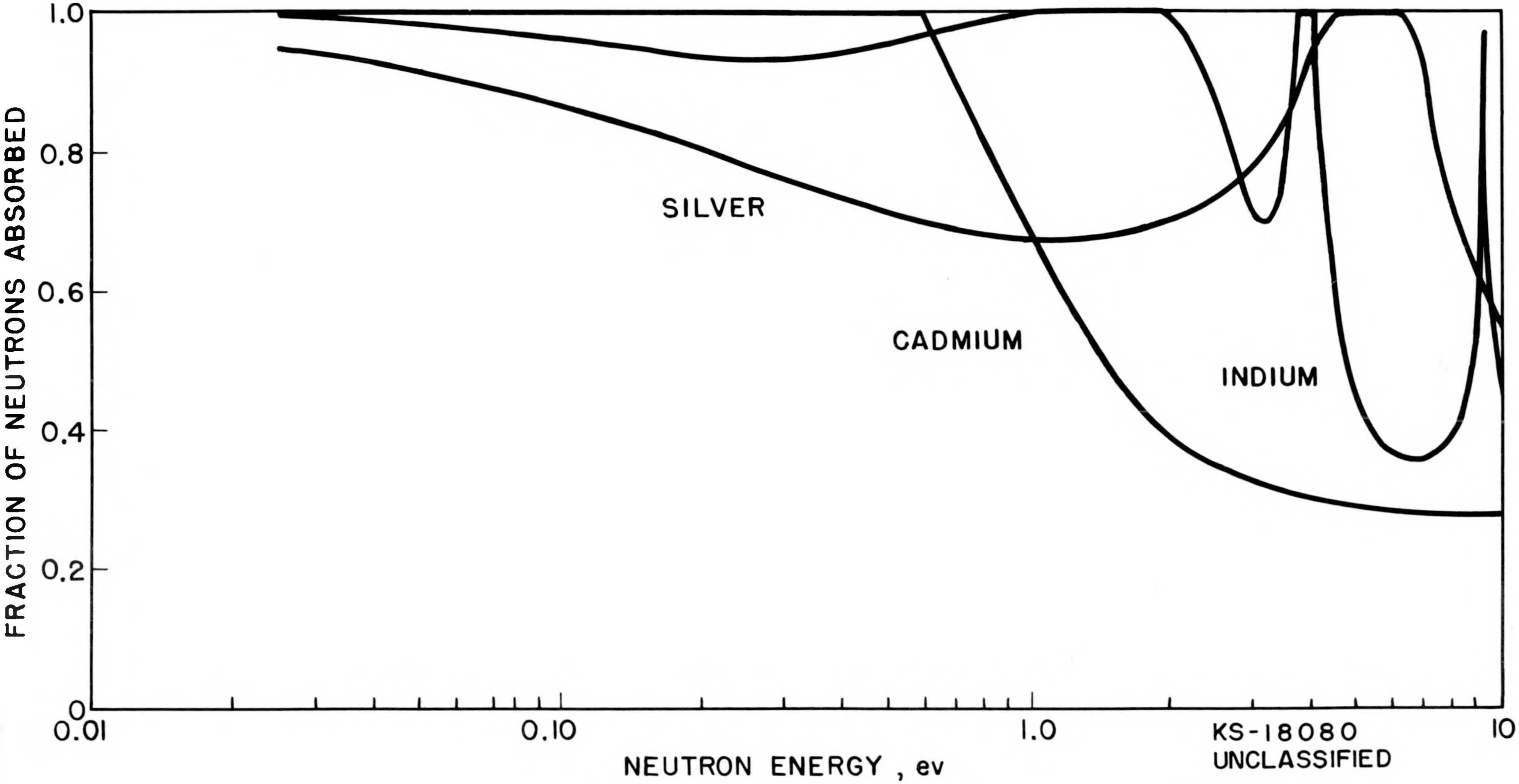

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FIGURE 9

MIXING ABSORBING MATERIALS WITH COMPLEMENTARY RESONANCES
.200" THICK SLABS

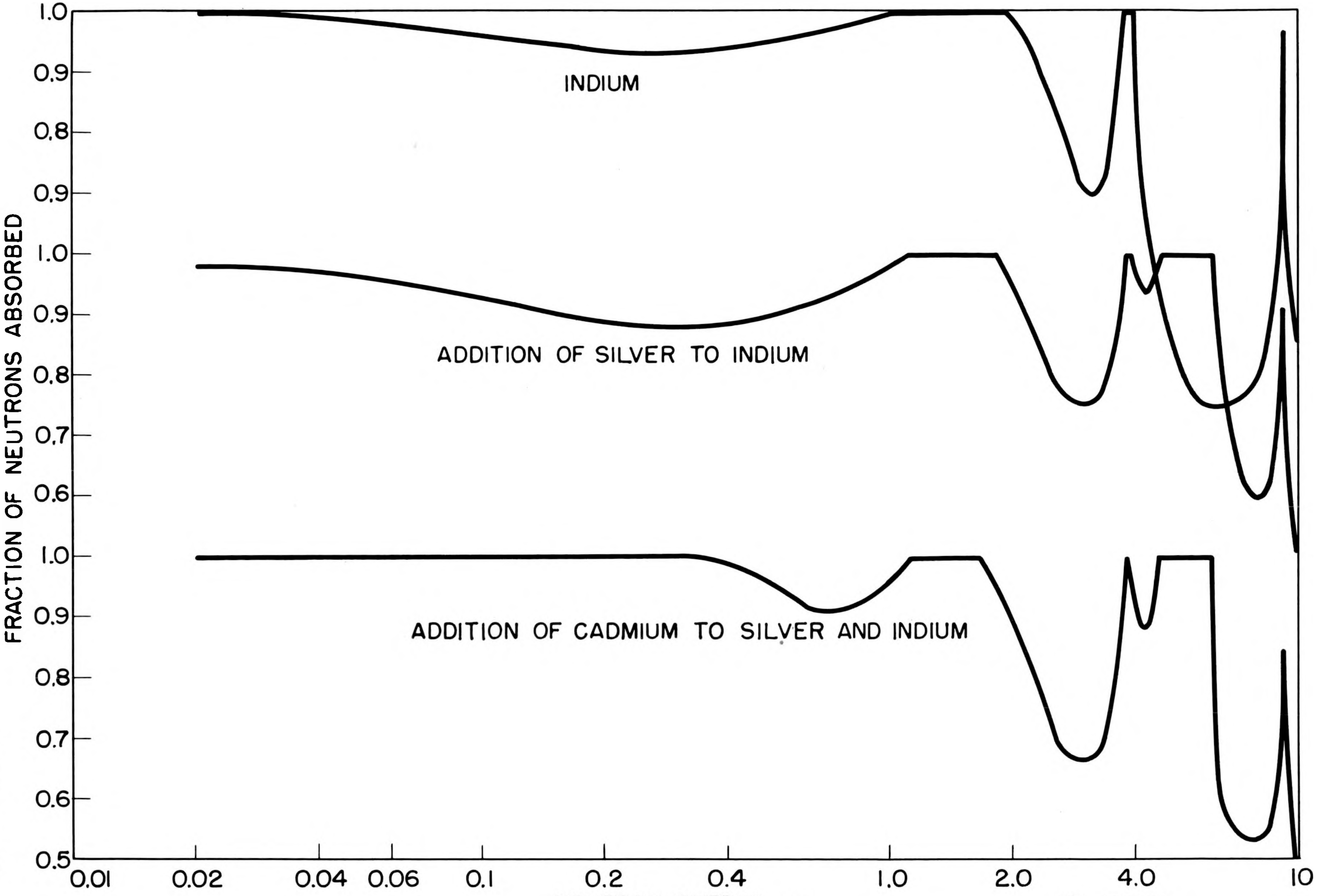

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FIGURE 10

LOSS OF EFFECTIVENESS BY NEUTRON SCATTERING IN THE CLADDING

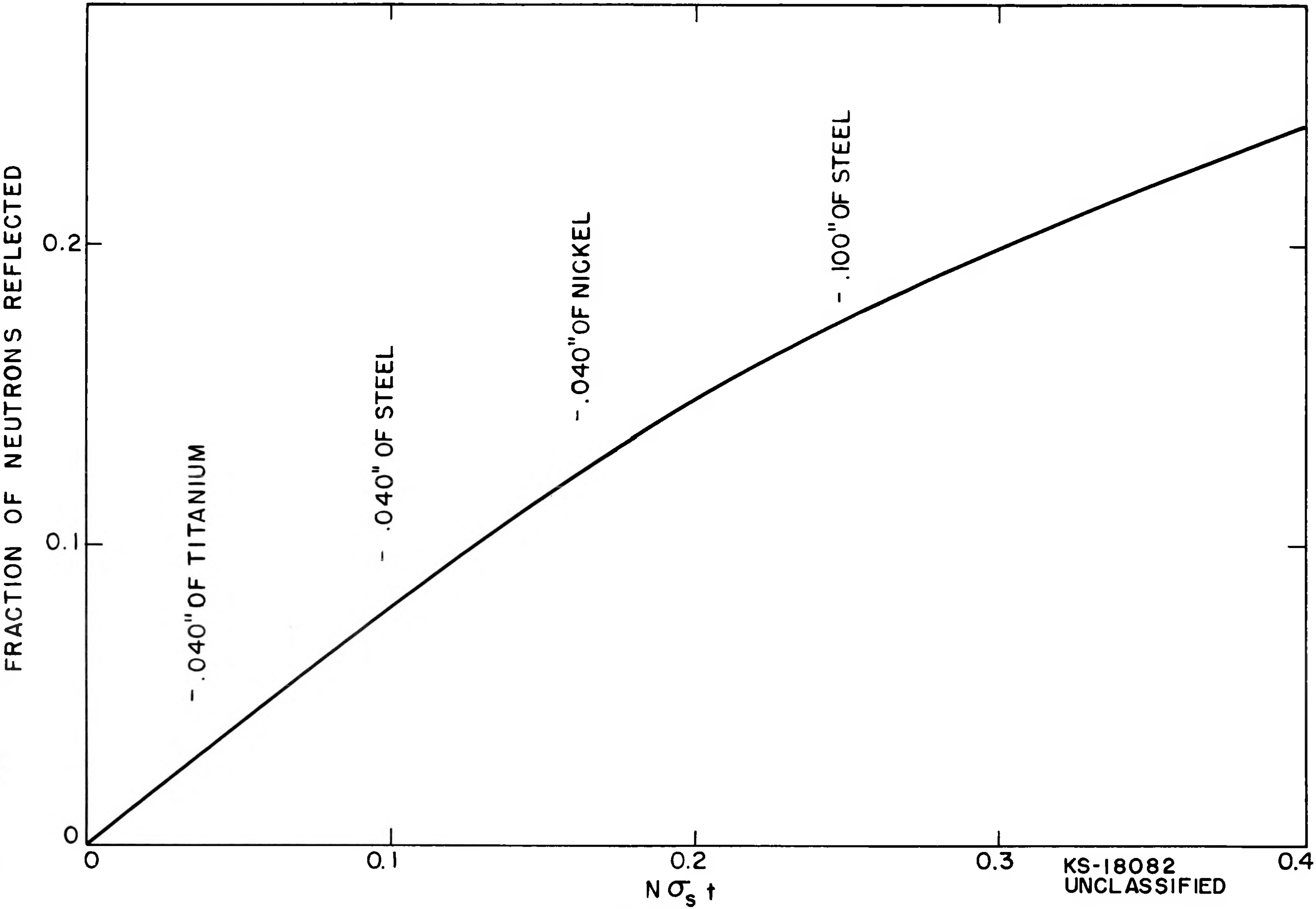

FIGURE 11

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LOSS OF EFFECTIVENESS DUE TO INTERNAL SCATTERING IN ABSORBER SLABS

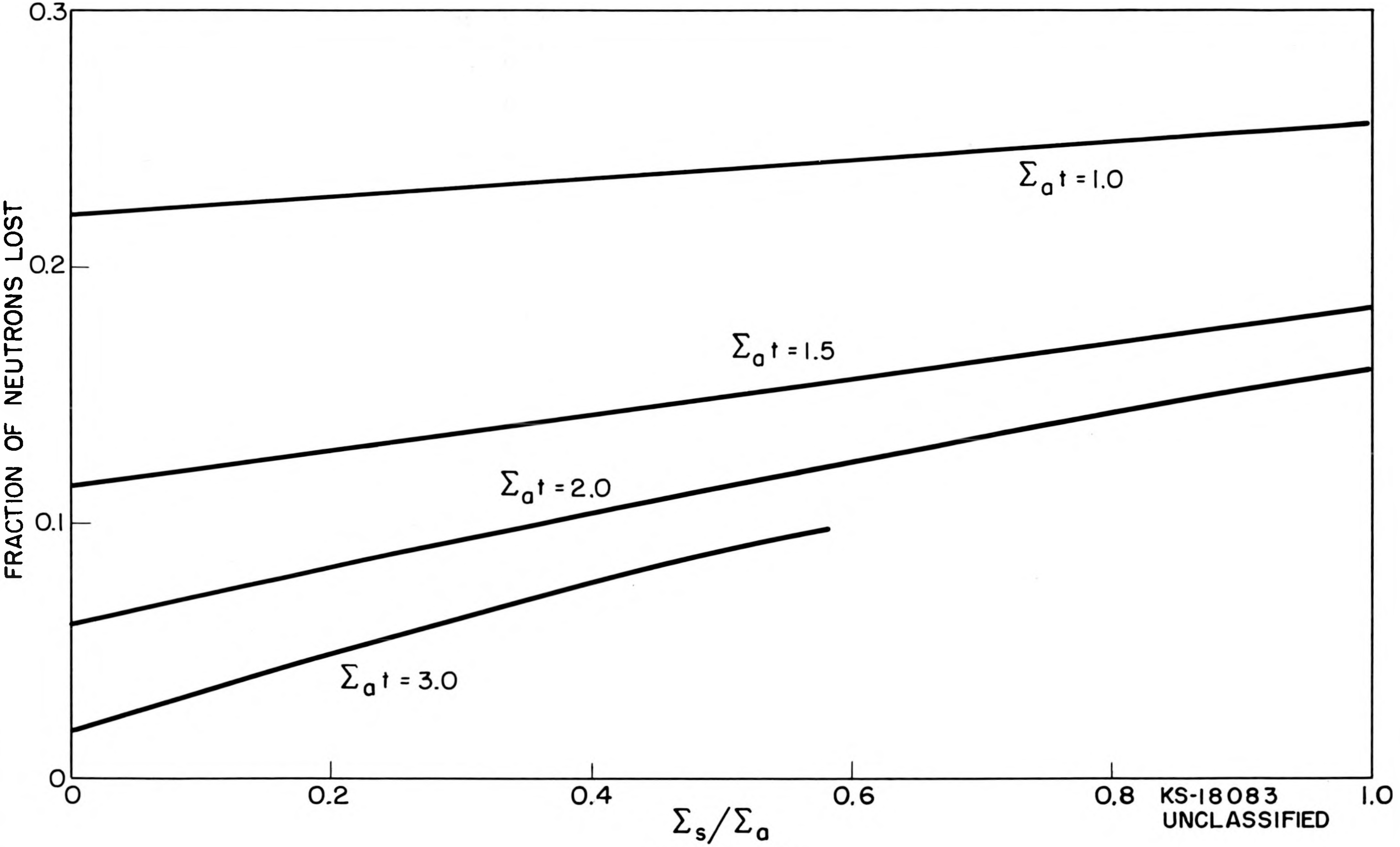

FIGURE 12

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