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SAVANNAH RIVER LABORATORY

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Neutron emission, DWPF, glass

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MEMORANDUM

January 3, 1986

TO: M. J. PLODINEC

FROM: D. J. PELLARIN, W. L. MATNEY, N. E. BIBLER
(for DJP, et al.)

(α, n) NEUTRON EMISSION FROM DWPF GLASS

INTRODUCTION

In the Defense Waste Processing Facility (DWPF), SRP waste will be immobilized in borosilicate glass. A knowledge of the neutron emission from DWPF glass is necessary to assess shielding requirements for the DWPF canister and to determine the response characteristics of the Neutron Transmission Glass Level Detection System. Fast neutrons are produced in DWPF glass from (α, n) reactions and spontaneous fission. The nuclear waste incorporated into the borosilicate glass consists of α-decaying actinides (primarily ^{238}Pu, ^{244}Cm and ^{241}Am) that accounts for 85% of the total neutron emission. The α-particles lose their energy in the immediate surroundings with a small fraction of them inducing (α, n) reactions. This neutron production increases with α-particle energy and depends on the material composition. It is generally only important for low atomic number elements where the Coulomb barrier for α-particles is low. The relevant elements in DWPF glass are mainly B, Li, O, Si, Al, Mg, F and Na.

It is not a straightforward matter to evaluate the thick target (α, n) neutron yield from a complex mixture such as DWPF glass. For this reason a program was initiated to measure the actual yield from three different glasses doped with known amounts of ^{238}Pu. The glasses covered a range of Pu-238 mole fractions that spanned expected DWPF conditions.

The experimentally determined values of total neutron emission and yield (neutrons per alpha) were compared to computations performed by two different calculational methods. The purpose of this program was to evaluate the calculational techniques and establish confidence in their use to predict the (α, n) neutron source term for DWPF glass.

SUMMARY

Excellent agreement was obtained between measured and calculated neutron emissions (yields) from Pu spiked black frit glasses using West's method of weighting components based on relative stopping power. The calculated values for the three glasses were 2-7% higher than measured. Calculations using a Nj Zj weighting method were 19-22% lower than measured. The good agreement between measurement and calculation using West's method lends confidence in its use to calculate the neutron source term for DWPF glass.

DISCUSSION

Measurement of (α, n) Yield

The neutron emission was measured from three glass samples prepared by N. E. Bibler. The samples consisted of Ferro black frit simulated waste glass spiked with PuO_2. The glasses, numbered 4, 5 and 6, are described in Table I and their compositions are given in Table II. Elemental weight fractions are given in Table III. The ^{238}Pu weights are based on alpha assay measurements of three samples from each glass. These samples were dissolved in HF-HCl, and the α activity of resulting solutions measured. The data are given in Table IV. Also, included in Table IV is the total alpha disintegration rate based on the total amount of Pu present. The small amount (~ 10 wt%) of $^{239}PuO_2$ in the PuO_2 has a negligible effect on the alpha assay because of the long alpha decay half life (2.44×10^4 yr) of ^{239}Pu.

A calibrated neutron detector was used to measure the fast neutron emission from each of the glass samples. The detector consisted of a 1 ft cube of plexiglas covered with 0.032-inch thick Cd. A vertical center hole accommodated the glass sample and another vertical hole, 3-3/4 inches away, accommodated a Nancy-Wood BF_3 counter.

The system was calibrated with fast neutrons from the SRCF-270 ^{252}Cf source. The neutron emission from SRCF-270 was measured by N. P. Baumann on 12/5/85 against a secondary standard. A value of 2.29×10^5 n/sec was obtained. An effort was made in the calibration measurements to simulate the effects of neutron scattering and absorption in the glass samples. The ^{252}Cf source was centered in a plastic cylinder, identical to that used for the glass samples, and surrounded by black frit. Auxiliary experiments established that elevation corrections were not required since the midplane of the BF_3 counter was aligned with the midplane of the glass samples and ^{252}Cf source. Rotation, however, was necessary to average small heterogeneities and positional inaccuracies. The counting rate data and calculated neutron emissions are summarized in Table V. The counter dead time was determined by the two source technique and a value of 1.96×10^{-7} min obtained. The counting rates were corrected for dead time and for a small 1 c/min background. Inherent in the measurement is the assumption that the energy spectrum of neutrons from (α, n) reactions in the glass is very similar to the enrgy spectrum of neutrons from ^{252}Cf spontaneous fission. In fact the (α, n) spectrum produced by 5.5 Mev α-particles on a thick B target peaks at 3.0 Mev[2] while the spectrum for ^{252}Cf neutrons peaks in the 1.0-2.0 Mev range[3]. It is estimated that the neutron emission from the glass samples may be underestimated by at most 10% as a result of spectrum effects.

<u>Calculation of (α, n) Yield</u>

A method of calculating the neutron yield (neutrons per alpha) in the mixture of compounds represented in glass from the thick target (α, n) yields of the separate constituents is provided by West[1]. The method is based on the Bragg additive law of stopping powers and determines the yield from a mixture (Y mix) by the expression

$$Y\ mix = \frac{\sum_j wJ\ CjAl\ Yj}{\sum_j Wj\ CjAl}$$

where, W_j = weight fraction of element j in the mixture

$$Cj, Al = \frac{\left(\frac{dE}{d\rho x}\right)_j}{\left(\frac{dE}{d\rho x}\right)_{Al}} = \text{relative stopping power}$$

Yj = thick target yield of element j

Tables of stopping power, $\frac{dE}{d\rho x}$, for α-particles incident on thick targets are given in the literature. Stopping powers relative to Al (Cj_{Al}) are given in Tables VI and VII from the compilation by Northcliffe and Schilling[4]. The data are for various solids and gases at an alpha energy of 5.49 Mev., corresponding to the weighted average energy of alphas from ^{238}Pu decay (72% at 5.50 Mev and 28% at 5.46 Mev). It is conventionally assumed that the stopping power varies smoothly with the atomic number (Z). The relative stopping power is plotted vs Z for solids in Figure 1 and for gases in Figure 2. The relative stopping powers of light elements represented in the glass were obtained from these plots.

Only a limited amount of thick target (α, n) yield (Yj) data is available in the literature. Fortunately data exists for most of the light elements represented in the glasses that were measured. Available values at 5.5 Mev are given in Table VIII. The (α, n) contributions from other elements such as Fe, Ni, Mn, Ca, Cl, and Pb cannot be included but numerically are small either because the weight fraction is small or because the element has a high Z (low yield). Calculated neutron emissions for Al, B, F, Li, Mg, Na, O and Si and the total emission for each of the glasses are presented in Table IX.

An alternate method of calculating the (α, n) production that has been used at SRP determines the yield from the mixture by the expression

$$Y\ mix = \frac{\sum j\ Nj\ Zj\ Yj}{\sum j\ Nj\ Zj}$$

where, Nj = atom density of element j in the mixture α Wj/Aj
Aj = atomic weight of element j
Zj - atomic number of element j
Yj = thick target yield of element j

Neutron emissions calculated by this weighting method are given in Table X. The calculated neutron emissions and yields are summarized and compared with measured values in Table XI.

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1. West D., "The Calculation of Neutron Yields in Mixtures and Compounds From The Thick Target (α, n) Yields in the Separate Constituents," Ann. Nucl. Energy, 6, 1970.

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5. Blair J. K. and Gomez del Campo J., "Neutron Yields From Alpha-Particle Bombardment," Nucl. Science and Engr., 71, (1979).

TABLE I

Glass Description

Glass	Wt, g	^{238}Pu Wt, g*	Diam, in.	Length, in.
4	24.542	1.08×10^{-2}	0.80	1 1/8
5	22.676	1.03×10^{-3}	0.80	1 1/8
6	24.657	3.64×10^{-4}	0.80	1 1/4

*See Table IV

TABLE II

Weight of Components in Glass Samples

Component	Weight, g		
	Glass 4	Glass 5	Glass 6
SiO_2	13.33	12.32	13.40
Fe_2O_3	3.02	2.79	3.03
Na_2O	2.53	2.34	2.54
B_2O_3	1.67	1.54	1.68
Li_2O	1.15	1.07	1.16
Al_2O_3	1.01	0.93	1.01
MnO_2	0.71	0.66	0.72
CaO	0.37	0.34	0.37
NiO	0.22	0.20	0.22
MgO	0.20	0.18	0.20
ZrO_2	0.29	0.27	0.30
F	0.015	0.014	0.015
Cl	0.012	0.011	0.012
Pb	0.012	0.011	0.012
$^{238}PuO_2$	1.23×10^{-2}	1.17×10^{-3}	4.13×10^{-4}

TABLE III

Elemental Weight Fractions

Element	Wt Fractions W_j
Al	.0218
B	.0211
F	6.1×10^{-4}
Li	.0218
Mg	4.9×10^{-3}
Na	.0765
O	.464
Ca	.0108
Cl	4.9×10^{-4}
Fe	.0861
Mn	.0183
Ni	7.0×10^{-3}
Pb	4.9×10^{-4}
Si	.254
Zr	8.8×10^{-3}

TABLE IV

Alpha Assay Measurements

Glass No.	Sample No.	Alpha/min/g of glass	^{238}Pu wt,* g	Total α dis. Rate, dis/sec
4	1	1.62×10^{10}		
	2	1.74×10^{10}		
	3	1.65×10^{10}		
	avg	$1.67 \times 10^{10} \pm .06 \times 10^{10}$	1.08×10^{-2}	6.83×10^{9}
5	1	1.64×10^{9}		
	2	1.74×10^{9}		
	3	1.82×10^{9}		
	avg	$1.73 \times 10^{9} \pm .09 \times 10^{9}$	1.03×10^{-3}	6.54×10^{8}
6	1	5.93×10^{8}		
	2	5.38×10^{8}		
	3	5.53×10^{8}		
	avg	$5.61 \times 10^{8} \pm .28 \times 10^{8}$	3.64×10^{-4}	2.31×10^{8}

*Based on ^{238}Pu alpha decay half life of 87.8 yr.

TABLE V

Counting Rate Data and Neutron Emission

Source	Position 1	Position 2	Position 3	Position 4	Avgd & Corr. Counting Rate c/min.	Neutron Emission n/sec.	Yield n/α
	Counting Rate, c/min						
SRCF-270	55,763	55,059	53,265	54,443			
	55,651	55,465	53,121	54,298			
	55,895	54,583	53,221	54,278			
		55,295					
avg	55,770	55,101	53,202	54,336	55,186	2.29×10^5	--
	Counting Rate, c/min						
Glass #4	1,130	1,120	1,179	1,153			
	1,189	1,167	1,167	1,226			
	1,222	1,143	1,086	1,138			
	1,219	1,109	1,121	1,127			
	1,261	1,120	1,170	1,158			
avg	1,204	1,132	1,145	1,160	1,160	4.81×10^3	7.04×10^{-7}
	Counting Rate, c/3 min						
Glass #5	321	391	336	370			
	299	319	365	379			
	331	341	382	331			
	323	357	359	355			
	321	345	347	343			
avg	319	351	358	356	114	4.73×10^2	7.23×10^{-7}
	Counting Rate, c/5 min						
Glass #6	214	209	225	222			
	228	213	243	203			
	197	233	205	195			
	201	225	211	234			
	194	225	223	193			
avg	207	221	221	209	42	1.7×10^2	7.40×10^{-7}

TABLE VI

Relative Stopping Power of Solids

Element, j.	At. No. Z	Relative Stop. Power cj, Al
Be	4	1.286
C	6	1.367
Al	13	1.000
Ti	22	0.825
Ni	28	0.735
Ge	32	0.687
Zr	40	0.618
Ag	47	0.573
Eu	63	0.453
Ta	73	0.410
Au	79	0.387
U	92	0.347

TABLE VII

Relative Stopping Power of Gases

Element, j	At. No. Z	Relative Stop. Power Cj, Al
H	4	4.809
He	2	1.829
N	7	1.308
O	8	1.249
Ne	10	1.114
Ar	18	0.841
Kr	36	0.595
Xe	54	0.482
Rn	86	0.367

TABLE VIII

Summary of (α, n) Yields at 5.50 Mev.

Element J	Yield, n/α Yj	Reference
Al	7.47×10^{-7}	2
B	2.06×10^{-5}	2
F	9.50×10^{-6}	2
Li	2.15×10^{-6}	5
Mg	1.33×10^{-6}	2
Na	1.5×10^{-6}	
O	6.46×10^{-8}	2
Si	1.13×10^{-7}	2

TABLE IX

Calculated Neutron Emission by WjCjAl Weighting

Element j	Wt. Fract Wj	Rel.S.P. CjAl	Yield, Yj	Neutron Emission, n/s Glass 4	Glass 5	Glass 6
Al	2.18×10^{-2}	1.00	7.47×10^{-7}	101	10	3
B	2.11×10^{-2}	1.26	2.06×10^{-5}	3,410	327	115
F	6.10×10^{-4}	1.19	9.50×10^{-6}	43	4	1
Li	2.18×10^{-2}	1.3	2.15×10^{-6}	388	37	13
Mg	4.9×10^{-3}	1.03	1.33×10^{-6}	42	4	1
Na	7.65×10^{-2}	1.05	1.5×10^{-6}	750	72	25
O	4.64×10^{-1}	1.26	6.46×10^{-8}	235	23	8
Si	2.54×10^{-1}	0.98	1.13×10^{-7}	175	17	6
Cl	4.90×10^{-4}	0.87	--			
Fe	8.61×10^{-2}	0.76	--			
Mn	1.83×10^{-2}	0.77	--			
Ni	7.0×10^{-3}	0.73	--			
Pb	4.9×10^{-4}	0.73	--			
Ca	1.08×10^{-2}	0.85	--			
Zr	8.8×10^{-3}	0.61	--			
		Total, n/s		5144	494	174

$\Sigma WjCj_{Al} = 1.097$

$Y\ mix = 7.53 \times 10^{-7}\ n/\alpha$

TABLE X

Calculated Neutron Emission by Bj NjZj Weighting

Element j	At. Wt Aj	Wt. Fract. Wj	At. No. Zj	Yield Yj	Neutron Emission, n,s Glass 4	Glass 5	Glass 6
Al	26.98	2.18×10^{-2}	13	7.47×10^{-7}	110	11	4
B	10.82	2.11×10^{-2}	4	2.06×10^{-5}	2,253	216	76
F	19.00	6.10×10^{-4}	9	9.50×10^{-6}	38	4	1
Li	6.94	2.18×10^{-2}	3	2.15×10^{-6}	284	27	10
Mg	24.32	4.9×10^{-3}	12	1.33×10^{-6}	45	4	2
Na	22.991	7.65×10^{-2}	11	1.5×10^{-6}	770	74	26
O	16.0	4.64×10^{-1}	8	6.46×10^{-8}	210	20	7
Si	28.09	2.54×10^{-1}	14	1.13×10^{-7}	201	19	7
Cl	35.457	4.90×10^{-4}	17	--			
Fe	55.85	8.61×10^{-2}	26	--			
Mn	54.94	1.83×10^{-2}	25	--			
Ni	58.71	7.0×10^{-3}	28	--			
Pb	207.21	4.9×10^{-4}	82	--			
Ca	40.08	1.08×10^{-2}	20	--			
Zr	91.22	8.8×10^{-3}	40	--			
	Total, n/s				3911	375	133

$$\sum_j \frac{W_j}{A_j} Z_j = 0.4871$$

TABLE XI

Summary of Measured and Calculated Neutron Emissions and Yields

	Measured		Calculated			
			WjCjAl Method		NjZj Method	
	Neutron Emission, n/s	Neutron Yield, n/α	Neutron Emission, n/s	Neutron Yield, n/α	Neutron Emission, n/s	Neutron Yield, n/α
Glass 4	4,810	7.04×10^{-7}	5,144	7.54×10^{-7}	3,911	5.74×10^{-7}
Glass 5	473	7.23×10^{-7}	494	7.54×10^{-7}	375	5.74×10^{-7}
Glass 6	170	7.36×10^{-7}	174	7.54×10^{-7}	133	5.74×10^{-7}

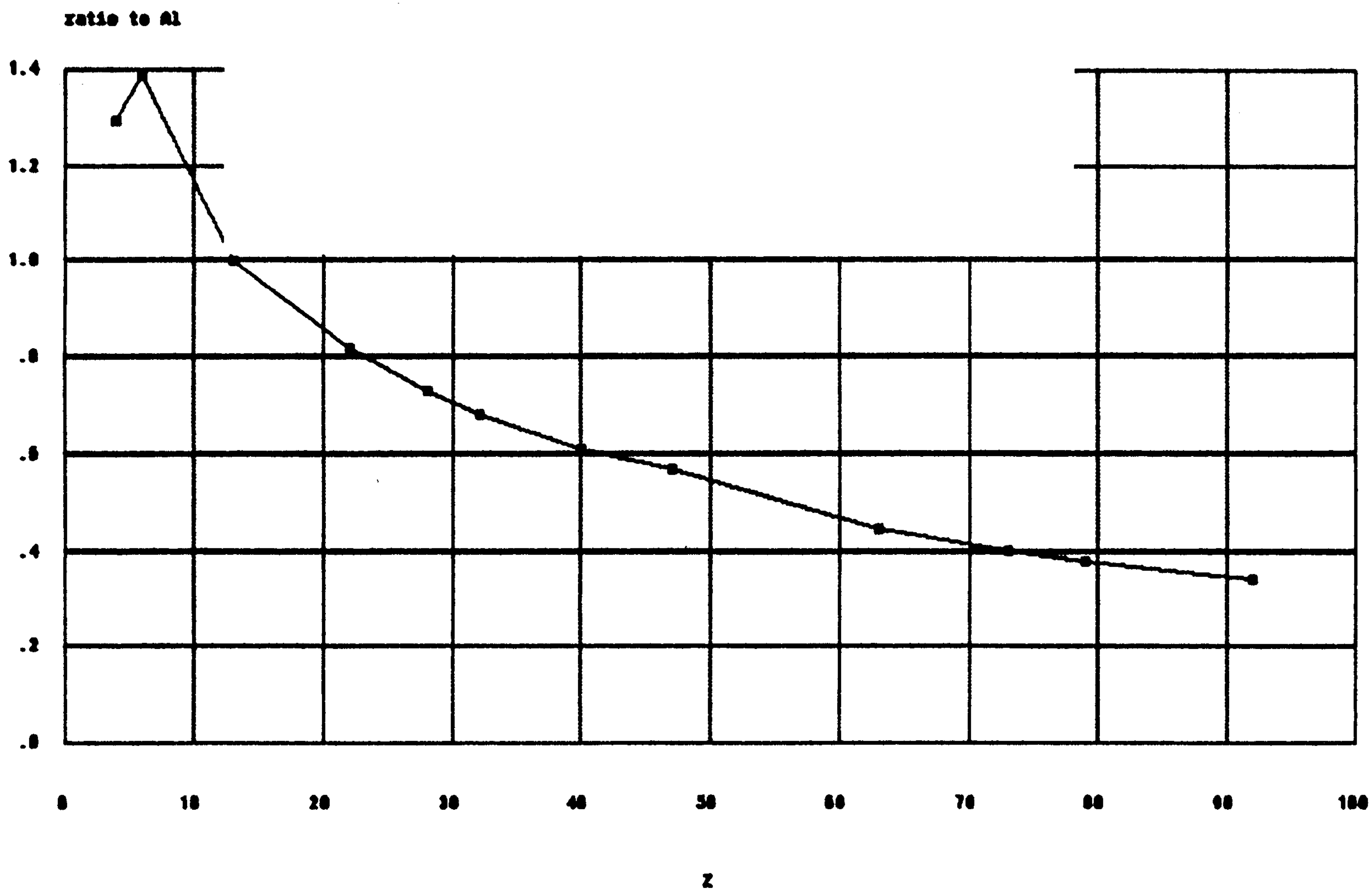

FIGURE 1

STOPPING POWERS VS Z FOR SOLIDS

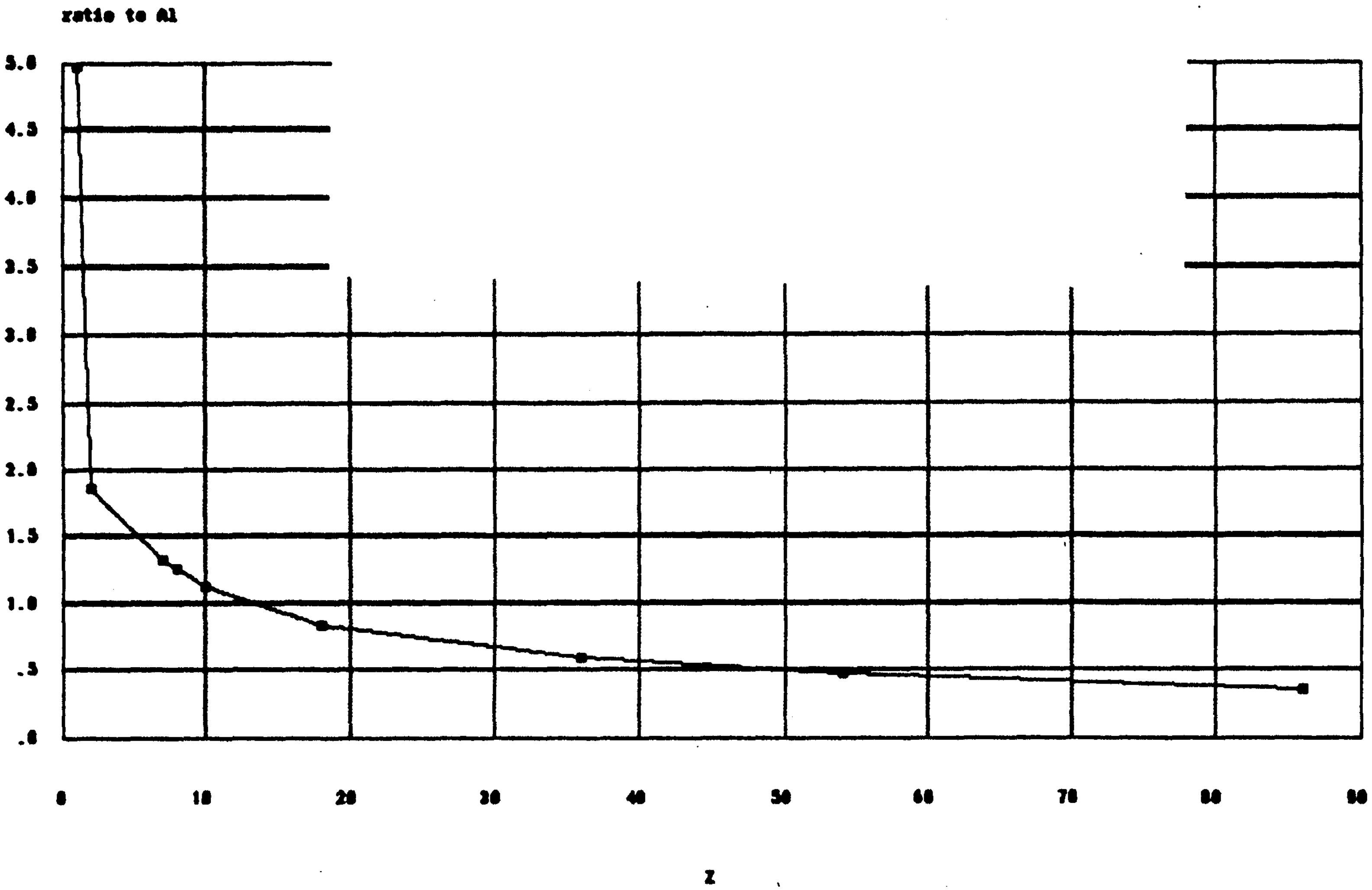

FIGURE 2

STOPPING POWERS VS Z FOR GASES