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A THEORETICAL INVESTIGATION OF ABSOLUTE NEUTRON DETECTION

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ABSTRACT

A survey was made of the detection systems classified as 'flat response' in order to establish their use in high accuracy measurements of absolute neutron intensities for a wide range of neutron sources. Two detection systems involving neutron moderation in either graphite or an hydrogenous material were selected as having the most suitable characteristics.

Numerical solutions of the neutron transport equations for these systems were computed to determine the conditions required to produce minimum variation of efficiency with energy. It was shown that, with the suitable choice of moderator composition and size, it should be possible to reduce these errors to less than one per cent over the energy range 100 eV to 4 MeV.

*Attached to the A.A.E.C. from the University of Western Australia for part of this investigation.

CONTENTS

(continued)

CONTENTS (continued)

1. INTRODUCTION

At the present time accuracies of better than one per cent are required in the measurement of absolute neutron intensities, for example, in the calibration of neutron sources, in (γ,n) reactions and in the determination of the mean number of neutrons emitted per fission event. The major obstacle to achieving this accuracy arises from the variation of detection efficiency with neutron energy, accompanied by a lack of detailed knowledge concerning the energy spectrum of the neutron sources. The effect of this limitation can be significantly reduced if most of the neutrons are thermalised in a moderating material before counting. In addition, errors from anisotropic neutron emission can be minimised by situating the source at the centre of such a moderator and detecting the neutrons in 4π geometry.

For the actual detection of neutrons, one normally measures their capture rate in an absorbing material whose cross section varies inversely with neutron velocity (1/v), for example BF_3 counters. For neutrons thermalised in a purely moderating material this can be considered as a measurement of the thermal neutron density at the point of detection since the absorption cross section decreases rapidly with energy and the neutron intensities are relatively low above thermal energies.

Two types of detection systems were considered in this investigation. The first is referred to as the neutron leakage detector because the 1/v counters are positioned at the boundary of the moderator; the second is called the neutron density detector because the counters are positioned throughout the volume of the moderator in order to obtain the neutron capture rates as a function of moderator radius.

The aim of the investigation was to determine

(a) the major factors which govern the variation of neutron detection efficiency with source energy for both detection systems, and

(b) the minimum variation in detection efficiency with source energy that could be achieved in the two types of detection systems.

For this purpose a numerical analysis of both systems was executed using energy group condensed cross sections from the GYMEA library (Pollard and Robinson 1966) together with the neutron transport equation embodied in the computer programmes DSN (Clancy et al. 1965) and WDSN (Francescon 1963).

2. SURVEY OF EXISTING DETECTION SYSTEMS

2.1 Neutron Leakage Detectors

The possible fates of neutrons emitted from the source are fast leakage, thermal leakage and thermal capture. Previous experiments (see below) have shown the existence of optimum moderator radii at which the fast leakage and thermal capture compensate over a wide energy range. Hence for etector of this size the ratio of thermal leakage to source strength is only a very slowly varying ction of initial energy. No detailed calculations of this optimum radius could be found in the rature.

For graphite moderators, Macklin (1957), using age diffusion theory, predicted that the iation in neutron leakage was a minimum for a sphere of radius about 80 cm. Experimental asurements (using BF_3 counters) confirmed his calculations. The efficiency of the system was nstant to within 2 per cent over the energy range 1 keV to 2 MeV, and with eight counters uniformly stributed over the moderator boundary, maximum anisotropy of the source produced less than 1.4 per nt change in efficiency up to 8 MeV.

A similar system was suggested by Rossi and Staub (1949) who predicted that a detector sing a paraffin moderator would give a flat response if the source–detector separation was care– ully selected. No theoretical calculations were made but this principle was used in the detection ystem of Halpern et al. (1952) who assumed a flat response for the same source–detector separation s used by Rossi and Staub. The best performance to date achieved with this type of detector was that of Marion et al. (1960) in which the response was flat to within 5 per cent over the energy range 100 keV to 2.5 MeV.

2.2 Neutron Density Detectors

For the neutron density detector the moderator radius and concentration of 1/v absorber are chosen to reduce the neutron leakage to any required value over the range of neutron energies being studied. Numerical integration of the neutron capture rates over the volume of the moderator should then provide energy-insensitive measurements of the neutron source strengths within this energy range.

The manganese bath (Axton et al. 1963, 1965) is a typical example of this technique. The source is situated at the centre of the detector and the emitted neutrons are thermalised and captured in an aqueous solution of manganese sulphate. The detector is made large enough for the neutron leakage to be negligible. A measurement of the neutron source strength is obtained from the induced activity of the Mn^{56}. Although capable of high accuracy this method is not ideally suitable for general use in research work because it requires elaborate radiochemical techniques and a steady source intensity over the period of the measurement.

An efficiency of greater than 90 per cent (varying by less than two per cent from thermal to fission neutron energies) has been achieved using an entirely different method of neutron detec– tion in an hydrogenous moderator. This is the liquid scintillator (Hopkins and Diven 1963, Asplund– Nilsson et al. 1963, Mather et al. 1964, and Boldeman and Dalton 1967). Its disadvantages are firstly, a high sensitivity to gamma rays (making it unsuitable for photoneutron work), and secondly, the considerable decrease in its detection efficiency for neutron energies above about 2 MeV (25 per

cent decreases at 8 MeV). Monte Carlo calculations were used by Hopkins and Diven (1963) and Mather et al. (1964) to calculate the variation of efficiency with neutron energy.

The boron pile (Colvin and Sowerby 1958) has a detection efficiency of about 64 per cent for fission spectra, as well as a flat response (again only 2 per cent from thermal to fission energies). The relatively high efficiency results from using over two hundred BF_3 detectors distributed uniformly in a large block of graphite, the source being situated at the centre of the block. Colvin and Sowerby quote errors of ±½ per cent in measuring the number of neutrons emitted per fission, but the effici– ency decreases considerably for neutrons of higher energy. In addition the high cost of such a detector prohibits its general use in research work.

A similar neutron density detector, but one having a much lower efficiency (1 per cent or less) has been suggested by Thies (1959). The neutron density distribution within the moderator surrounding the source is measured using a small number of BF_3 counters. The neutron leakage is reduced to a very low value by the homogeneous addition of 1/v absorber (poison) to the moderator. Numerical integration of the measured neutron densities over the volume of the moderator should produce an energy–insensitive measurement of source intensity.

3. PHYSICAL REQUIREMENTS FOR A DETECTION SYSTEM

3.1 Moderator Materials

The use of high intensity beams of gamma rays in photoneutron investigations precludes the use, in a detection system, of moderators containing nuclei with a low (γ,n) threshold, such as Be^9 (1.7 MeV) and D (2.2 MeV). For pure neutron source measurements the use of such moderators is impractical owing to the nuclear reactions $Be^9(n,\alpha)$, $Be^9(n,2n)$ and $D(n,2n)$ with thresholds at about 1, 2 and 3.3 MeV respectively (Stehn et al. 1964). Since a versatile detection system was being sought, only graphite and hydrogenous moderators were further considered.

The absorption of neutrons in the moderator will produce an energy dependence in the detection efficiency only if the variation of the capturing cross section is non–1/v. For hydrogen and graphite the error introduced by assuming a 1/v variation up to high energy (MeV region) is negligible when used in conjunction with a thermal neutron energy distribution (see Sections 5 and 6). This assumption could be invalidated if non–1/v cross section impurities were present in the moderator, and it is therefore essential that the amount of such materials present should be kept as small as possible.

3.2 Geometry

Spherical geometry would give maximum applicability to the calculations because the computer codes are one–dimensional only. The uniform distribution of neutron counters about a source centrally situated in a spherical moderator would reduce the errors arising from aniso–

tropic emission of neutrons from the source. However, of the two available transport codes, WDSN and DSN, only the latter could handle spherical geometry. Therefore, in order to compare the results from both codes, all calculations were carried out in cylindrical geometry.

3.3 Neutron Counters

The neutron counters should be so chosen that their composition and size produce negligible perturbation to the neutron density in the moderator. When situated within the moderator (for neutron density measurements) perturbations produced by the counters can be reduced by matching the concentrations of $1/v$ absorber in the moderator and in the neutron counters. When situated outside the moderator (for neutron leakage measurements), back scattering of neutrons from the counters may perturb the neutron balance in the moderator. Hence materials used in these counters must be of low scattering cross section.

4. FACTORS INVESTIGATED

4.1 Neutron Leakage Detector System

The variation of leakage out of the moderator boundary was determined as a function of source energy, moderator radius, moderator absorption cross section and moderator density. The variations caused by the addition of small amounts of poison were also calculated.

4.2 Neutron Density Detector System

A $1/v$ poison was mixed homogeneously with the moderator and variation of absorption was determined as a function of source energy, moderator radius and poison concentration.

4.3 General Considerations

It is desirable to restrict each source energy to narrow limits in order to simulate mono-energetic sources. The best that could be done in this investigation was to represent the neutron source by a single GYMEA energy group, and at higher energies this corresponded to a relatively wide spread in the source energy (see Table 1). To economise in computer time, only three neutron energy groups were used in the analysis. The energy boundaries of these groups were different for each source energy and are also given in Table 1, while the corresponding energies at mean lethargy are listed in Table 2.

5. GRAPHITE MODERATED DETECTORS

It would appear that graphite satisfies most requirements for moderating materials used in detection systems. Its (γ,n) and $(n,2n)$ thresholds are outside the range of the neutron source energies and its absorption cross section is known to vary as $1/v$ up to an energy of about 10 keV; its mass number is sufficiently high for isotropic scattering to be a good approximation and from the practical point of view it can be obtained in a very pure s... at reasonable cost.

The absorption cross section of graphite was represented in the GYMEA library as follows: at thermal energies its value was 3.8 mb decreasing as $1/v$ to 1.0 mb at 0.5 eV, then remaining constant at 1.0 mb up to about 2 MeV. For the type of problem normally considered (reactor systems) this crude approximation was adequate owing to the relatively small value of the cross section. However for problems involving small localised sources in a large volume of graphite a more accurate representation was found to be required, namely, a thermal value of 3.4 mb (Stehn et al. 1964) and a $1/v$ variation with energy over the whole range.

5.1 Neutron Leakage Detector

The variation with radius of the thermal neutron leakage from the boundary of a graphite cylinder is shown in Figures 1a and 1b. For both densities (1.60 g cm^{-3} and 1.63 g cm^{-3}) minimum dependence on the energy of the neutron sources considered occurs near 84 cm (details in Table 3).

5.1.1 Graphite density

The variation of thermal neutron leakage with source energy and graphite density is illustrated in Figure 2 for cylinders of three different radii. For each radius the variation with source energy decreases with decrease in graphite density, the effect being more marked for 80 cm and 85 cm radii. For both these radii a linear extrapolation of the leakages indicates that a small change in the graphite density to 1.56 g cm^{-3} (N = .078) would be sufficient to reduce the variation with energy to 0.5 per cent.

5.1.2 Poison concentration

Small additions of $1/v$ poison to the moderator affect the thermal neutron leakage as shown in Figure 3. Although the radius at which minimum variation with source energy occurs decreases with increasing poison concentration, there is no apparent change in the magnitude of the variation which equals about 1.1 per cent in both cases (compare also with zero poison case, Figure 1a).

The addition of poison could also be considered theoretically as an increase in the absorption cross section of the moderator. On this basis it would be interesting to investigate the effect of a smaller absorption cross section. Figure 4 shows that for a graphite cylinder of 90 cm, the thermal leakages associated with neutron sources at the three energies considered converged to a common value when linearly extrapolated to 2.5 mb. For an absorption cross section of this magnitude, zero variation in detection efficiency would be obtained.

5.1.3 Effect of a non-$1/v$ cross section

The least complicated and most convenient way of representing a non-$1/v$ cross section was to use the data for graphite which appeared in the GYMEA library. Using this data in conjunction with graphite of density 1.63 g cm^{-3} the radius for minimum energy dependence was found to be 86 cm (Figure 5). This is more clearly shown in Table 4. The results indicate that for the leakage detector, the presence of a non-1/v absorbing material in the moderator does not necessarily increase the energy dependence, and could in certain cases even reduce it.

5.2 Neutron Density Detector

The volume integral of the neutron density is equal to the difference between the neutron source intensity and the total leakage from the boundary of the moderator. As this leakage is greater for higher neutron source energies, it is necessary to consider only source energies of the maximum value for which the detector is to be used.

5.2.1 Reduction of neutron leakage

For neutrons of all energies, the leakage out of the detection system can be reduced by increasing the radius of the moderator. For thermal neutrons the leakage can also be reduced by the addition of 1/v poison. Figure 6 shows the effect of increasing moderator radius on the leakage from a centrally situated neutron source of 4.2 MeV. The effect produced by the addition of poison to a graphite cylinder of 120 cm radius is illustrated in Figure 7. The associated variation of neutron leakage with radius is given in Table 5 for zero poisoning and in Table 6 for a poison concentration equivalent to that in a typical BF_3 counter (see Appendix). From these results it is possible to specify the conditions required to reduce the total leakage from the neutron density detector to the required level over a specified range of source energies.

5.2.2 Effect of a non-1/v cross section

Using the GYMEA representation of the graphite absorption cross section, the neutron leakage from an unpoisoned graphite cylinder of 120 cm radius was calculated (Table 7). The variation of the neutron leakage with poison concentration for a 120 cm radius moderator is shown in Figure 8, which indicates that from a 4.2 MeV source a total neutron leakage of only 0.6 per cent is achieved for a poison concentration of $P = 0.05$ (equivalent to a typical BF_3 counter). Analysis of the neutron absorption within the moderator produced the results shown in Table 8. Thus although the total neutron absorption varies by only 0.6 per cent, the absorption of neutrons in the non-1/v cross section at different energies would produce a variation of 2.4 per cent in the detection efficiency (which is proportional to the absorption in the 1/v poison). These calculations illustrate the fact that, in the presence of a non-1/v cross section, the absorption reaction rate is not proportional to neutron density alone but to some function with an energy dependence. Hence the accuracy with which the energy response of this type of detection system can be known is limited by the extent to which non-1/v absorbing materials can be excluded from the graphite.

6. HYDROGENOUS MODERATED DETECTORS

One of the disadvantages of a graphite moderator is the large size necessary to produce thermalisation. Achieving a similar effect with a smaller detection system would necessitate

using a more effective moderating material such as hydrogen. Some calculations were carried out on water and paraffin wax (represented for simplicity as CH_2) to estimate the relative decrease in size resulting from the use of these materials.

6.1 Calculations

It was found that with these hydrogenous moderators, the calculations for the lower source energies would not converge. It was therefore impossible to investigate the leakage detection system because optimum radius determinations need the results for neutron sources over the entire energy range. However, it was feasible to investigate the neutron density detector as only the calculations for neutrons of the maximum energy are required.

Figures 9 and 10 show the variation of leakage with radius for cylindrical moderators of water and paraffin wax taking the neutron source energy as 4.2 MeV.

6.2 Discussion

The radius at which the total leakage from hydrogenous moderators is reduced to less than one per cent is much smaller than that for graphite. The reduction in size results from the fact that both the scattering and the absorption cross sections of hydrogen are larger than those for graphite. The smaller radius obtained for paraffin wax is due to the relatively higher concentration of hydrogen nuclei (see Appendix). No significant reduction in the radius would be achieved by poisoning an hydrogenous moderator because the epithermal leakage is greater than the thermal leakage at all radii. Since the effective poison concentration inherent in the hydrogen absorption cross section lies in the range covered by enriched BF_3 counters, it is possible to minimize their perturbation by:

(a) selecting BF_3 counters with an equivalent amount of poison, or

(b) adding small amounts of 1/v absorber to the moderator so that the capture rate in the moderator per unit volume is the same as that in the detector.

7. CONCLUSIONS

The investigations showed that using computer techniques it was possible to specify the major factors governing the variation of detector efficiency with source energy for two different detection systems.

An optimisation study of both thermal neutron leakage detectors and neutron density detectors showed that an accuracy of greater than 1 per cent should be attainable in neutron intensity measurements over the energy range 100 eV to 4 MeV.

8. RECOMMENDATIONS

The scope and accuracy of the calculations would be improved if:

(a) cross section data and finer group structure were used to represent the source at higher source energies,

(b) many more neutron energy groups were used in the neutron transport calculations,

(c) anisotropic cross sections were included,

(d) the number of spatial mesh points was increased, especially in the vicinity of the source (all calculations might then converge), and

(e) molecular scattering cross sections were used for the study of hydrocarbons.

However, in principle, the available computer codes gave a reasonable description of the detection systems considered.

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APPENDIX

NUCLEAR CONCENTRATIONS

For their use in the computer programmes, nuclear concentrations of the various materials were specified in units of nuclei per 10^{-24} cm^3, and were represented in this paper by the symbol N; these are given below together with their thermal absorption macroscopic cross sections.

(a) Carbon

Values of the density of carbon range from 1.60 to 1.63 g cm^{-3}. These correspond to values of N = 8.03 to 8.18 x 10^{-2}.

The thermal absorption cross section assumed in the GYMEA library was 3.8 mb; the latest recommended value (Stehn et al. 1964) is 3.4 mb. To cover both these values and the range of densities mentioned above, macroscopic cross sections of 2.73 x 10^{-4} to 3.05 x 10^{-1} cm^{-1} were used.

(b) Hydrocarbons

For hydrogen in water and in paraffin, N = 6.69 x 10^{-2} and 8.60 x 10^{-2} respectively. Using a density of 1.0 g cm^{-3} and 1.627 g cm^{-3} for water and paraffin, together with a microscopic cross section of 0.33b for hydrogen, gives macroscopic absorption cross sections of 2.21 x 10^{-2} cm^{-1} for water and 4.62 x 10^{-2} cm^{-1} for paraffin.

(c) 1/v absorbers

In the GYMEA library the cross section of a 1/v absorber is represented in terms of the equivalent nuclear concentration of an imaginary material having unit microscopic cross section at thermal energies. For this report the degree of poisoning is expressed simply in terms of this material and its concentration is denoted by the symbol P, its units being nuclei per 10^{-24} cm^3.

For a specific absorber such as boron enriched to 90 per cent in B^{10} (whose microscopic cross section is 3532b), the average value of N over the volume of a typical enriched BF_3 counter is 1.87 x 10^{-5}, which is equivalent to a macroscopic cross section of 6.59 x 10^{-2} cm^{-1}. Expressing this in terms of poisoning gives P = 6.59 x 10^{-2}.

TABLE 1

ENERGY BOUNDARIES OF THE NEUTRON SOURCES AND THEIR ASSOCIATED NEUTRON GROUPS

Source Energy	Neutron Energy Groups 1	2	3
10 MeV – 1.74 MeV	10 MeV – 1.74 MeV	1.74 MeV – 0.51 eV	0.51 eV – 0.001 eV
143 keV – 86.5 keV	143 eV – 86.5 keV	86.5 keV – 0.51 eV	0.51 eV – 0.001 eV
130 eV – 78.9 eV	130 eV – 78.9 eV	78.9 eV – 0.51 eV	0.51 eV – 0.001 eV

TABLE 2

CHARACTERISTIC ENERGIES OF THE NEUTRON SOURCES AND THEIR ASSOCIATED NEUTRON GROUPS

Source Energy	Neutron Groups 1	2	3
4.16 MeV	4.16 MeV	937 eV	0.023 eV
111 keV	111 keV	209 eV	0.023 eV
101 eV	101 eV	6.3 eV	0.023 eV

TABLE 3

RELATIVE DIFFERENCES IN THERMAL NEUTRON LEAKAGE FROM GRAPHITE CYLINDERS OF THE OPTIMUM RADII

Graphite Density (g cm^{-3})	Optimum Radius (cm)	Source Energy 101 eV	111 keV	4.2 MeV
		Relative change in leakage (%)		
1.60	84.0	0.0	1.1	0.0
	88.8	0.0	1.3	1.3
1.63	83.3	0.0	1.3	0.0
	87.5	0.0	1.6	1.6

TABLE 4

RELATIVE DIFFERENCES IN THERMAL NEUTRON LEAKAGE FOR A GRAPHITE CYLINDER CONTAINING A NON–1/v ABSORBER

Graphite Density (g cm^{-3})	Radius (cm)	Source Energy 101 eV	111 keV	4.2 MeV
		Relative difference in leakage (%)		
1.63	85.0	0.0	−0.3	0.5
	86.0	0.0	0.7	0.0
	87.0	0.0	0.8	0.3

TABLE 5

VARIATION IN NEUTRON LEAKAGE FROM A GRAPHITE CYLINDER (DENSITY 1.60 g cm^{-3}) FOR A 4.2 MeV SOURCE

Radius (cm)	Energy of Neutron Group 4.2 MeV	937 keV	0.023 eV
	Relative leakage (%)		
80	0.10	5.85	68.02
90	0.04	3.58	64.13
100	0.02	2.19	59.39
120	0.00	0.87	49.13
140	0.00	0.39	39.51

TABLE 6

VARIATION IN NEUTRON LEAKAGE FROM A POISONED GRAPHITE CYLINDER (DENSITY 1.60 g cm^{-3} AND P = 0.05) FOR A 4.2 MeV SOURCE

Radius (cm)	Energy of Neutron Group 4.2 MeV	937 keV	0.023 eV
	Relative leakage (%)		
80	0.10	5.72	0.59
90	0.04	3.58	0.36
100	0.02	2.02	0.21
120	0.00	0.69	0.07
140	0.00	0.23	0.03

TABLE 7

VARIATION IN NEUTRON LEAKAGE FROM AN UNPOISONED GRAPHITE CYLINDER (120 cm RADIUS, GYMEA ABSORPTION CROSS SECTION)

Source Energy	Neutron Energy Group		
	1	2	3
	Relative Leakage (%)		
4.2 MeV	0.00	0.73	43.81
111 keV	0.00	0.20	43.12
101 eV	0.00	0.05	42.32

TABLE 8

ABSORPTION OF NEUTRONS IN A POISONED GRAPHITE CYLINDER (120 cm RADIUS, GYMEA ABSORPTION CROSS SECTION, P = 0.05)

Source Energy	Absorption in 1/v Poison (%)	Absorption in Poison + Graphite (%)
4.2 MeV	96.6	99.4
111 keV	97.9	99.9
101 eV	98.9	100.0

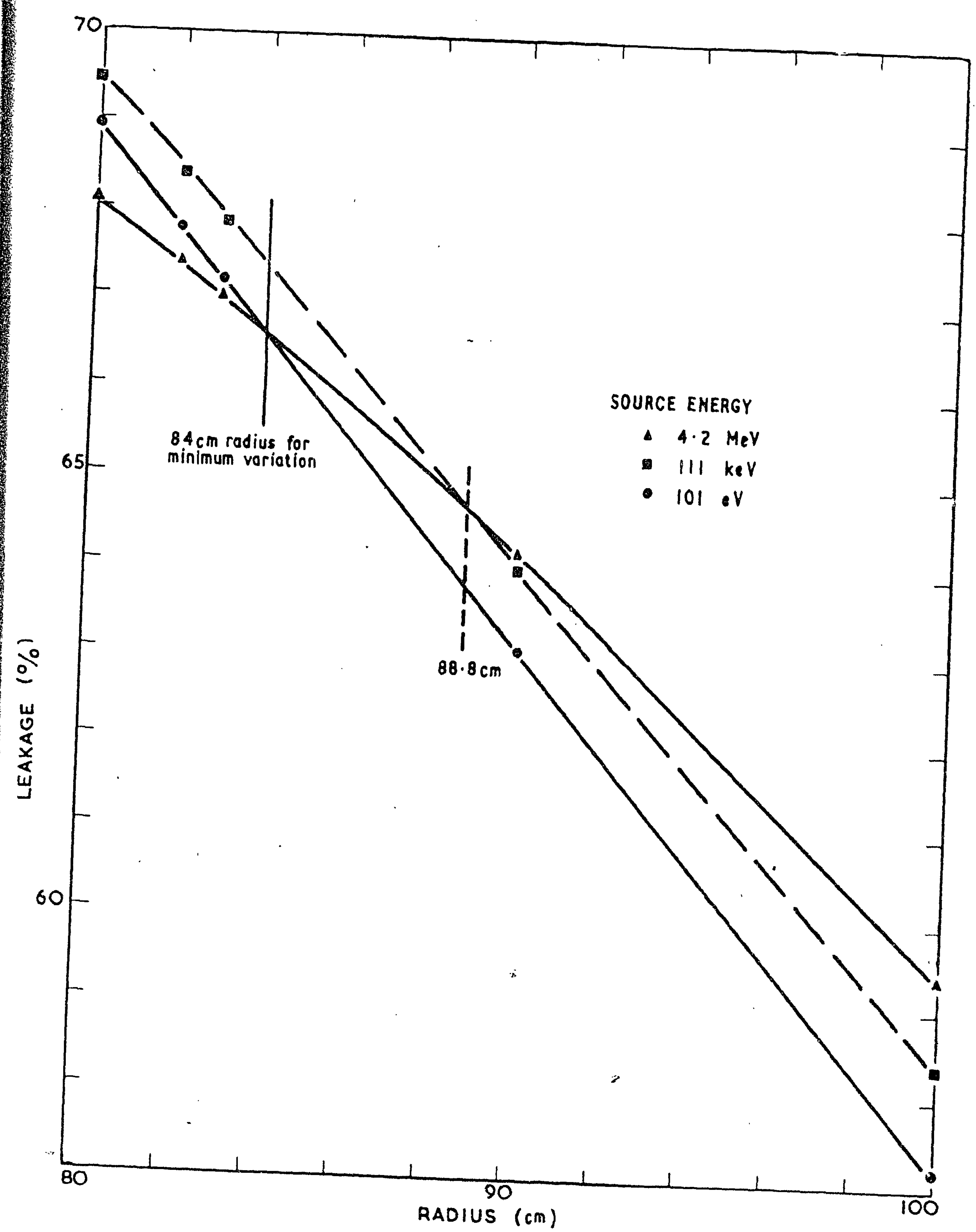

FIGURE 1a. VARIATION OF THERMAL NEUTRON LEAKAGE WITH RADIUS FOR A GRAPHITE CYLINDER (DENSITY 1.60 g cm^{-3})

P1204

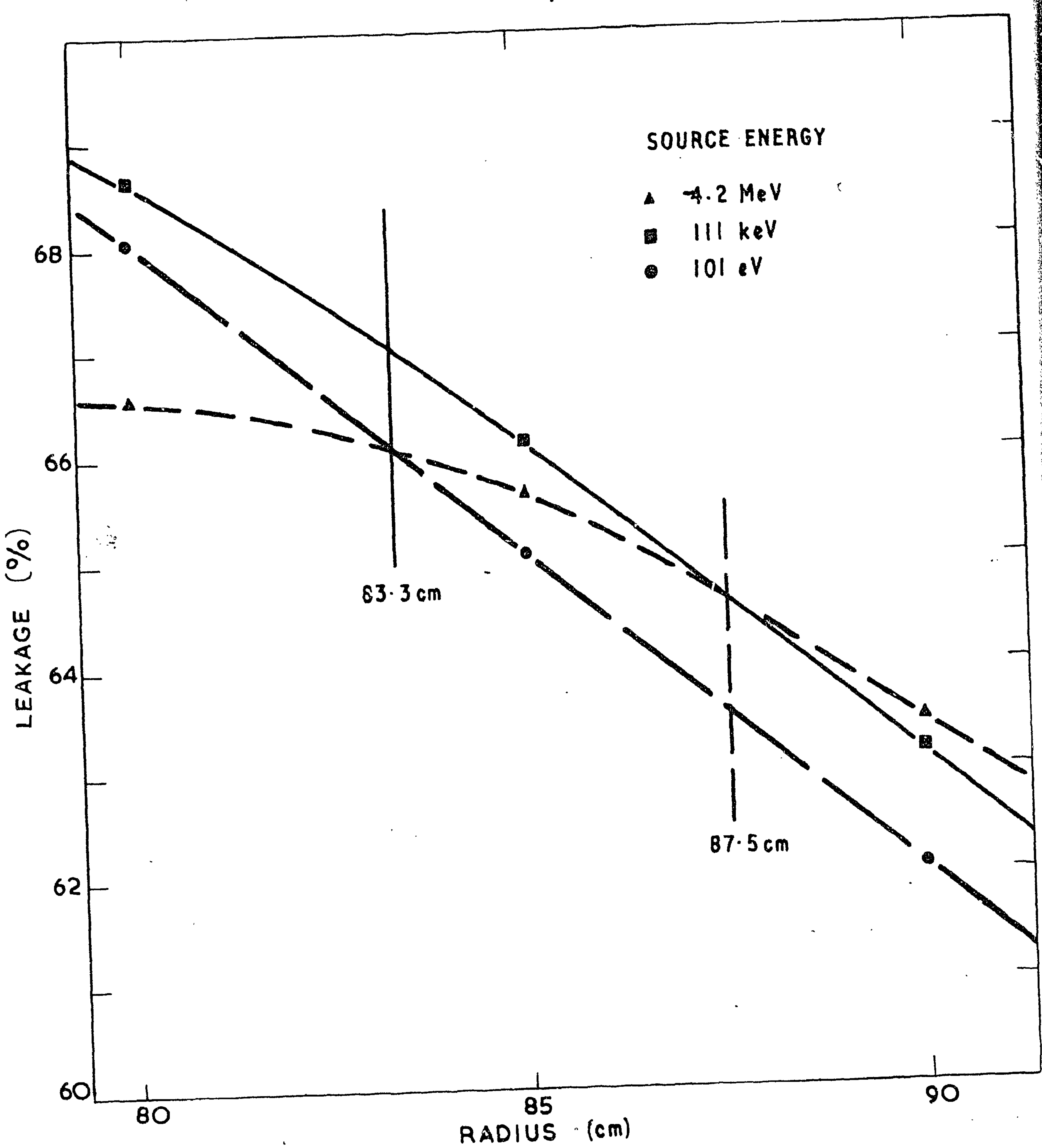

FIGURE 1b. VARIATION OF THERMAL NEUTRON LEAKAGE WITH RADIUS FOR A GRAPHITE CYLINDER (DENSITY 1.63 g cm^{-3})

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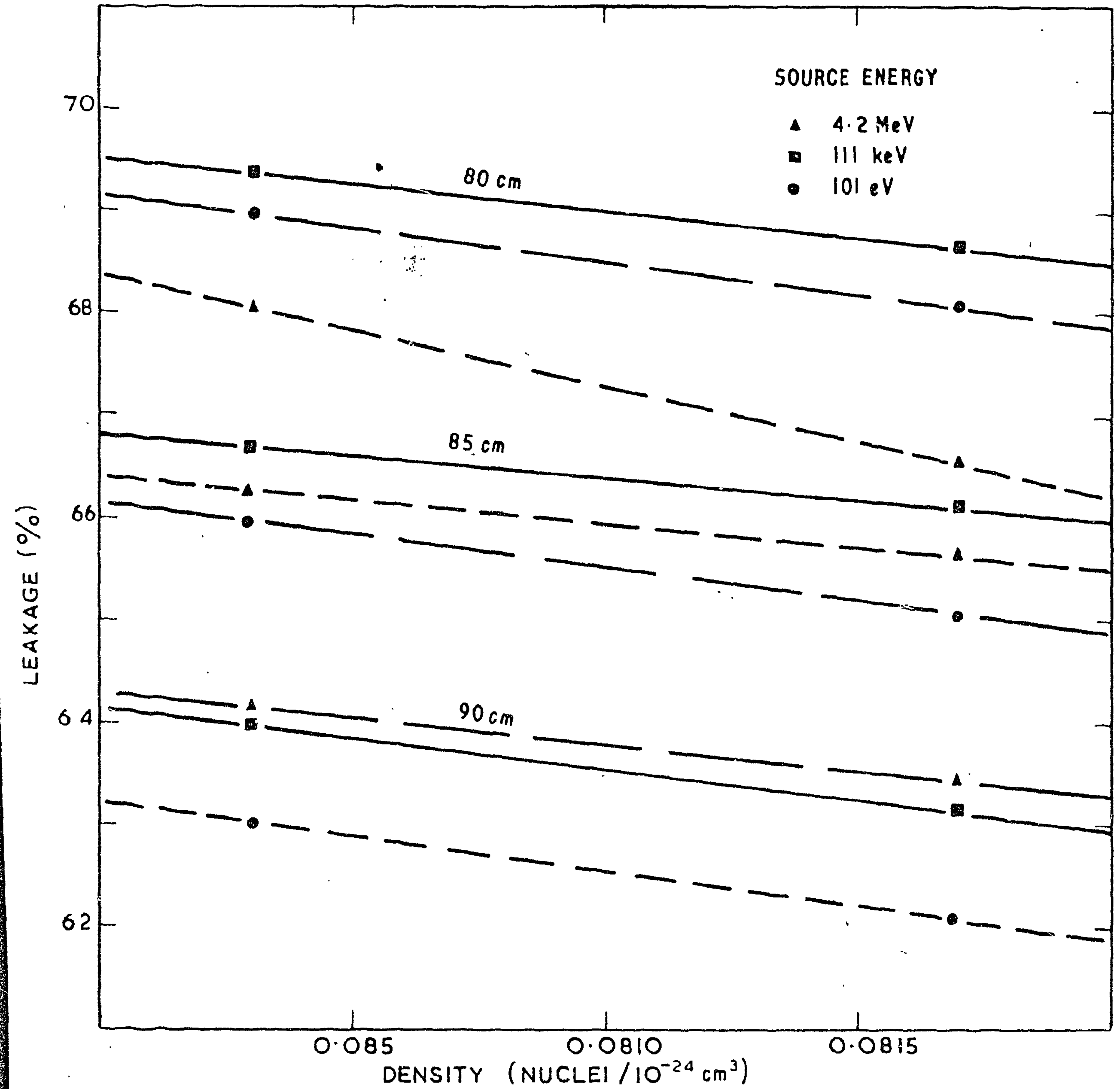

FIGURE 2. VARIATION OF LEAKAGE WITH DENSITY FOR GRAPHITE CYLINDERS OF THREE DIFFERENT RADII

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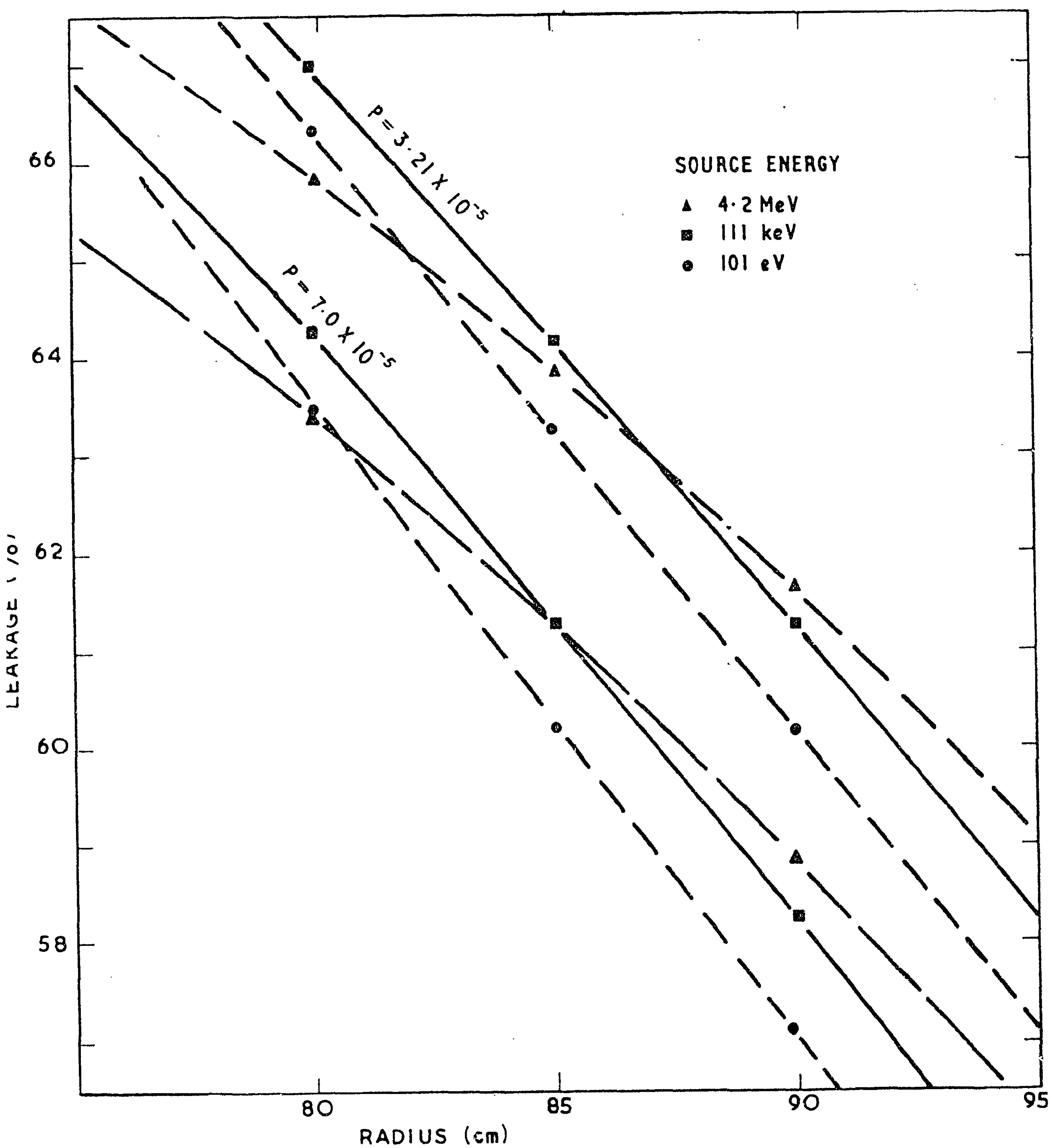

FIGURE 3. VARIATION OF LEAKAGE WITH RADIUS OF A GRAPHITE CYLINDER (DENSITY 1.60 g cm^{-3}) CONTAINING SMALL CONCENTRATIONS OF POISON

P1204

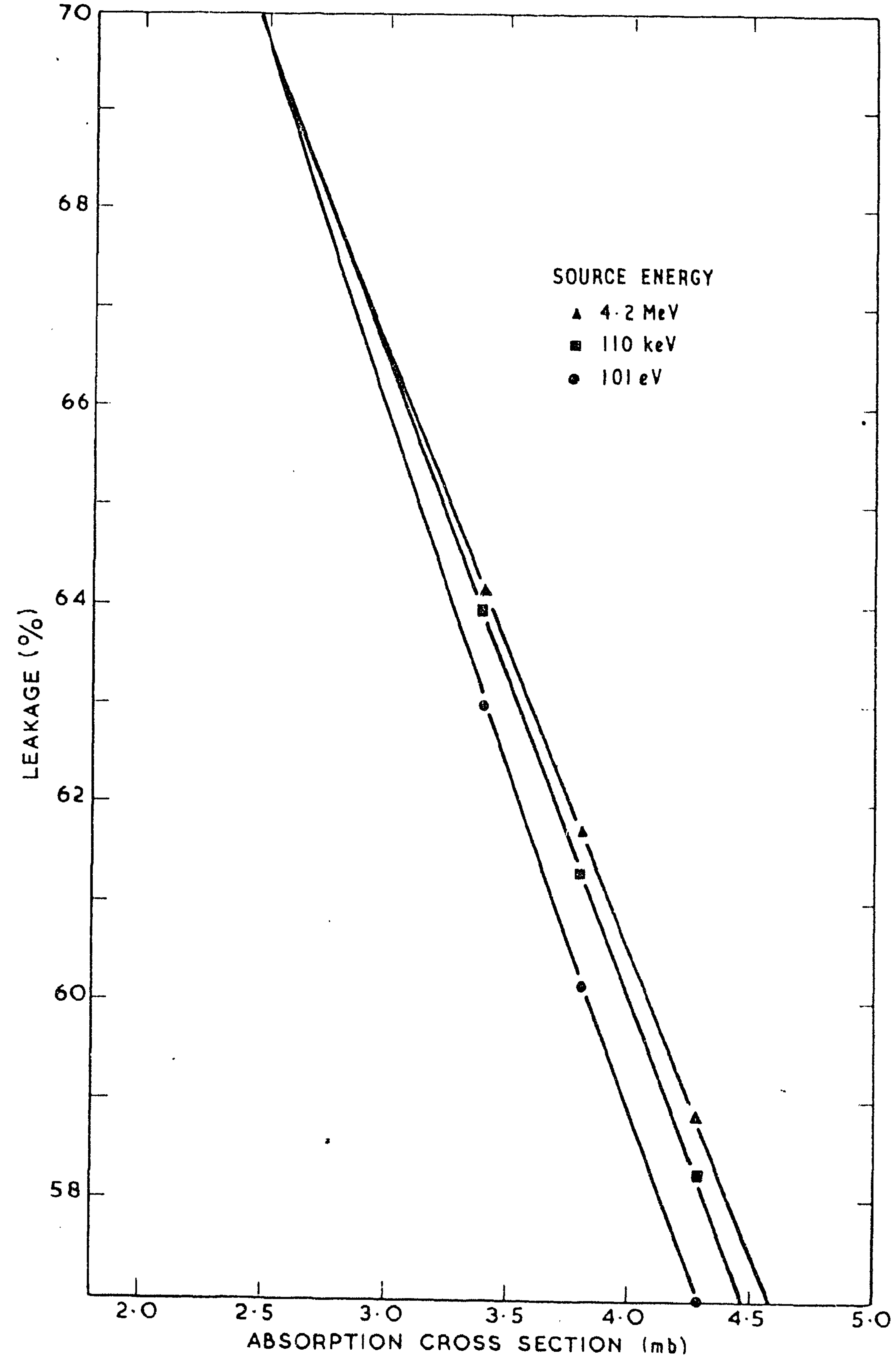

FIGURE 4. VARIATION OF LEAKAGE WITH EFFECTIVE 1/v ABSORPTION CROSS SECTION FOR A GRAPHITE CYLINDER (DENSITY 1.60 g cm^{-3}, RADIUS 90cm)

P1204

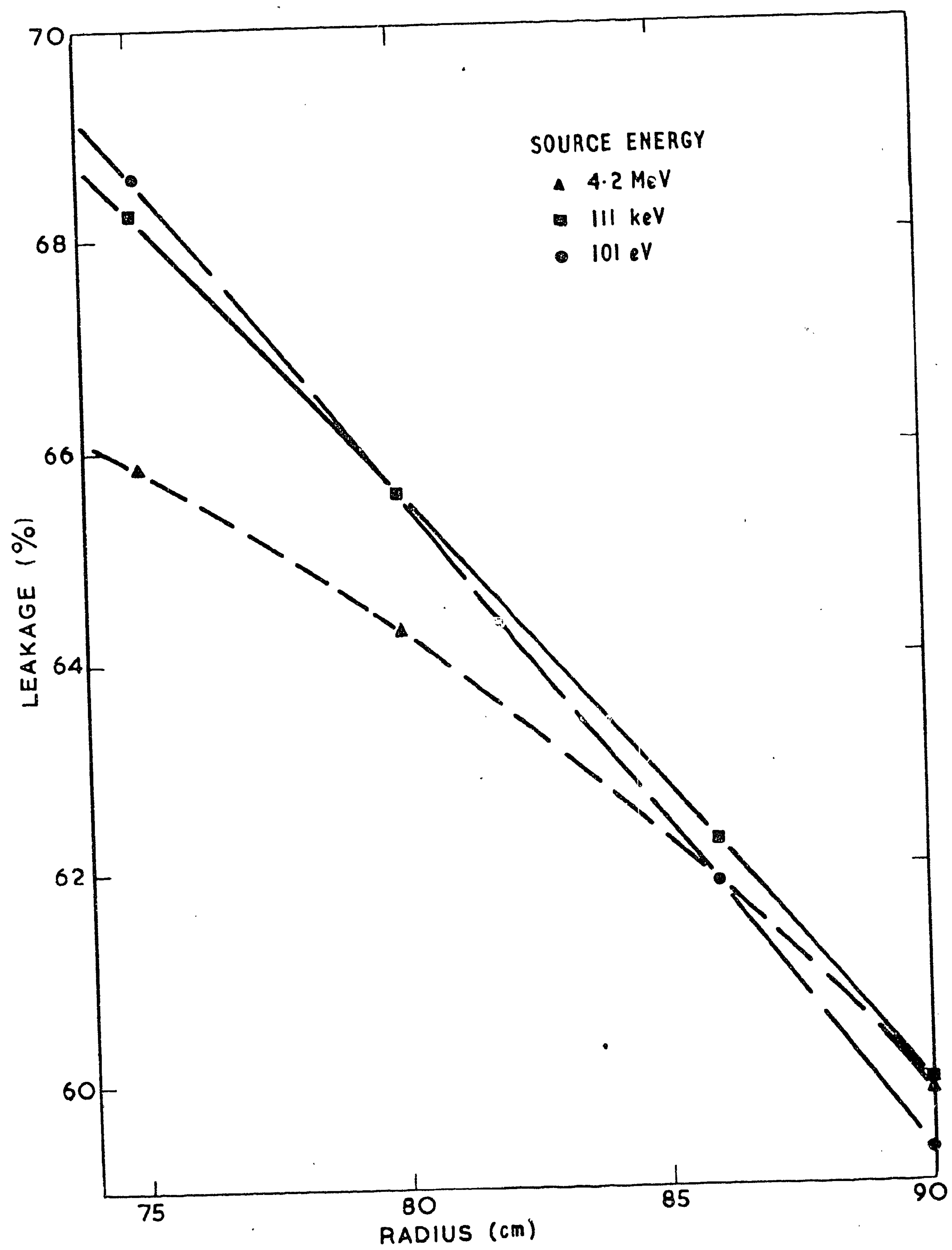

FIGURE 5. VARIATION OF LEAKAGE WITH RADIUS FOR A GRAPHITE CYLINDER (DENSITY 1.63 g cm^{-3}) CONTAINING A NON-1/v ABSORPTION CROSS SECTION MATERIAL

P1204

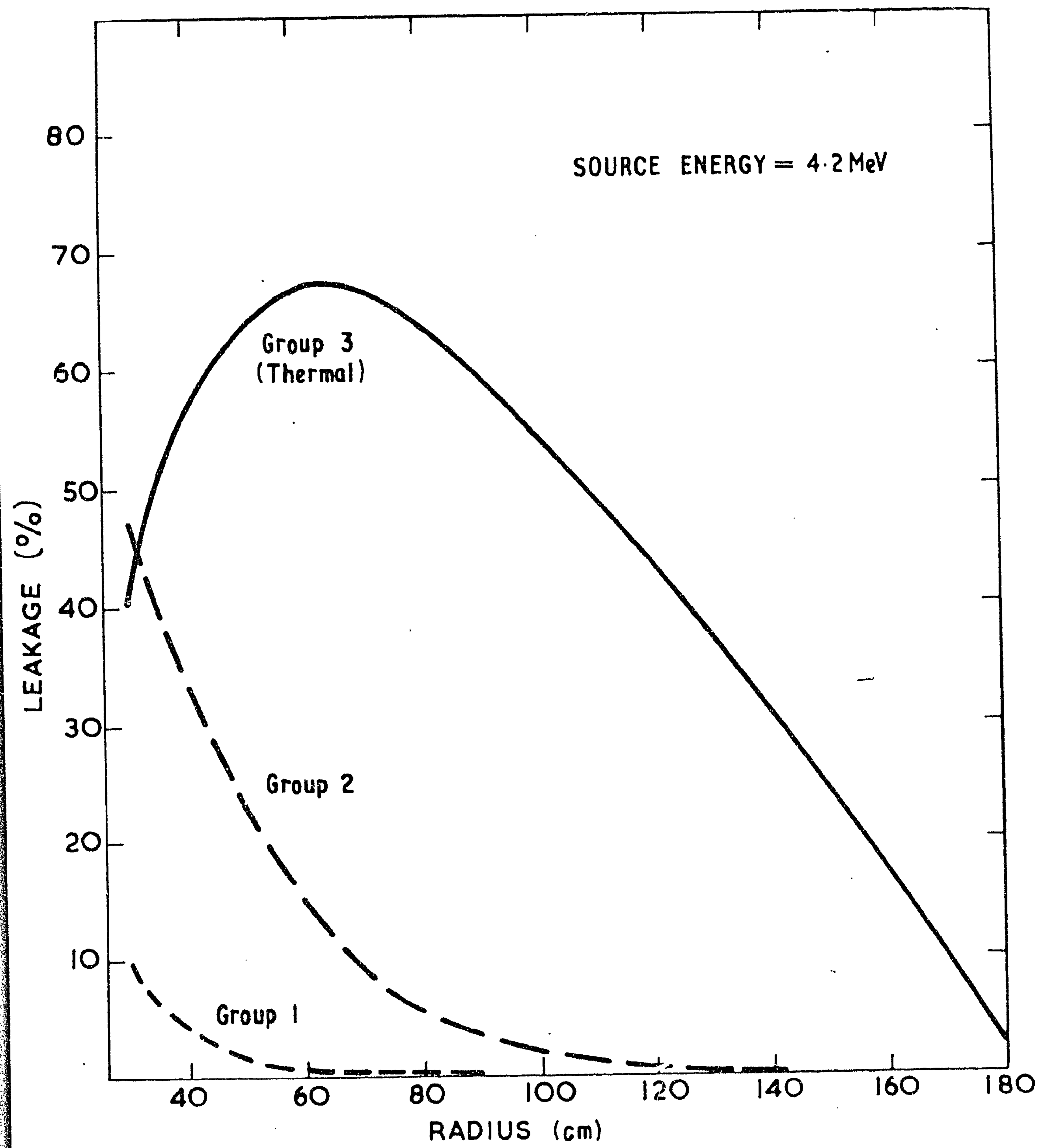

FIGURE 6. REDUCTION IN LEAKAGE WITH INCREASE IN RADIUS OF A GRAPHITE CYLINDER (DENSITY 1.60 g cm^{-3})

P1204

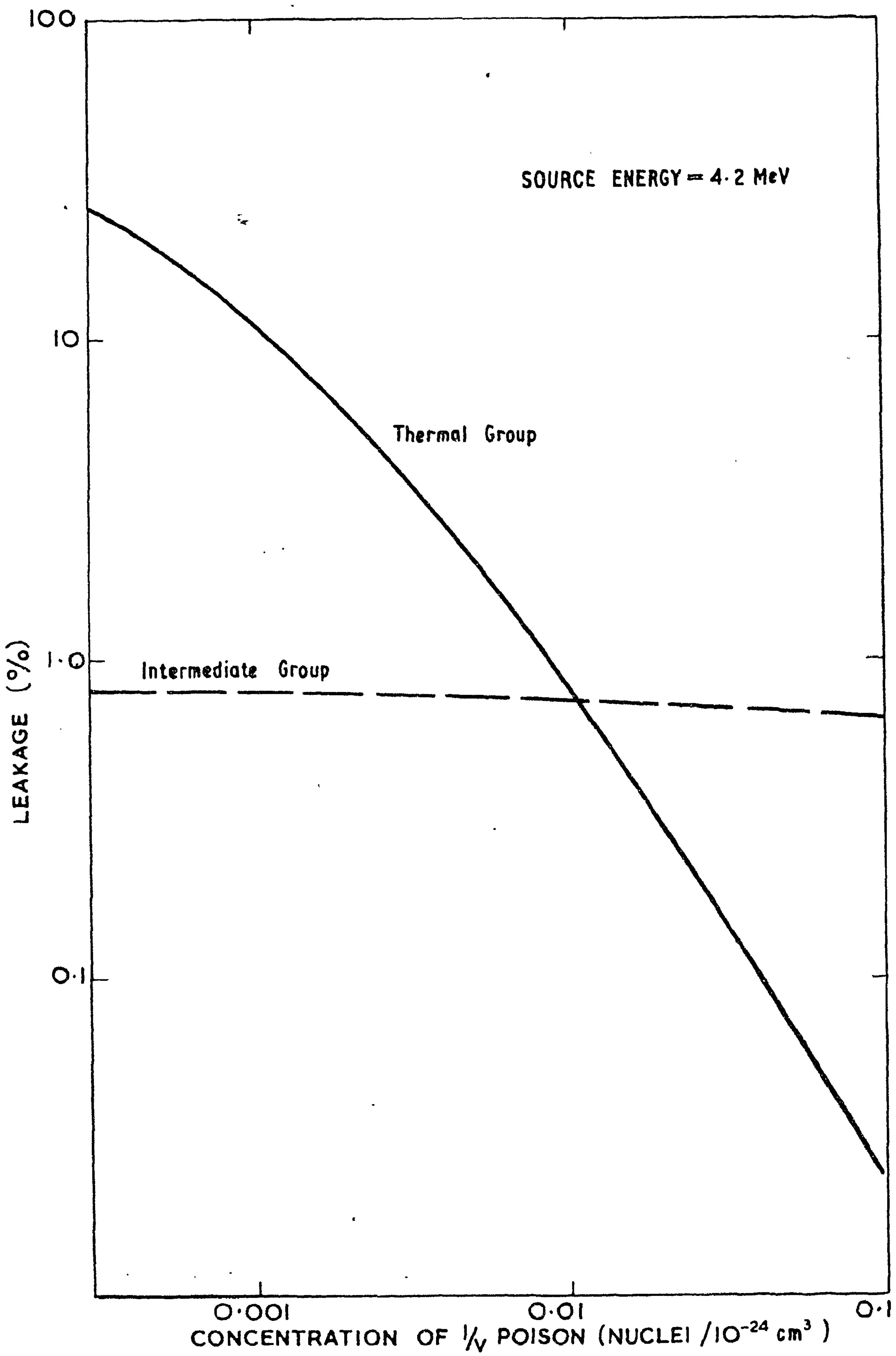

FIGURE 7. VARIATION OF LEAKAGE WITH POISON CONCENTRATION FOR A GRAPHITE CYLINDER (DENSITY 1.60 g cm^{-3} RADIUS 120 cm)

P1204

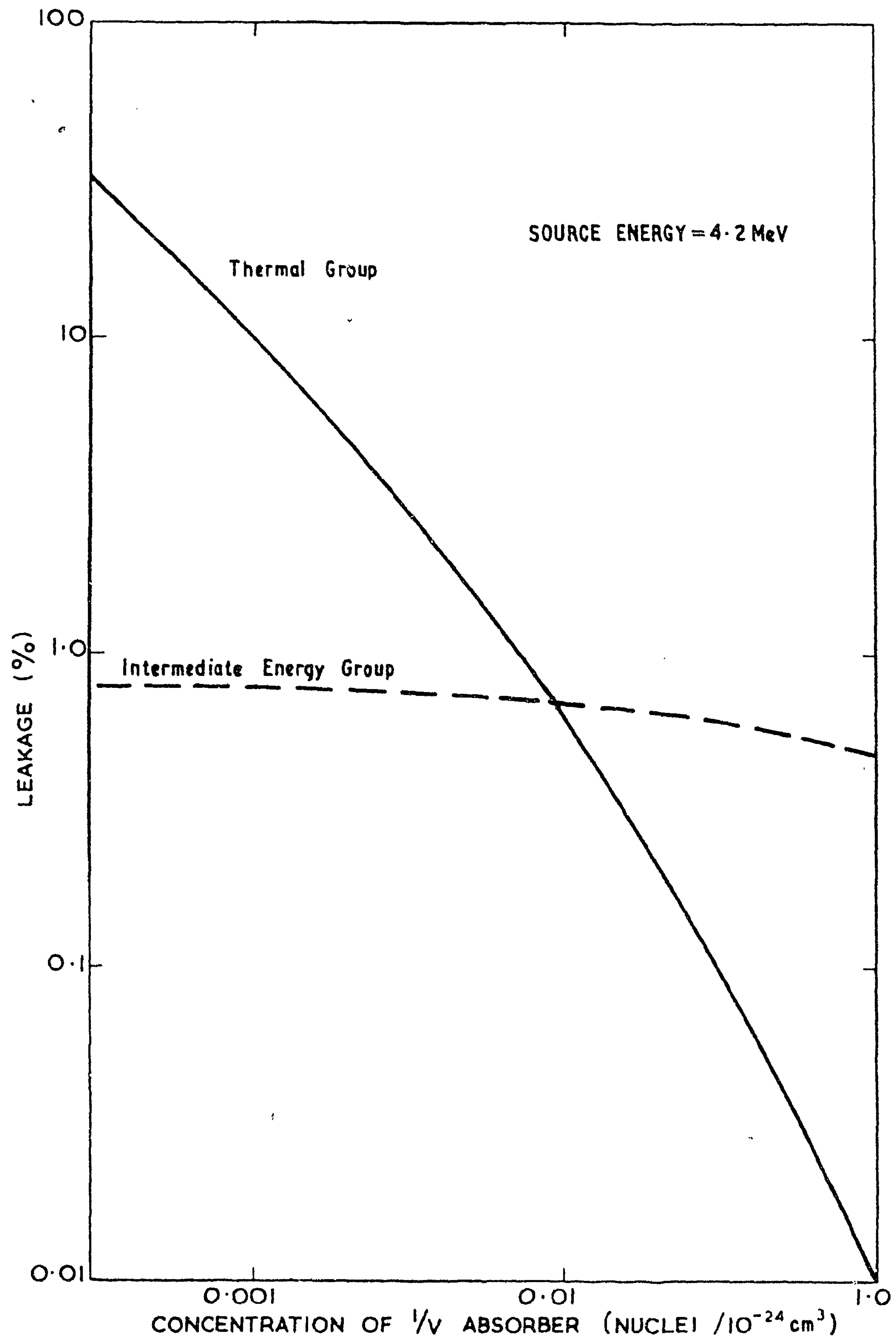

FIGURE 8.

VARIATION OF LEAKAGE WITH POISON CONCENTRATION FOR A GRAPHITE CYLINDER (DENSITY 1.60 g cm^{-3} RADIUS 120 cm) CONTAINING A NON-1/v ABSORPTION CROSS SECTION MATERIAL

P1204

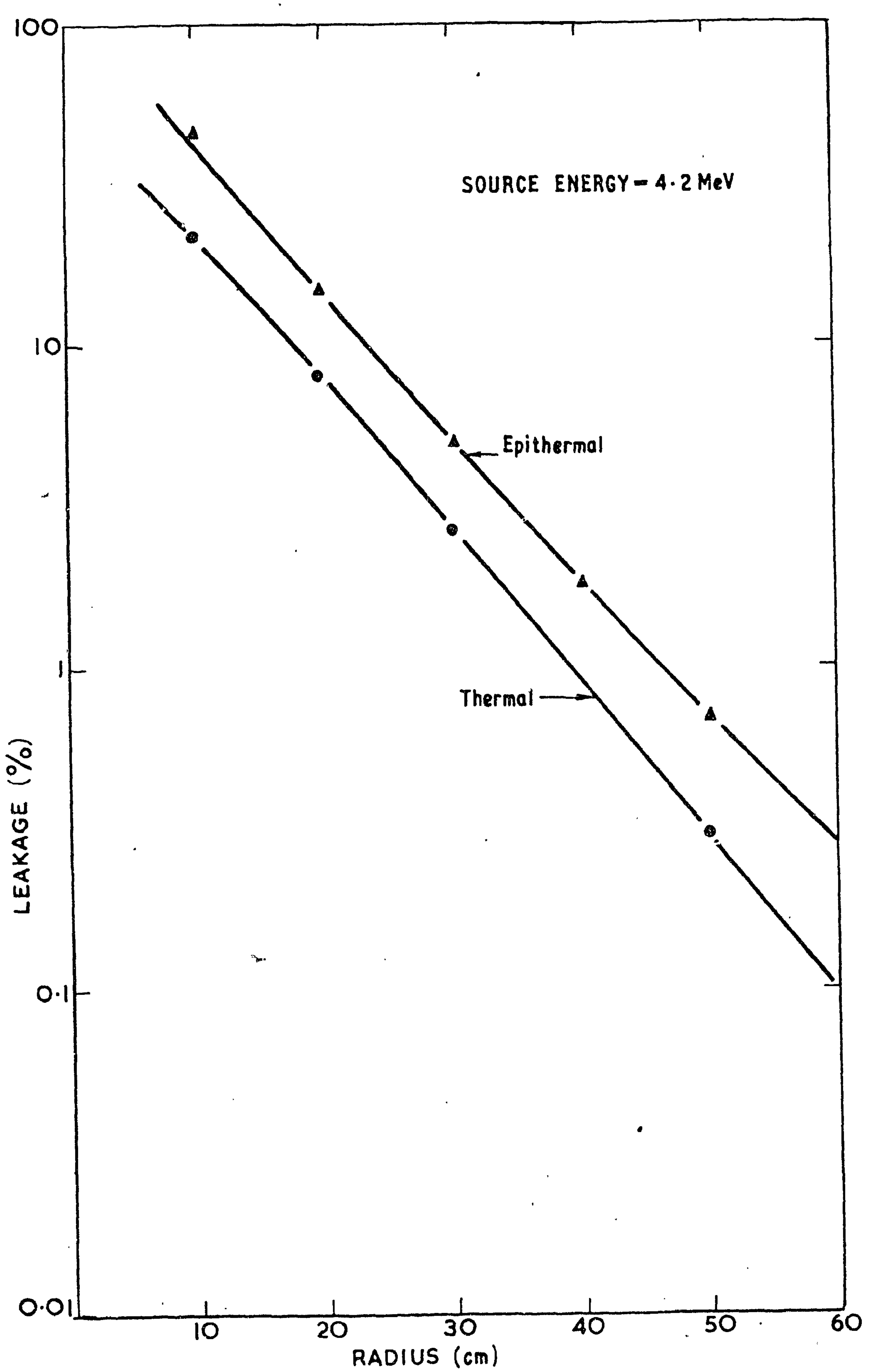

FIGURE 9. VARIATION OF LEAKAGE WITH RADIUS FOR A WATER CYLINDER

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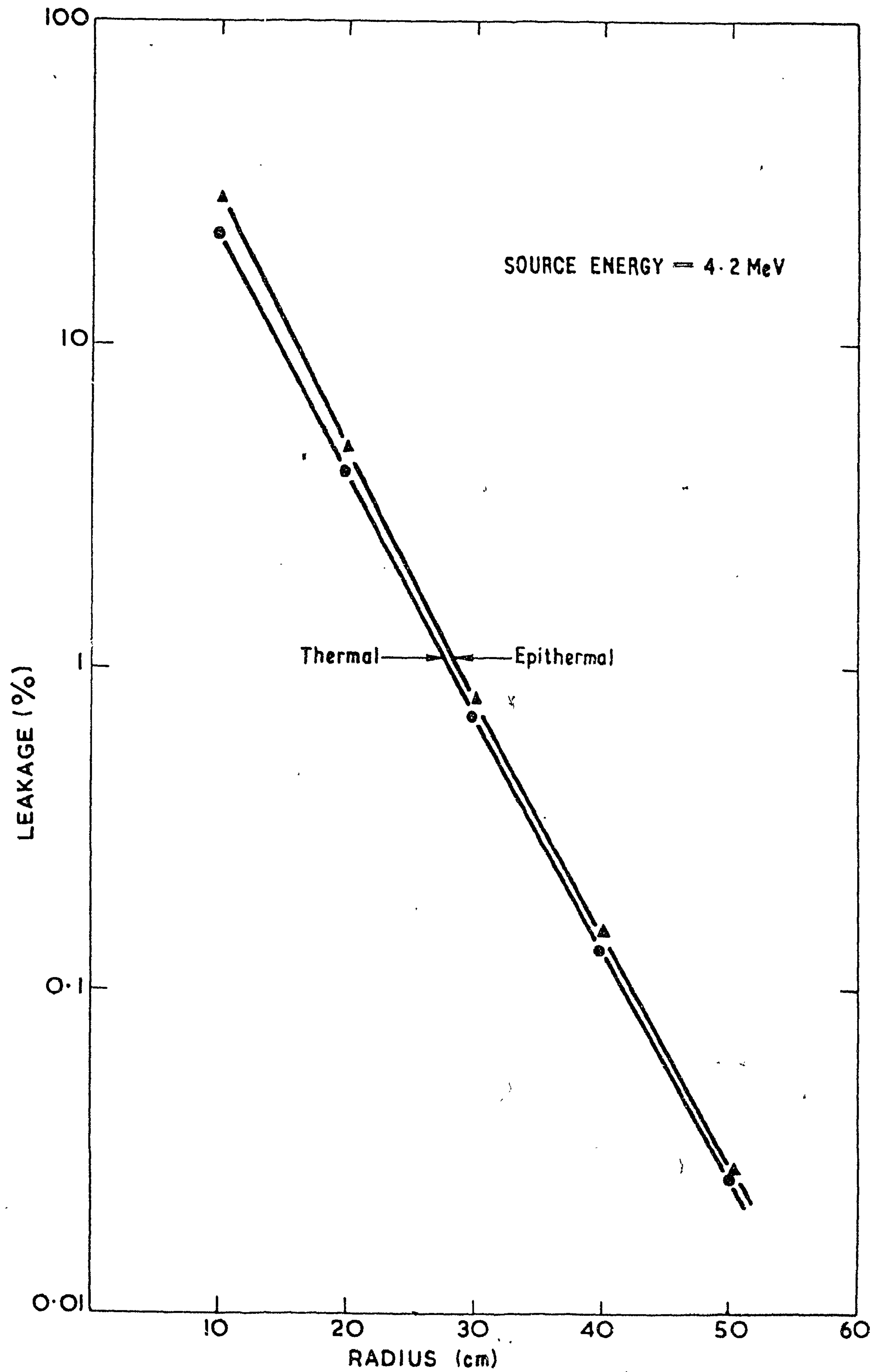

FIGURE 10. VARIATION OF LEAKAGE WITH RADIUS FOR PARAFFIN WAX CYLINDER

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