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« ABSORBED ENERGY IN THERMOLUMINESCENT POWDER EMBEDDED IN A BINDER »

P. BASSI, G. BUSUOLI, L LEMBO* et O. RIMONDI**

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SUMMARY

The energy absorbed in the grains of a thermoluminescent powder embedded in a solid binder was evluated. The calculations were made with the Howarth method and by means of a computer. The influence of the various physical, geometric parameters (in particular the grain diameter) on which the absorbed energy depends, has been shown. The following cases evaluated : CaF_2 and lithium borate powders embedded in silicone rubber or telfon, in various percentages. The calculations referred to CaF_2 powder embedded in silicone rubberwere experim entally controlled too.

RÉSUMÉ

L'énergie absorbée par un grain de poudre thermoluminescente, enrobée par un liant solide a été évaluée à l'aide d'un ordinateur en adoptant la méthode de calcul de HOWARTH.

On a montré l'influence des paramètres physiques et géométriques (en particulier le diamètre des grains) sur l'énergie absorbée.

Les évaluations ont été effectuées dans les cas suivants :

Fluorure de calcium et borate de lithium en proportions variées, noyées dans un liant de caoutchouc aux silicones ou en téflon.

Les calculs relatifs à la poudre de fluorure de calcium ont été vérifiés expérimentalement.

I - INTRODUCTION

Thermoluminescent powders incorporated in a solid binder are frequently employed in dosimetry of electromagnetic radiation. The binder causes a change in the response of the powder. This research evaluates the influence of the various parameters that affect the response as a function of energy, of the embedd-

* Laboratorio Fisica Sanitaria, C.N.E.N., Centro Calcolo-Bologna (Italia)
** Instituto di Fisica dell' Universita - Bologna (Italia)

ed powder i.e. the atomic composition of the powder and binder, the respective percentages by weight and the grain size of the powder.

II - CALCULATION PROCEDURE

As is well know the energy dependence [1] of a medium is expressed by :

$$f = \frac{(\mu_{en}/\varrho) \text{ medium}}{(\mu_{en}/\varrho) \text{ air}}$$

Where (μ_{en}/ϱ) medium and $(\mu en/\varrho)$ air are the mass energy-absorption coefficients of the medium and air respectively, f depends on the atomic composition of the medium, which may be one chemical compound or homogeneous mixture of two or more compounds (with the respective molecules distributed homogeneously). The energy dependance f of pure CaF_2 (curve *a*), of pure silicone rubber (curve *d*) and of two homogeneous mixtures (curve *b* and *c*) are shown in fig. 1. The silicone rubber RTV-615 employed is produced by the G.E.Co., and it has the following composition by weight : C = 31.3 %, N = 7.3 %, O = 21.9 %, Si = 38.7 %.

In the case of a thermoluminescent powder incorporated in a binder the f value calculated for the homogeneous mixture cannot give the energy absorbed in the powder grains. This has to be evaluated with other calculation techniques [2] [3] for instance with the method of J.L. HOWARTH [4] (*). The energy absorbed in a grain may be subdivided into two components : one imparted by the electrons generated in the binder and entering the grains, the second imparted by the electrons arising in the same grains. When the grain size increases, in comparison with the electron range, the first component becomes much lower than the second. Obviously, by keeping constant the grain diameter and by varying the radiation energy, we have the former or the latter situation.

The Howarth method gives the dose distribution inside a spherical grain. The distribution function [4] is expressed by :

$$h(x) = 1 + \Sigma_i K_i G(T_i, x)$$

where

$$K_i = \frac{1}{\mu_{en}/\varrho} \left[\frac{(\mu'_{en}/\varrho)i}{m^s} - (\mu_{en}/\varrho)i \right]$$

The meanings of the symbols is given in the Appendix.

$h(x)$ is the ratio between the dose actually absorbed at a point P, which has a distance x from the spherical grain surface and the dose there would be at P, if the grain were surrounded by the same substance that the grains are made of.

The sum is extended to any group i of electrons having initial energy T_i; G_i is a geometrical factor that depends on T_i and x; K_i is a function of the mass energy absorption coefficients of the binder and grains.

(*) In Appendix we give a summary of the Howarth method.

The mean value $\bar{h}$ of $h(x)$ is expressed by : $\bar{h} = \frac{1}{D}\int_0^D H(x)\,dx$ and, within a grain, gives the ratio between the energy actually absorbed in a grain embeddep in the binder and the energy that a grain would absord if it were surrounded, not by the binder but by the same substance that the grains are made of.

The $\bar{h}$ values have been calculated by supposing that the grains are spherical. When the mean distance between the grain is shorter than the electron range, the electrons emerging from a grain may reach the neighbours. This situation may be approximated by supposing, for ease of calculation, that the grains are embedded in a homogeneous mixture made ot the same substance as the powder and the binder. From now on we shall call this case " altered binder " to distinguish it from the case " pure binder " where the grains are supposed to be very far from one another.

To make the calculation easier, the order of magnitude of the mean distance between the grains may be evaluated [5] by supposing that the grains are not spherical but cube-shaped and arranged at the lattice points of a single cubic space lattice of side L. For instance in the case of CaF_2 embedded in silicone rubber (5 % and 95 % by weight respectively) it results $L \simeq 8$ D, where D is the grain diameter. The $\bar{h}$ values calculated for monodisperse powders of CaF_2 embedded in an " altered binder " (silicone rubber plus CaF_2) are shown in fig. 2.

The $\bar{h}$ values are relative to that at 1 MeV.

The parameter D is the diameter of the powder grains. The larger the diameter, in comparison with the range of the electrons, the more the ratio $\bar{h}$ approaches the unit value, *i.e.* the response of the loose powder. It must be noted that the approximation of the parameters employed in the calculations for CaF_2 brings about $\bar{h}$ values a little higher than the unit value [6].

Other calculations for various kinds of powders an binders were made. The f values of teflon, teflon plus BaF_2 and of silicone rubber, silicone rubber plus lithium borate are shown in figs. 3 and 4 respectively. The h values for CaF_2 embedded in teflon are shown in fig. 5 (case of " pure binder ") and in figs. 6 and 7 (case of " altered binder "). Figures 8, 9 and 10 are referred to calculations for the lithium borate powder embedded in silicone rubber.

III - EXPERIMENTAL TEST OF SOME CALCULATIONS

The authors carried out an experimental test of some calculations referred to CaF_2 powder, 5 % by weight, embedded in silicone rubber. The preparation method is described in a previous paper [7]. The detectors are disc-shaped 7.5 mm in diameter and 0.9 mm thick.

The powder grains have a mean weight diameter $\bar{D} = 2/\mu m$. $\bar{D}$ is defined by [8] :

$$\bar{D} = \sqrt[3]{\frac{\Sigma_i\, n_i\, D_i^3}{\Sigma_i\, n_i}}$$

where n_i is the number of particles with diameter D_i. The incorporated powder was extracted from the commercial one by means of calibrated sieves. The powder is not monodispersed and the size distribution n_i was mesured on microphotographs by means of a Zeiss particle analyszer TGZ3 [9]. From the size distribution n_i and from the complete set of the curves $\bar{h}$ (part of which are shown in fig. 2) it was possible to calculate the weighted mean $\bar{h}_g$ for a certain energy of the radiation :

$$\bar{h}_g(E) = \frac{1}{m} \Sigma_i \, m_i \, \bar{H}_i(E)$$

where E is the energy of the incident radiation and m_i is the total mass of the particles with a diameter D_i, $m = \Sigma_i \, m_i$; $\bar{H}_i$ is the ordinate of the curve having the parameter D_i. The product $\bar{h}_g(E) \times f$ represents the calculated response of the detectors.

The detectors were exposed to Co-60 and to filtered X-rays. The characteristics of the beams are shown in Tab. I. The exposure measurements are accurate to within a few percent [10].

TABLE I

FILTERED X-RAYS

Excit. Voltage (KV)	Current (mA)	Filter (mm)	H.V.L. (mm)	Effective Energy (KeV)
37	10	4.0 Al	1.6 Al	26
85	10	8.0 Al	5.8 Al	42.5
120	10	1.0 Cu + 1.0 Al	0.51 Cu	61
150	10	1.63 Cu + 1.0 Al	0.97 Cu	77

Before irradiation the detectors were annealed at 300 °C for ten minutes. After irradiation, before the reading, the detectors were kept at 10 °C for 10 min. The TL reader was a Harshaw mod. 2000.

TABLE II

RESPONSE OF 5 % BY WEIGHT CaF_2 POWDER EMBEDDED IN SILICONE RUBBER

$\overline{D} = 2 \ \mu m$

Eff. Energy (keV)	Exper. Response per Roentgen	$\bar{h}_g(E) \times f$	$\bar{h}_g(E)$	f for CaF_2
A	B	C	D	E
26	9.1 ± 10 %	7.1	0.49	14.4
42.5	5.5 ± 10 %	4.4	0.35	12.5
77	2.5 ± 10 %	2.0	0.38	5.25
Co^{60}	1.0 ± 6 %	1.0	1.00	1.00

The maximum reading temperature was 300 °C and the heating time 60 sec. The experimental results are compared with the calculated values in Table II.

The experimental values (column B) are normalized to that of Co-60; the error is the standard deviation of the ratio. Column E shows the energy dependence of CaF_2. The $\bar{h}_g$ values are shown in column D. Column C shows the product of the values of column D and column E.

TABLE III

RESPONSE OF 30 % BY WEIGHT LiF POWDER EMBEDDED IN SILICONE RUBBER

$\overline{D} = 6\ \mu m$

Eff. Energy (keV)	Exper. Response per Roentgen	$\bar{h}_g(E) \times f$	$\bar{h}_g(E)$	f for LiF
A	B	C	D	E
26	1.9 ± 12 %	1.7	1.28	1.29
42.5	2.1 ± 12 %	2.0	1.63	1.22
61	1.9 ± 12 %	1.8	1.60	1.13
77	1.7 ± 13 %	1.5	1.36	1.07
Co^{60}	1.0 ± 10 %	1.0	1.00	1.00

In table III other results obtained in a previous work [5] are shown. In that case *LiF* powder was embedded in silicone rubber. The experimental results of both tables confirm the calculations with good approximation.

IV - CONCLUSIONS

As been shown the mean weight diameter of the powder is a determining parameter for the dependence of the response as a function of energy of the embedded grains. From the practical point of view if we want to keep this response similar to the one of the loose powder, the embedded grains must have a diamater larger than 70-80/μm.

The satisfactory agreement between the experimental results and the calculations confirms the practical utility of the Howarth method for the dose calculation in transition zones. For this reason calculations for other kinds of embedded powders are shown. All the calculations have been made with the computer. The computer program is given in another report [11].

ACKNOWLEDGEMENT

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APPENDIX

SUMMARY OF THE HOWARTH METHOD

INTRODUCTION

J.L. HOWARTH presented a simplified method for the calculation of the absorbed dose in soft-tissue cavities is bone [4]. The method takes into account the variation of *LET* with electron energy, the continuous energy spectrum of the Compton electron, and the energy transferred to the medium by Auger electrons.

We used the same method for the calculation of the absorbed dose in a thermoluminescent powder embedded in a binder. In the following exposition we use the same symbols used by HOWARTH in his paper, in particular : the symbols that he used for soft-tissue and bone here are employed for the TL powder and the binder respectively.

CONTRIBUTION TO THE ABSORBED DOSE IN A GRAIN OF A TL POWDER, FROM THE ELECTRONS ARISING IN THE BINDER

Consider spherical grains of diameter D embedded in a binder. The distance between the grains is supposed larger than the electron range. Let n_0 secondary electrons, each with initial energy T_0, and corresponding streight-line range R, be generated per unit volume in a grain and let n_0' be the number of electrons, each with the initial energy T_0, generated per unit volume of the binder. Consider a point P inside a grain at a distance X from the nearest point on the spherical interface. Assuming a relation of the form $R = AT^m$ between the range R and energy T of the secondary electrons, CHARLTON and CORMACK [12] derived an expression equivalent to the following :

$$E_b(x) = \frac{n_0' T_0}{s} G \qquad (1)$$

where $E_b(x)$ is the contribution of the electrons arising in the binder to the energy absorbed per unit volume at P, s is the ratio (assumed constant) of the electron range in grains to that in the binder and G a geometrical factor, wich is x a function of $\frac{x}{R_0}$ and $\frac{D}{R_0}$.

Calculation of the Total Absorbed Dose at P

Now suppose that the binder in replaced by the same material of the TL powder. In this case, by comparison with equation (1), the contribution from the external region of the spherical interface, would be $n_0 T_0 G$. But with this replacement, we would have a uniform medium, and electronic equilibrium

would exist, so that the total absorbed energy per unit volume at P, would be $n_o T_o$.

The contribution $E_t(x)$ to the absorbed dose at P from electrons arising inside the grain can be expressed with :

$$E_t(x) = n_o T_o (1 - G) \tag{2}$$

and the total absorbed energy per unit volume at P :

$$E(x) = E_b(x) + E_t(x) = n_o T_o + \left(\frac{n'_o T_o}{s} - n_o T_o \right) G \tag{3}$$

The equilibrium value of $E(x)$ (in the absence of the binder) is $n_o T_o$, so that the ratio of the energy absorbed per unit volume to the equilibrium energy absorbed per unit volume is :

$$b(x) = 1 + \left(\frac{n_o T_o}{n'_o T_o} \cdot \frac{1}{s} - 1 \right) G = 1 + \left(\frac{\mu_{en}}{\mu'_{en}} \cdot \frac{1}{s} - 1 \right) G \tag{4}$$

Where μ_{en} and μ_{en} are the linear energy absorption coefficient for the binder and the TL powder respectively. Thus :

$$b(x) = 1 + \left(\frac{\mu'_{en}/\varrho}{\mu_{en}/\varrho} \cdot \frac{1}{m^s} - 1 \right) G \tag{5}$$

Where μ'_{en}/ϱ and μ_{en}/ϱ are the corresponding mass energy absorption coefficients, and m^s is the ratio of the mass stopping powers for the binder and the powder.

The quantity $b(x)$ is a direct measure of the number of times the absorbed dose is increased by the presence of the binder.

By multiplying $b(x)$ by the ratio f of the equilibrium absorbed dose in the powder to the exposure, we way obtain the absorbed dose at P under non equilibrium conditions.

In the above analysis it has been assumed that electrons of only one energy, T_o, are produced. If electrons of a number of discrete energies are produced, the calculation must be made separately for each group.

For electrons of group i, we have, corresponding to equation (3) :

$$E_i(x) = (n_o T_o)_i + \left[\frac{(n'_o T_o)_i}{s} - (n_o T_o)_i \right] G_i \tag{6}$$

where $(n'_o T_o)_i$ and $(n_o T_o)_i$ are the total energy of group i electrons produced per unit volume of the binder and of the powder, respectively, and G_i is the value of G for the electrons of this range.

Thus, corresponding to equation (4), the ratio of the part of the absorbed dose at P due to group i electrons alone to the total equilibrium absorbed dose is :

$$H_i(x) = \frac{E_i(x)}{E_{eq}} = \frac{(n_o T_o)_i}{n_o T_o} + \left[\frac{(n'_o T_o)_i}{n_o T_o} \frac{i}{s} - \frac{(n_o T_o)_i}{u_o T_o} \right] G_i =$$

$$= \frac{(\mu_{en})_i}{\mu_{en}} + \frac{1}{\mu_{en}} \left[\frac{(\mu'_{en})_i}{s} - (\mu_{en})_i \right] G_i$$

where $(\mu'_{en})_i$ and $(\mu_{en})_i$ are the components for production of group i electrons of the linear energy absorption coefficients for the binder and the powder respectively. Thus :

$$h_i(x) = \frac{(\mu_{en}/\varrho)_i}{\mu_{en}/\varrho} + \frac{1}{\mu_{en}/\varrho}\left[\frac{(\mu'_{en}/\varrho)_i}{m^s} - (\mu_{en}/\varrho)_i\right] G_i \tag{7}$$

where $(\mu'_{en}/\varrho)_i$ and $(\mu_{en}/\varrho)_i$ are the components of the mass energy absorption coefficients for group i electrons for the binder and the powder, respectively.

Summing over all the groups of electrons, the ratio of the total absorbed dose at P to the equilibrium absorbed dose in the powder is given by :

$$h(x) = 1 + \Sigma_i K_i G_i \tag{8}$$

where

$$K_i = \frac{1}{\mu_{en}/\varrho}\left[\frac{(\mu'_{en}/\varrho)_i}{m^s} - (\mu_{en}/\varrho)_i\right] \tag{9}$$

and

$$\mu_{en}/\varrho = \Sigma_i (\mu_{en}/\varrho)_i$$

The form of the eq. (8) makes relatively easy to take account of Auger electrons as well as photoelectrons and to allow for the continuous range of energy of the Compton electrons.

In the calculation of K_i for the photoelectrons (*), the photoelectric energy energy absorption coefficient has been reduced by a fraction corresponding to the average farction of the photon energy which goes to Auger electrons. It has not, however, been reduced to allow for the energy that appears as fluorescence radiation. It is assumed that the energy to the fluorescence radiation is distributed in the same way as that of the photoelectrons.

With these simplifications and those relative to Compton electrons, equation (8) takes the form :

$$h(x) = 1 + \widehat{K}_\tau G_\tau + K_A G_A + K_C \bar{G}_C \tag{10}$$

where

$$\widehat{K}_\tau = \frac{1}{\mu_{en}/\varrho}\left[\frac{\tau'/\varrho\,(1-\alpha')}{m^s} - \tau/\varrho\,(1-\alpha)\right] \tag{11}$$

$$K_A = \frac{1}{\mu_{en}/\varrho}\left[\frac{\tau'/\varrho\,\alpha'}{m^s} - \tau/\varrho\,\alpha\right] \tag{12}$$

τ/ϱ and τ'/ϱ are the total photoelectric absorption cross sections per unit mass of powder and binder. G_τ and G_A are the values of the geometrical factors for electrons with ranges corresponding to the average energy of photoelectrons and average Auger electron energy; α and α' are the average fraction of the photon energy appearing as energy of an Auger electron per photoelectric inter-

(*) It will be assumed that only K photoelectrons are produced and a simple average energy will be assigned to them.

action. $\bar{G}$ is a suitable weighted mean of the function G over the range of energies of the Compton electrons :

$$K_C = \frac{e\sigma_a}{\mu_{en}/\varrho}\left(\frac{N'}{m^s} - N\right)$$

where $_e\sigma_a$ is the Compton absorption cross section per electron; N, N' the numbers of electrons per unit mass of powder and binder.

In order to select the appropriate value of the geometrical factor G_τ for the photoelectrons, it is necessary to choose a value for the initial photoelectron energy form which R_o may be calculated.

A considerable simplification of equation (10) can also made in handling of the Auger electron term. At distances from the spherical interface greater than the Auger electron range ($\sim$ 0.3 μ) $G_A = 0$ and the Auger term disappears.

If the diameter of the interface is large compared with 0.3 μ, that is $D/_{Ro}$ is large for the Auger electrons, the interface is close to being a plane surface and G may be taken as 0.5 for the Auger electrons. This leads to the expressions :

$$H(x) = 1 + \widehat{K}_\tau\, G_\tau + K_C\, \bar{G}_C \qquad x > 0.3\ \mu$$

$$H(o) = 1 + \widehat{K}_\tau\, G_\tau(o) + 0.5\ K_A + K_C\, \bar{G}_C$$

Calculation of the Physical Parameters

The parameters K_τ, K_A and K_c have been calculated by using the experimentally determined total photoelectric cross sections tabulated by Storm and Israel [6]; Klein-Nishina cross sections are taken from Attix and Roesch [13].

The value of m^s was calculated with the expression given by K.Z. MORGAN [14].

Values of the K fluorescence yield, F_K, and of the fluorescence (or Auger) energy $\bar{E}_K$ were taken from Storm and Israel [6]; α and α' (equations 11 and 12) were calculated by averaging the value of $\bar{E}_K$ $(1 - F_K)/h\nu$, where $h\nu$ is the photon energy, weighted in proportion to the photoelectric cross section, over the appropriate atomic compositions.

In order to test the computer program, the marrow dose in spherical bone cavity 50 μ in diameter has been calculated. The results obtained for a 50 Kev electromagnetic radiation is 2.4 rad/R. F.W. SPIERS gives 2.28 rad/R [2] for the same situation. The slight difference is probably due to the different choice of the parameters employed.

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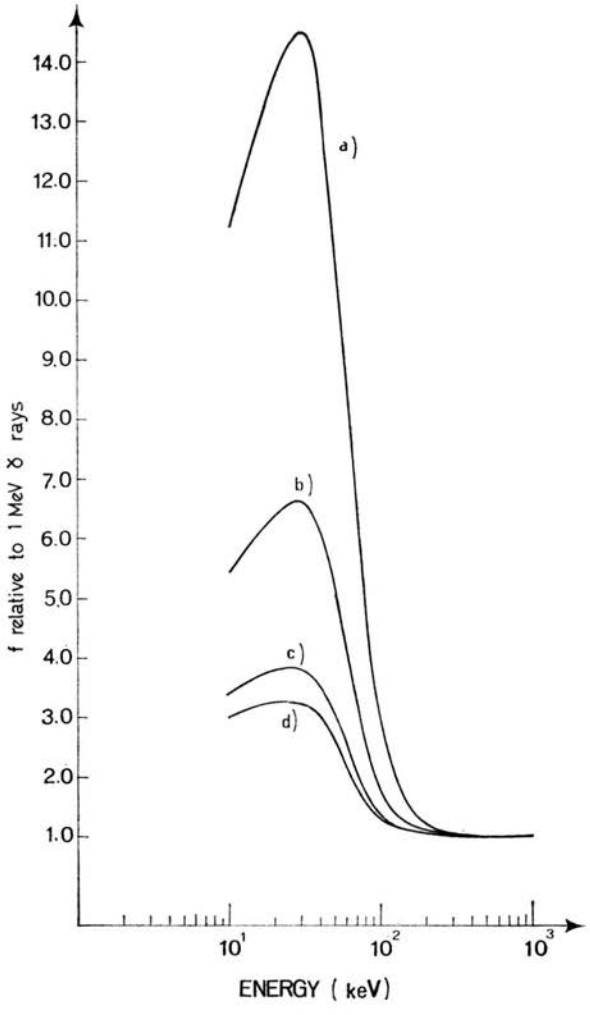

FIG. 1. — Calculated energy dependence $f = \frac{(\mu_{en}/\varrho)\ \text{medium}}{(\mu_{en}/\varrho)\ \text{air}}$ for CaF_2 (curve *a*), for silicone rubber (curve *d*), for homogeneous mixture of silicone rubber plus 5 % or 30 % by weight of CaF_2 (curve *c* and *b* res pectively)

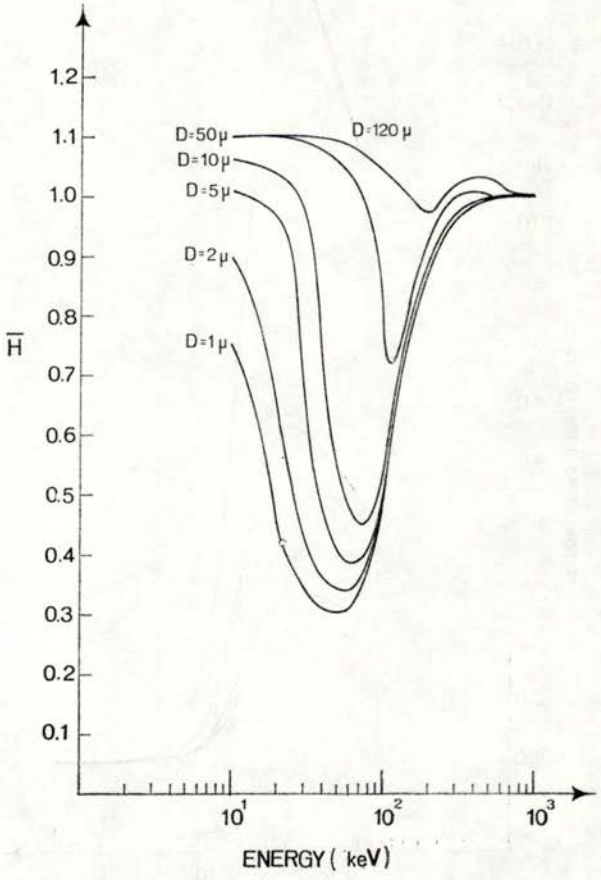

FIG. 2. — $\overline{H}$ values calculated for CaF_2 embedded in " altered binder " (5 % CaF_2 plus 95 % silicone rubber)

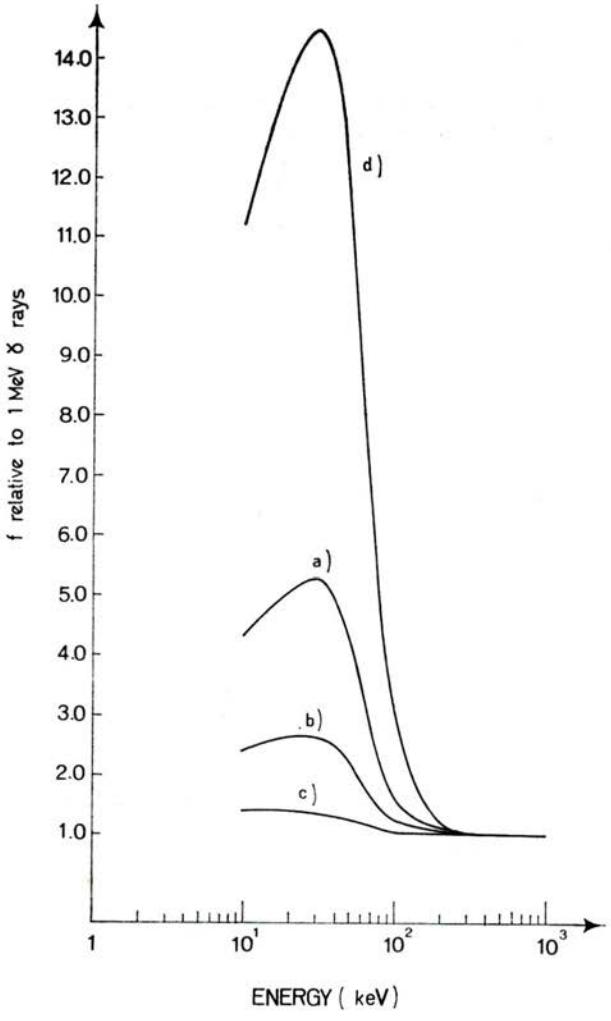

FIG. 3. — f values calculated for teflon (curve c), for homogeneous mixture of teflon plus 10 % or 30 % by weight of CaF_2 (curve c and a respectively), for CaF_2 (curve d).

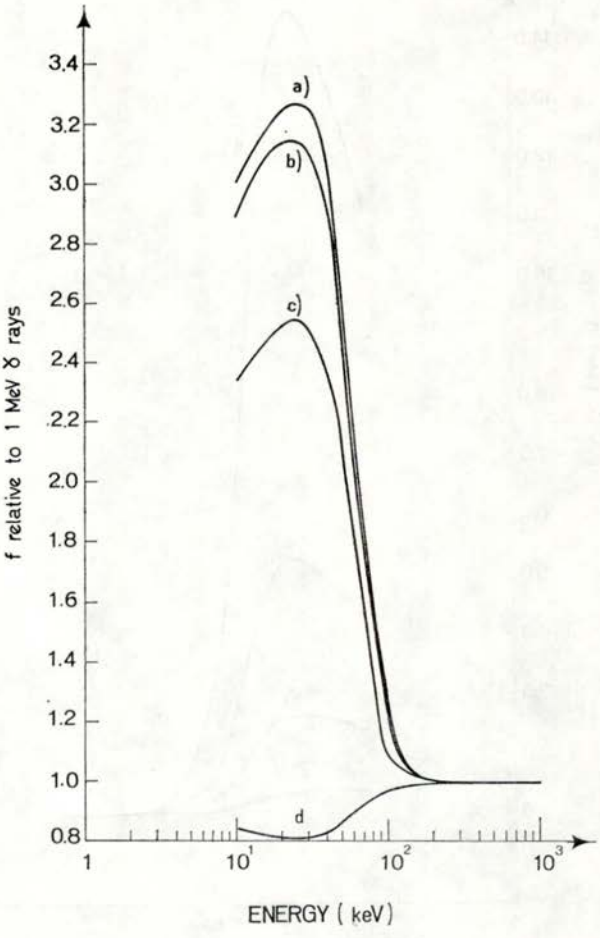

FIG. 4. — f values calculated for lithium borate (curve d), for homogeneous mixture of silicone rubber plus 5 % or 30 % by weight of lithium borate (curve b and c respectively), and for silicone rubber (curve a).

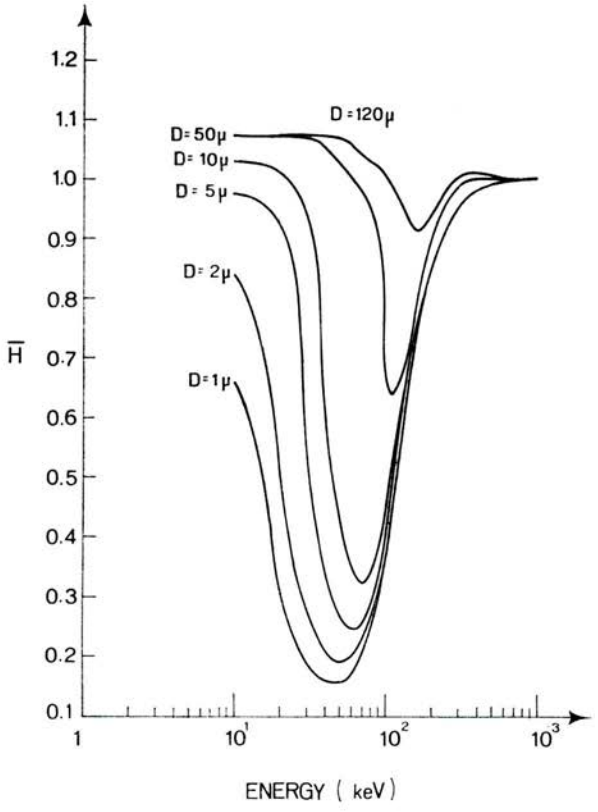

FIG. 5. — $\overline{H}$ calculated for CaF_2 embedded in " pure binder " (teflon)

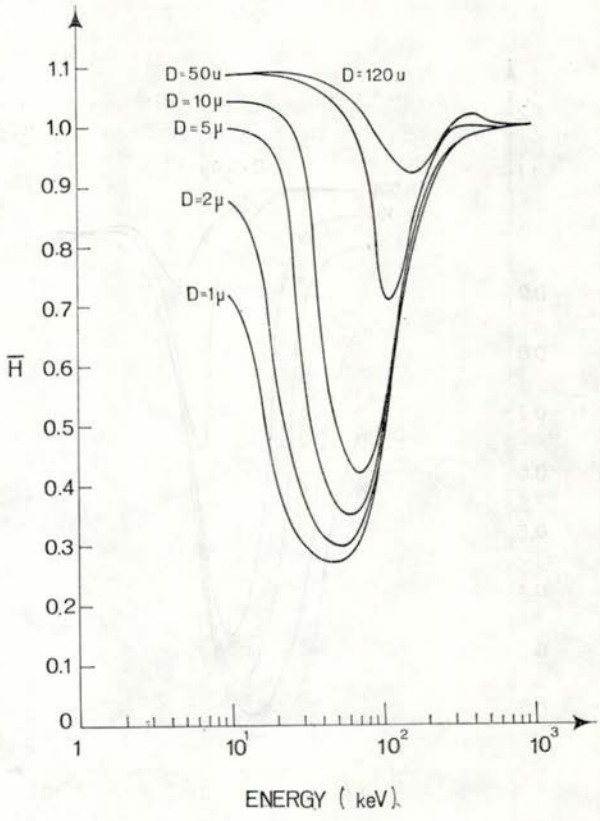

FIG. 6. — $\overline{H}$ calculated for CaF_2 embedded in "altered binder" (teflon plus 10 % CaF_2 by weight).

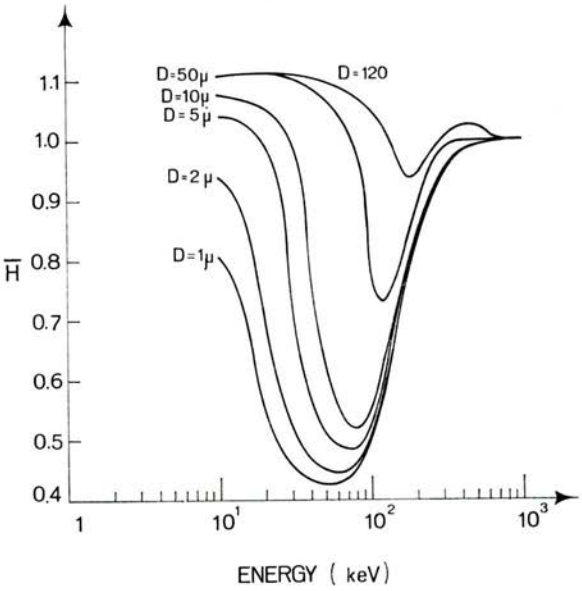

FIG. 7. — $\overline{H}$ calculated for CaF_2 embedded in " altered binder " (teflon plus 30 % CaF_2 by weight).

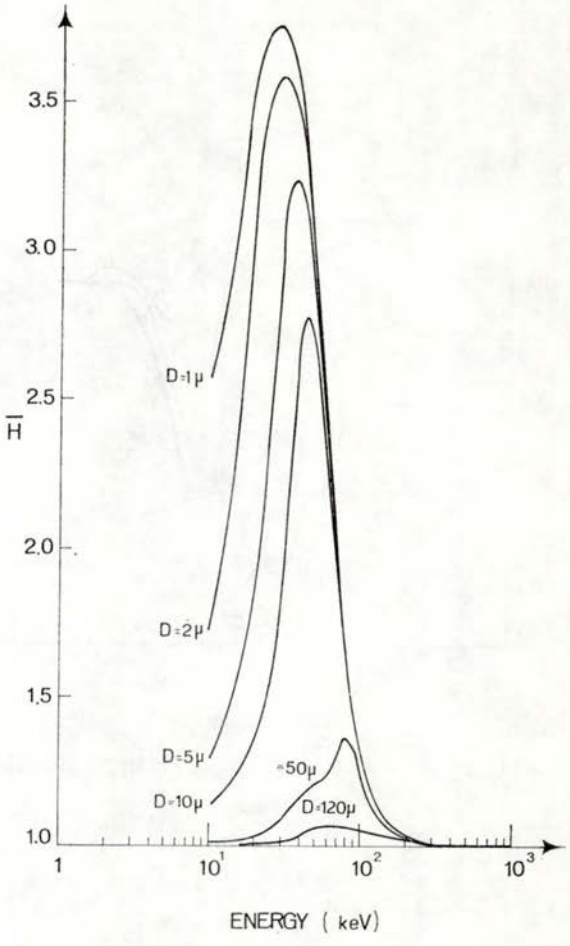

FIG. 8. $\overline{H}$ calculated for lithium borate embedded in "pure binder" (silicone rubber)

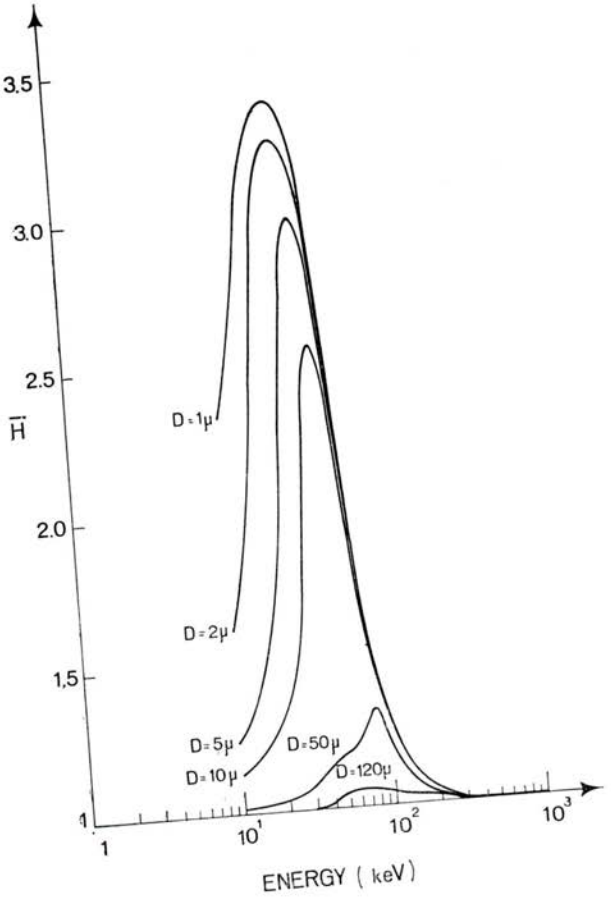

FIG. 9. $\overline{H}$ calculated for lithium borate embedded in " altered binder " (silicone rubber plus 5 % lithium borate by weight).

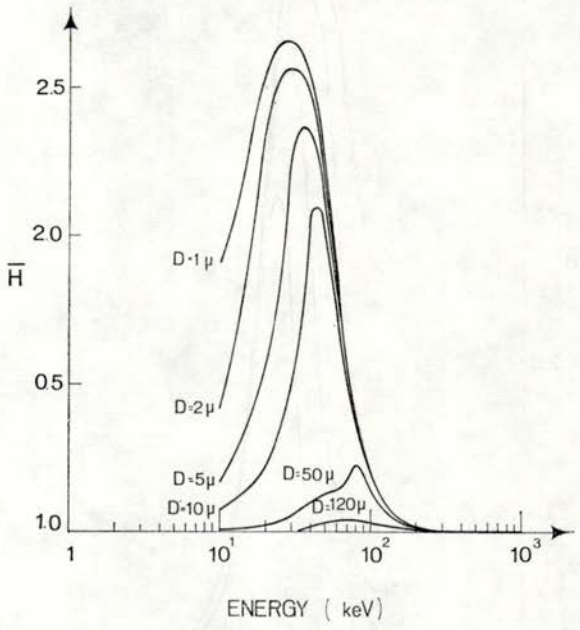

FIG. 10. — $\overline{H}$ calculated for lithium borate embedded in "altered binder" (silicone rubber plus 30 % lithium borate by weight).