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ANALYSIS OF RESPONSE DATA FOR SEVERAL ORGANIC SCINTILLATORS

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Data on the scintillation response (light output) of anthracene, stilbene, NE-213, NE-102 and Pilot-B to electrons and protons and of NE-230 to electrons and deuterons reported recently by Smith et al.[1]) have been fitted by the method of least squares to the parametric formula

$$L(E) = S\int_0^E d\varepsilon[1 + kB(dE/dx) + C(dE/dx)^2]^{-1}$$

where E is the initial particle energy, dE/dx is the specific energy loss, and S, kB and C are the adjustable parameters. Required values of dE/dx were computed from available data[16,17]). Scintillation efficiencies S were deduced from the electron data for which $dE/dx \approx 0$. One-parameter (kB variable, $C=0$) and two-parameter (kB and C variable) fits for the heavy-particle data were obtained and compared with other results reported in the literature.

1. Introduction

Organic scintillators are widely used for detecting nuclear radiations. Different types of scintillators – crystals (e.g. anthracene), liquids (e.g. NE-213) and plastics (e.g. NE-102) – are commercially available and have been developed for various applications. Measurements of the response of these scintillators to different radiations are of practical importance because of the technological applications which have evolved. Furthermore, response data must be available in order to make progress toward understanding scintillation mechanisms.

Smith et al.[1]) recently reported the results of response measurements on six commonly used organic scintillators: anthracene, stilbene, NE-213, NE-230, NE-102 and Pilot B. The objective of the present work is to fit these data using a parametric formula based on theoretical work by Birks[2]) and Chou[3]). These fitted curves can be used for extrapolation of the scintillator response to particle energies for which experimental data are not available. Certain adjustable parameters of the formula have physical interpretations, and their "best-fit" values may provide insight into the scintillation mechanisms.

Scintillators respond directly to ionization generated by charged particles. Neutral radiations (e.g. gamma rays and neutrons) are detected if they produce recoil electrons or recoil nuclei within the scintillator material. A relatively small fraction of the ionization energy lost by a charge particle in a scintillator is converted into flourescent light energy. The rest of the energy is dissipated non-radiatively, mainly as heat[2]). The flourescence is characteristic of the molecular structure of the scintillator. Organic scintillators generally exhibit a fast (2–30 nsec) decay component. An additional slow (several μsec) decay component is observed for some organic scintillators, and several of these scintillators posses pulse-shape discrimination properties[1,2]). The scintillation efficiency, fraction of ionization energy converted to flourescent light energy, differs for each type of scintillator and also depends on the type of charged particle producing the ionization. Electrons generate more light than do heavy particles (e.g. protons and ^{4}He particles) of equal energy when stopped in organic scintillators. The response to electrons is linear for particle energies above $\approx$ 125 keV [2,4]); however, the response to heavier particles is nonlinear up to much higher energies. It is the objective of scintillator response theories to explain these and other observed phenomena.

Birks[2]) has derived a formula which has been extremely successful in explaining many of the features of the experimental data for organic scintillators. His treatment assumes that high ionization density along the particle track is responsible for the quenching effect which is observed for protons and other heavily ionizing particles. The quenching of the primary ionization energy is believed to occur within a time that is short compared with the flourescent decay times ($<$ 1 nsec). The following expression is obtained:

$$dL/dx = S(dE/dx)\{1 + kB(dE/dx)\}^{-1}, \qquad (1)$$

where

dL/dx = flourescence light energy emitted per unit path length;

dE/dx = specific energy loss for the charge particle (a function of particle type and energy);

S = scintillation efficiency.

In this theory, the molecules along the ionization column are grouped into two categories labeled

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"damaged" and "undamaged" for convenience. The ratio of the number of "damaged" molecules to the number of "undamaged" molecules is assumed to be $B\,\mathrm{d}E/\mathrm{d}x$ and k is the corresponding relative quenching probability. The molecules labeled "damaged" are those with the greater capacity for dissipating the ionization energy nonradiatively (quenching). Those molecules labeled "damaged" actually occupy highly excited or ionized states, and they de-excite quickly (< 1 nsec) to the "undamaged" condition. Some permanent damage will occur, and this can contribute to long-term deterioration of the properties of the scintillator, but it is not responsible for the quenching effect being considered.

The product kB is treated as a single adjustable parameter since there is no means available at present for measuring k or B separately. Chou[3]) proposed a modified form of eq. (1) with one additional adjustable parameter

$$\mathrm{d}L/\mathrm{d}x = S(\mathrm{d}E/\mathrm{d}x)\,[1+kB(\mathrm{d}E/\mathrm{d}x)+C(\mathrm{d}E/\mathrm{d}x)^2]^{-1}. \tag{2}$$

Eq. (2) approaches eq. (1) for small values of $\mathrm{d}E/\mathrm{d}x$. There have been several other theoretical studies of scintillation mechanisms and quenching in organic scintillators which have yielded formulas for $\mathrm{d}L/\mathrm{d}x$, ref.[5-14]). These formulas also reduce to eq. (1) for small values of $\mathrm{d}E/\mathrm{d}x$. Since scintillation processes in organic scintillators are still not entirely understood, these formulas should be considered as semiempirical.

Eq. (2) can rewritten in the form

$$\mathrm{d}L/\mathrm{d}E = S[1+kB(\mathrm{d}E/\mathrm{d}x)+C(\mathrm{d}E/\mathrm{d}x)^2]^{-1}, \tag{3}$$

where $\mathrm{d}L$ is the quantity of flourescence light generated when a charged particle with energy E loses a quantity of energy $\mathrm{d}E$ through ionization along a path increment $\mathrm{d}x$ within the scintillator. The integral of eq. (3) with respect to particle energy is

$$L(E) = S\int_0^E \mathrm{d}\varepsilon\,[1+kB(\mathrm{d}E/\mathrm{d}x)+C(\mathrm{d}E/\mathrm{d}x)^2]^{-1}, \tag{4}$$

where $L(E)$ is the total light emitted by a charge particle which expends all of its energy E within the scintillator. For electrons with energies $\geqslant$ 125 keV, $\mathrm{d}E/\mathrm{d}x \approx 0$ and eq. (4) reduces to the linear expression

$$L_e(E) \approx SE + L_0. \tag{5}$$

2. Analytic procedure

The data of Smith et al.[1]) have been fitted with the formulas in eqs. (4) and (5) using the method of differential corrections[15]). A computer program was developed to search for the best-fit parameters. The adjustable parameters were chosen to minimize the quantity

$$\chi^2 = \sum_i [\{L(E_i)-L_i\}/\Delta L_i]^2, \tag{6}$$

where $L_i \pm \Delta L_i$ is the measured value (including experimental uncertainty) of the response for particle energy E_i and $L(E_i)$ is computed from eq. (4) for heavy particles or from eq. (5) for electrons. The summation in eq. (6) extends over the available data points. Smith et al.[1]) measured the electron- and heavy-particle responses under identical conditions so that for a particular scintillator, the same value of the scintillation efficiency parameter S applies for both the electron- and heavy-particle data. Smith et al.[1]) expressed the measured scintillator responses on a relative scale for which the value $L_e = 1000$ was assigned arbitrarily to the response of each scintillator to 0.525-MeV electrons. Because of this imposed normalization, there is actually only one adjustable parameter S in eq. (5), and this equation may be written as

$$L_e(E) = S(E-0.525)+1000. \tag{7}$$

The parameters S for each scintillator were extracted from the fits of eq. (7) to the electron data. Two independent fits were obtained to the heavy-particle data for each of the six scintillators studied. First, the best one-parameter [Birks formula[2]): kB variable, $C = 0$] fits to the experimental points were determined for each of the scintillators. Next, the best two-parameter [Chou formula[3]): kB and C variable] fits were obtained.

The specific energy losses $\mathrm{d}E/\mathrm{d}x$ required for this numerical work were computed from atomic stopping number data published by Whaling[16]) and by Hirschfelder and Magee[17]). The main constituents of the organic scintillators studied are carbon and hydrogen (deuterium for NE-230) with negligible traces of other elements. The following formula was used to compute the specific energy losses for protons (or deuterons)

$$\mathrm{d}E/\mathrm{d}x = \{4\pi e^4/(m_0 V^2)\}(N_H B_H + N_C B_C), \tag{8}$$

where

e = electron charge;
m_0 = electron rest mass;
V = velocity of the particle;
B_H = atomic stopping number for hydrogen (or deuterium);

TABLE 1
Computed specific energy losses for protons in several organic scintillators.

Proton energy (MeV)	dE/dx(MeV·cm^2·g^{-1})				
	NE-102	NE-213	Pilot B	Stilbene	Anthracene
0.005	290.5	291.4	290.4	284.0	281.3
0.010	524.2	528.7	523.9	510.4	508.0
0.015	730.5	736.9	730.1	711.6	707.5
0.02	901.1	908.0	900.6	879.4	875.8
0.03	1047.7	1054.9	1047.3	1025.1	1021.4
0.04	1081.5	1088.5	1081.0	1051.8	1055.7
0.05	1073.2	1079.9	1074.8	1052.1	1048.7
0.10	894.4	901.3	894.0	878.2	876.1
0.11	869.8	876.3	869.5	854.6	852.7
0.12	845.3	851.3	845.0	831.0	829.4
0.13	820.8	826.4	820.4	807.5	806.1
0.14	802.0	807.3	801.7	789.5	788.4
0.15	778.7	783.7	778.5	766.9	765.9
0.16	756.7	761.5	756.4	745.3	744.4
0.17	735.7	740.2	735.4	725.1	724.3
0.18	715.9	720.2	715.7	705.8	705.2
0.19	698.4	702.5	698.2	688.9	688.3
0.20	676.9	680.9	676.7	667.7	667.2
0.25	598.7	601.9	598.5	591.2	591.0
0.30	533.9	536.6	533.7	528.5	527.4
0.35	494.4	496.3	494.2	488.9	489.0
0.40	450.3	452.3	450.2	445.7	446.0
0.45	418.6	420.4	418.6	414.5	414.7
0.50	386.7	388.4	386.7	383.0	393.2
0.55	364.7	366.1	364.6	361.4	361.7
0.60	339.4	340.8	339.3	336.1	336.4
0.70	307.0	308.1	306.9	308.2	304.8
0.80	280.4	281.4	280.3	278.0	278.3
0.90	259.1	260.0	259.1	257.0	257.4
1.00	238.1	238.9	238.0	236.2	236.6
1.10	224.3	227.6	226.9	225.3	222.7
1.20	211.5	212.1	211.5	210.0	210.3
1.30	201.4	202.0	201.3	200.0	200.2
1.40	190.3	190.0	190.3	189.1	189.4
1.50	180.8	181.2	180.7	179.6	180.0
1.60	174.5	175.0	174.5	173.4	173.8
1.70	167.6	168.0	167.6	166.6	167.0
1.80	160.0	160.4	160.0	159.0	159.4
1.90	154.7	155.0	154.6	153.8	154.1
2.00	148.4	148.8	148.4	147.6	147.9
2.50	127.0	127.3	127.0	126.3	126.6
3.00	110.3	110.5	110.2	109.7	110.0
3.50	97.3	97.6	97.3	96.9	97.1
4.00	88.1	88.2	88.1	87.7	87.0
4.50	79.0	79.2	79.0	78.6	78.9
5.00	73.7	73.8	73.7	73.2	73.6
5.50	68.1	68.2	68.0	67.8	68.0
6.00	62.5	62.7	63.0	62.3	62.4
7.00	55.5	55.6	55.5	55.3	55.4
8.00	50.1	50.2	50.1	49.9	50.5
9.00	45.3	45.8	45.7	45.5	45.6
10.00	41.9	42.0	41.9	41.8	41.9
11.00	38.9	39.0	38.9	38.8	38.9
12.00	36.3	36.4	36.3	36.2	36.3
13.00	34.0	34.1	34.0	33.9	34.0
14.00	32.1	32.1	32.1	31.9	32.0
15.00	30.3	30.4	30.3	30.2	30.3

N_H = number of hydrogen (or deuterium) atoms/g;
B_C = atomic stopping number for carbon;
N_C = number of carbon atoms/g.

The H:C and D:C ratios were obtained from the manufacturers' specification sheets*. The computed specific energy losses for protons in anthracene, stilbene, NE-213, NE-102 and Pilot-B are presented in table 1 whereas the corresponding values for deuterons in NE-230 appear in table 2.

Uncertainties in the atomic stopping numbers contribute to the overall uncertainty of the fitting procedure in a complicated way. The quantities B_H and B_C are uncertain by $\pm 25\%$ at low energies (< 100 keV) and $\pm 5\%$ at high energies ($\gtrsim 10$ MeV) with gradual variation between these extremes for intermediate energies[16,17]). The numerical integration of eq. (4) is not taken to zero particle energy but carries a lower cutoff at 5 keV. This will contribute a very small systematic error to the fitted curves which can be neglected for the particle energies considered in the present work. The overall uncertainties of the fitting procedures will result in an estimated uncertainty of $\pm 10\%$ for the extracted values of kB.

3. Results and discussion

The fits to the experimental data of Smith et al.[1]) are exhibited in figs. 1–6. The dashed lines represent the best fits of eq. (7) to the electron response data points. The best one- and two-parameter fits of eq. (4) are identified respectively by crosses and smooth curves. The best-fit parameters are listed in table 3 along with the corresponding χ^2 values which provide quantitative comparison of the fits. It is apparent from figs. 1–6 that excellent fits to the experimental data over wide particle energy ranges can be obtained with eqs. (4) and (7). The one- and two-parameter fits to the anthracene data are nearly identical [very small value for C] in agreement with Chou[3]). However, the two-parameter fits are generally superior to the one-parameter fits for the other scintillators. It is not known what physical interpretation should be assigned to the parameter C. However, it is interesting to note that all of the best-fit values obtained for C in the present work are positive.

Several other researchers have fitted organic scintillator response data with eq. (1), e.g. refs.[2]) and[18–25]). The parameter kB has become the accepted basis for comparison of these results. Values of kB for anthra-

* Nuclear Enterprises, Inc., San Carlos, California (anthracene, stilbene, NE-213, NE-230 and NE-102);
Pilot Chemical Company, Cambridge, Massachusetts (Pilot-B).

TABLE 2

Computed specific energy losses for deuterons in NE-230.

Deuteron energy (MeV)	dE/dx (MeV·cm²·g⁻¹)	Deuteron energy (MeV)	dE/dx (MeV·cm²·g⁻¹)	Deuteron energy (MeV)	dE/dx (MeV·cm²·g⁻¹)
0.005	351.0	0.26	751.2	2.20	203.6
0.010	633.3	0.28	734.3	2.40	194.4
0.015	882.6	0.30	713.1	2.60	185.1
0.02	1088.6	0.32	693.0	2.80	175.0
0.03	1265.8	0.34	673.9	3.00	166.2
0.04	1306.6	0.36	655.9	3.20	160.5
0.05	1296.7	0.38	640.1	3.40	154.2
0.10	1080.6	0.40	620.4	3.60	147.2
0.11	1050.9	0.50	549.0	3.80	142.3
0.12	1021.2	0.60	489.6	4.00	136.6
0.13	991.6	0.70	453.6	5.00	116.8
0.14	969.0	0.80	413.4	6.00	101.5
0.15	940.8	0.90	384.3	7.00	89.6
0.16	914.2	1.00	355.1	8.00	81.1
0.17	888.8	1.10	335.0	9.00	72.7
0.18	865.0	1.20	311.6	10.00	67.9
0.19	843.9	1.40	282.0	11.00	62.7
0.20	817.9	1.60	257.6	12.00	57.6
0.22	795.7	1.80	238.1	14.00	51.1
0.24	773.4	2.00	218.8		

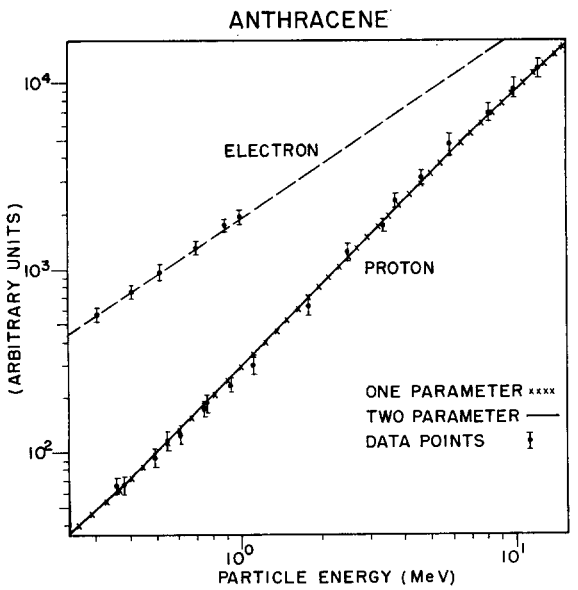

Fig. 1. Semiempirical fits to electron- and proton-response data for anthracene, measured by Smith et al.[1]).

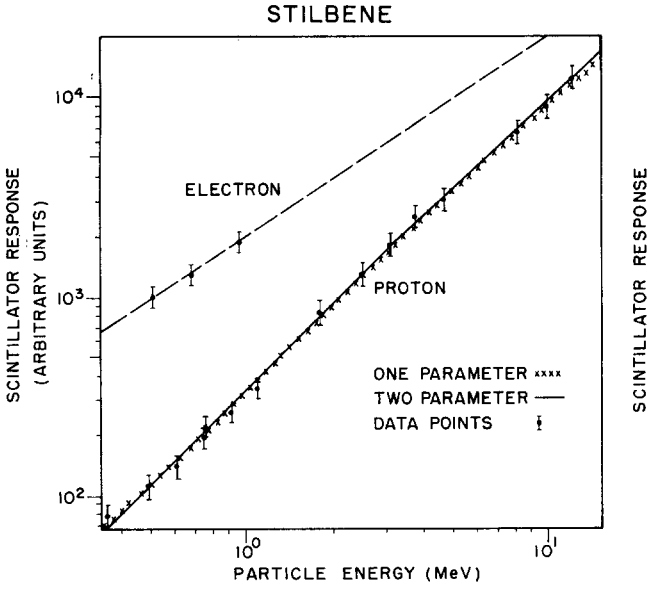

Fig. 2. Semiempirical fits to electron- and proton-response data for stilbene, measured by Smith et al.[1]).

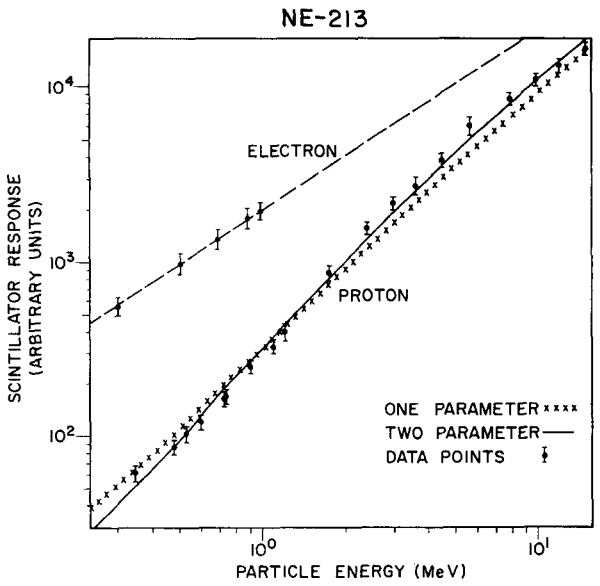

Fig. 3. Semiempirical fits to electron- and proton-response data for NE-213, measured by Smith et al.[1]).

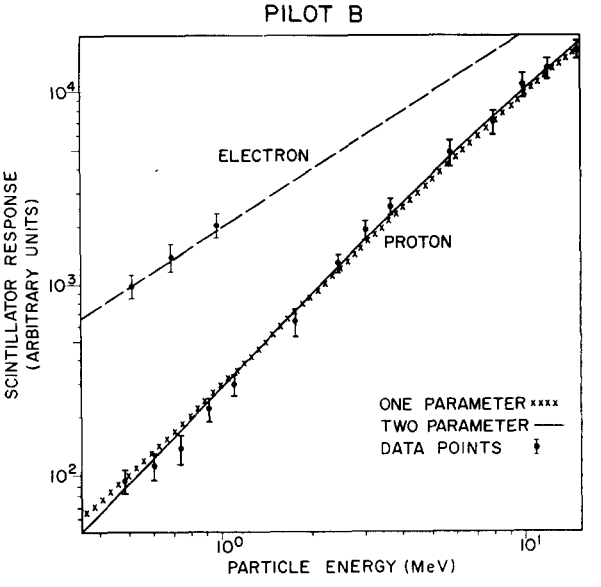

Fig. 5. Semiempirical fits to electron- and proton-response data for Pilot-B, measured by Smith et al.[1]).

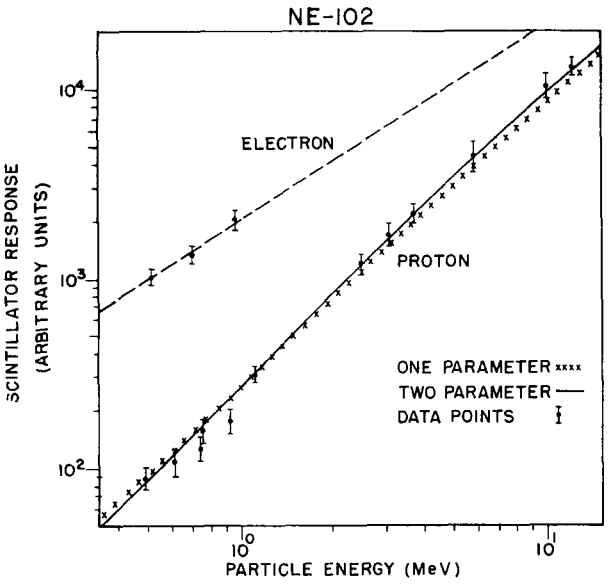

Fig. 4. Semiempirical fits to electron- and proton-response data for NE-102, measured by Smith et al.[1]).

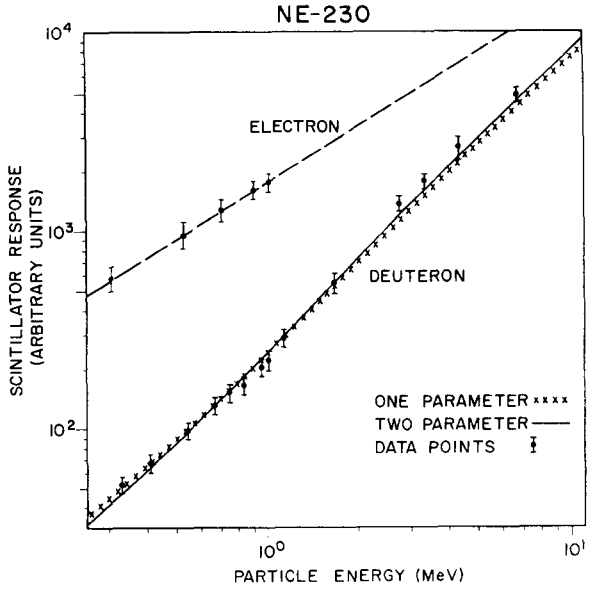

Fig. 6. Semiempirical fits to electron- and deuteron-response data for NE-230 measured by Smith et al.[1]).

cene and NE-102 which were obtained in the present work are compared in table 4 with several corresponding results reported in the literature. The disparity of these results indicates a need for more accurate measurements and for better theoretical understanding of the processes involved. There is considerable evidence that the response functions for organic scintillators are strongly dependent upon their chemical composition and purity (manufacturing process and storage history) and their histories of exposure to light and other radiations[2,23]). Thus, the response data for a particular scintillator can be fitted very well with the semiempirical formulas described in this paper, although care must be taken in using these formulas for the same scintillator after it has aged or for other scintillators of the same type.

TABLE 3

Parameters obtained from least-squares fits of the formulas to experimental data of Smith et al.[1]).

Scintillator	Particle	Particle energy (MeV)	dE/dx (MeV·cm^2·g^{-1})	S (Arb. units)	One-parameter fits		Two-parameter fits		
					$(kB)_1$ (g·cm^{-2}·MeV^{-1})	χ_1^2	$(kB)_2$ (g·cm^{-2}·MeV^{-1})	C_2 (g^2·cm^{-4}·MeV^{-2})	χ_2^2
NE-102	rec. prot.*	0.35–15.0	30.3–494	2113	1.31×10^{-2}	23.0	1.29×10^{-2}	9.59×10^{-6}	19.4
NE-213	rec. prot.	0.24–15.0	30.4–601	1929	1.25×10^{-2}	67.8	7.35×10^{-3}	1.45×10^{-5}	28.9
Pilot B	rec. prot.	0.35–15.0	30.3–494	2232	1.59×10^{-2}	18.0	1.29×10^{-2}	1.05×10^{-5}	14.2
Stilbene	rec. prot.	0.35–15.0	30.2–489	1717	9.55×10^{-3}	6.1	8.56×10^{-3}	3.99×10^{-6}	5.4
Anthracene	rec. prot.	0.24–15.0	30.3–591	2074	1.46×10^{-2}	10.4	1.45×10^{-2}	1.61×10^{-6}	10.4
NE-230	rec. deut.†	0.25–11.0	62.7–760	1915	1.10×10^{-2}	27.2	9.17×10^{-3}	4.60×10^{-6}	24.0

*rec. prot. = recoil protons, † rec. deut. = recoil deuterons.

TABLE 4

Comparison of kB values extracted from fits of the Birks formula to several sets of experimental response data*.

Reference	Scintillator	Particles	Particle energy (MeV)	dE/dx (MeV·cm^2·g^{-1})	kB (g·cm^{-2}·MeV^{-1})
Brooks[18])	anthracene	protons†	1–17	<237	6.6×10^{-3}
Present work	anthracene	recoil protons	0.24–15.0	30.3–591	1.46×10^{-2}
Daehnick and Fowler[19])	NE-102	recoil protons	<8.4	>50	2×10^{-3}
Evans and Bellamy[20])	NE-102	recoil protons	1.2–14	>34	$(1.0\pm0.1)\times10^{-2}$
Gooding and Pugh[21])+	NE-102	external protons	28–148	5.5–20	$(1.32\pm0.25)\times10^{-2}$
Gettner and Selove[22])	NE-102	recoil protons	<2.3	>150	1.0×10^{-2}
Prescott and Rupaal[23])	NE-102	recoil protons	<4	>97	$(9.1\pm0.6)\times10^{-3}$
Groom and Hauser[24])	NE-102	external protons	<100	>7	$(3.7\rightarrow7.5)\times10^{-3}$
Badhwar et al.[25])	NE-102	external protons	36–220	4.2–12.3	$(1.26\pm0.20)\times10^{-2}$
Present work	NE-102	recoil protons	0.35–15.0	30.3–494	1.31×10^{-2}

* Fits were made using either eq. (1) or eq. (4), with $C=0$.
† Data compiled from both recoil- and external-proton response measurements.
+ J. B. Birks, ref. [2]), p. 193 for a correction to these results.

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