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ABSTRACT

The use of cellulose nitrate films LR-115 type II (Kodak-Pathé) as a practical, exposure integrating device to measure the level of exposure to alpha particles in atmospheres which contain radon and radon-daughter products is investigated. The analysis of a number of cellulose nitrate films that have been exposed to calibrated radon test-chamber atmospheres has indicated good correlations between through-etched track density ρ and integrated alpha-particle exposure ε_α (Working-Level-Hour). It is shown that the response of the cellulose nitrate detector to radon-daughter alpha-particle exposures is linear, and that reliable conservative estimations of the Working-Level-Hour can be obtained from $\varepsilon_\alpha = 3.0(\rho - b)$, where ρ is expressed in tracks/mm^2 (b is the background level). These results recommend the use of the special red cellulose nitrate films as a convenient dosimeter for monitoring radioactive contaminants in mine atmospheres.

1. INTRODUCTION

It is well known that polymers are highly sensitive to track registration of low-energy alpha particles and protons, and that such a basic property makes feasible numerous applications of cellulose nitrate and many other plastic nuclear track detectors to several branches of nuclear science (Fℓ75). For instance, cellulose nitrate track detectors have been widely tested with satisfactory results as an alpha-particle exposure monitor (Ro69; Lo69; Au71; Be72; Ch72; Fr77; Pa77; Pa78; Ph79; To80).

In recent years, the availability of the special red cellulose nitrate films LR-115 type II, supplied by the Kodak-Pathē (Ba73) has opened new possibilities of solving numerous practical problems concerning the detection of low-energy alpha particles and protons. This particular track detector consists of a thin reddish layer ($\sim$ 13μm) of cellulose nitrate covering a $\sim$ 100μm thick polyester sheet. Such a configuration has the advantage of enhancing track contrast when tracks perforate the residual cellulose nitrate layer after prolonged etching, thus allowing for better recognization of tracks during track analysis.

In a previous paper (Kn80) some registration properties of the cellulose nitrate film LR-115 type II to low-energy alpha particles were studied in detail and it was commented on its possible use as a suitable tool in detecting alpha particles from contaminated atmospheres of radon[†] and radon daughter products.

[†] Unless otherwise specified, the term radon will be referred to as the ^{222}Rn isotope.

In the present work we examine the response of the cellulose nitrate detector to alpha-particle exposures from calibrated radon test-chamber atmospheres. Although it has been already tested in other laboratories, the technique of the through-etched track analysis we develop allows one to obtain a better correlation between track densities and integrated alpha particle exposures. These correlations permit us to obtain reliable estimations of integrated alpha-particle exposures due to short-lived radon-daughter products (working-level-hour). This result, combined with the general registration characteristics of cellulose nitrate detectors as discussed earlier (Ro69; Lo69) (good stability of latent alpha-particle tracks against air humidity, insensitivity to light, gamma and beta radiations, inability to record alpha particles from radon daughters plated-out on the detector surface) strongly recommend the use of such detectors as a personnel dosimeter for monitoring lung doses received by workers exposed to uranium mine atmospheres.

2. EXPERIMENTAL DETAILS

Clean sheets of the reddish, double-layer, cellulose nitrate film ∿13μm thick were exposed to alpha particles emitted from radon and radon-daughter products in various calibrated radon test-chamber atmospheres at the Denver Mining Research Center (Denver, Colo.). Data concerning information exposures are given in Table 1. Three different RaA, RaB and RaC mixtures were used during the exposures and they are indicated as a Group I, II and III. In Fig. 1 we show a trian-

gular graph with these three groups plotted relative to each other (filled circles). The triangle was constructed on the same scale as in Fig. 2 of the BuMines RI 8316 (Ho78), so that the mixtures used in the present chamber exposures can be compared to mixtures measured in various uranium mine atmospheres. Twelve exposures were made using two films, each film having six areas exposed making a total of twelve areas for each exposure.

The cellulose nitrate films were etched in a 4.0 N NaOH solution at a constant temperature of 60°C without stirring. Several etching times (1.50, 2.25, 2.50, 3.00 and 3.50 h) were selected in order to investigate the dependence of the measured through-etched track density on etching time. Track counting was carried out by using conventional optical microscopes (Leitz, Ortholux) fitted out with objective 25×1 and ocular periplan 6×B. In order to determine the alpha-particle incident energy on the detector surface the optics objective KS 45×1 and ocular periplan GF 25×M was used for track-diameter measurements (alpha particles which hit the detector surface at right angles produce high-contrast circular holes whose diameters decrease with increasing alpha-particle energy for a given etching time (Kn80)). Finally, calibrated eyepieces made it posible to determine accurately the scanning areas (within 1% uncertainly) of the various plates, so that the measured track densities could be reported as a number of tracks observed per unit area.

3. RESULTS

The through-etched track densities obtained under the

experimental conditions as described above are reported in Table 2. In Figs. 2-4 we have plotted the track densities *versus* etching time for each exposure. Some interesting features must be commented on. All curves exhibit similar trends independently of the radon-daughter mixtures used during exposures (groups I, II and III) and the particular exposure condition within each group. A threshold etching time of about 30 min is apparent, i.e. below this value through-etched tracks were not visible at all. This is because the detector itself has a period of latency of about 20 min due to a very small dissolution rate during the early stages of etching. Once the etching proceeds normally some more time must be expended for latent tracks produced by the lower energetic (strongly ionizing) alpha particles be revealed in such a way that the circular holes formed on the polyester surface become as large as $\sim$2μm in diameter in order that tracks can be safely recognized by the observer. As the etching time increases the track diameter also increases and, at the same time, tracks due to more energetic alpha particles are revealed. This makes the track density increase monotonically with etching time until a certain plateau value is reached when all latent alpha-particle tracks produced under the conditions of uncollimated and multienergetic alpha particles become visible. In fact, it is observed from Fig.2-4 that after about 3h of etching the track-density curves begin to show a tendency of saturation which seems to be completed after about 3.5h of etching (more prolonged etching times should be avoided since 3.5h of chemical attack cause the thickness of the cellulose nitrate layer to be reduced to about 30% of its original value).

Fig. 5 shows the distributions of through-etched track-diameter (which are related to the alpha-particle energy distributions) measured for two film exposures. To illustrate the incident energy spectrum of alpha particles detected by the system, we chose those exposures of maximum exposure time of the two extreme radon-daughter mixtures (groups I and III). As can be seen, for alpha particles of normal incidence to the detector surface the incident energy distribution is nearly isotropic in the energy-range of 2.4-5.6 MeV. This is the energy interval within which alpha particles can be recorded by the present detection technique. From the very similar track-diameter distributions obtained for the two extreme exposures the conclusion could be drawn that the detector does not allow one to identify alpha particles emitted from a particular alpha emitter (Rn, RaA, RaC').

From the above listed observations it is concluded that cellulose nitrate (LR-115, type II) films exposed to radon test-chamber atmospheres respond with maximum efficiency to both through-etched track density and incident energy-range whenever the films are processed in a 4.0N NaOH solution at 60°C during 3.5h. For this reason we take the above etching conditions as a standard one for future applications of film exposed to uranium mine atmospheres. Of course other possible combinations of etchant concentration, temperature, and etching time would give results very similar to the present ones. However, as has been demonstrated from our previous experience (Kn80), the special red cellulose nitrate detector works well under the specified development conditions. Thus, in what follows, we will concern ourselves with the discussion referred to in the above

mentioned etching conditions.

4. RADON CONCENTRATION AND RADON EXPOSURE DETERMINATIONS

Before entering on the description of the method we propose in evaluating cumulative alpha-particle exposures by the technique of through-etched track counting in the cellulose nitrate LR-115 (type II) track detector, let us first recall some concepts and units of routine use in radiation protection, safety and health problems concerning uranium-mine atmospheres and environment (a complete discussion on both instrumentation and measurement techniques of radon daughter mixtures can be found in the review article by Busigin *et al.* (Bu79)).

A contaminated atmosphere is a system containing air, radon and airbone radon-daughter products. The concentration of each radioactive component is, in general, expressed in units of pCi/ℓ (1pCi equals to 2.22 dpm). An equilibrium atmosphere (or air) is a system containing radon in radioactive equilibrium with its daughter products.

The short-lived radon daughters RaA, RaB, RaC and RaC' (see Table 3) are the significant ones for radiological health hazard purposes. To express the occupational exposure level to radon daughters the Working Level unit (symbol WL) was introduced and it is defined as "any combination of radon daughters in 1ℓ of air that will result in the ultimate emission of 1.3×10^5 MeV of the potential alpha energy"[†]. The potential alpha-particle energy contained in a mixture of RaA, RaB, RaC and RaC' defined by the concentrations C_A, C_B, C_C and C_C, (in pCi/ℓ),

[†]Federal Radiation Council, 1967, "Guidance for the Control of Radiation Hazards in Uranium Mining", Washington, D.C., Federal Radiation Council, Report 8 Revised, September.

respectively, of its components is given by

$$E_\alpha = \left[(6.00+7.68) \frac{C_A}{\lambda_A} + 7.68 \left(\frac{C_B}{\lambda_B} + \frac{C_C}{\lambda_C} + \frac{C_{C'}}{\lambda_{C'}} \right) \right] \times 2.22 \quad \text{MeV}/\ell , \tag{1}$$

where the λ's denote the decay constants expressed in min^{-1}. Substituting the λ's by their respective values, we obtain

$$E_\alpha = C_A(134 + 661\, r_B + 487\, r_C) \quad \text{MeV}/\ell , \tag{2}$$

where $r_B = C_B/C_A$ and $r_C = C_C/C_A$ define the concentrations of RaB and RaC relative to that of RaA (due to its shorter half-life the component RaC' does not contribute at all to the total potential alpha-particle energy; in other words, the alpha particles emitted by RaC' can be entirely ascribed to RaC). Thus, the exposure level ε, measured in WL, to an atmosphere containing RaA, RaB and RaC is obtained from

$$\varepsilon = \frac{C_A(134 + 661\, r_B + 487\, r_C)}{1.3 \times 10^5} \quad \text{WL} . \tag{3}$$

In the case of equilibrium atmospheres, 1 WL corresponds to a concentration of 100 pCi/ℓ of radon in equilibrium with its daughter products.

The time-integrated (or accumulated) exposure is defined by

$$\varepsilon_a = \int_0^t \varepsilon \, dt' \quad \text{WL-h} , \tag{4}$$

where t denotes the exposure time expressed in h. If the assumption is made that the concentrations of RaA, RaB and RaC do not vary significantly during the exposure time (a case not

completely verified in practical situations, although it can be assumed as a rough approximation for simplest calculation purposes) eqns (3) and (4) are combined to give

$$\varepsilon_a = C_A(1.03 + 5.08\, r_B + 3.75\, r_C)\, t \times 10^{-3} \quad \text{WL-h.} \qquad (5)$$

Mixtures of airbone radon daughters have been represented on triangular graphs (Ho78) like the one shown in Fig. 1. Any point within the triangle represents a mixture of RaA, RaB and RaC normalized to an exposure level of 1 WL. These graphs have the advantage of rapid comparing different radon daughter mixtures to each other. A quantity which measures the position of a given non-equilibrium mixture relative to the equilibrium atmosphere is the Working Level Ratio (WLR). This is defined as a 100 times the occupational exposure level ε in WL divided by the radon concentration C_{Rn} in pCi/ℓ, i.e.

$$\text{WLR} = 100\varepsilon/C_{Rn}\ , \qquad (6)$$

which figured with eqn (3) can be transformed into

$$\text{WLR} = (0.10 + 0.51\, r_B + 0.38\, r_C)\, \frac{C_A}{C_{Rn}}\ . \qquad (7)$$

For equilibrium air mixtures (the point + marked in Fig.1), we have $C_A = C_{Rn}$, $r_B = r_C = 1$ and, therefore, WLR = 1.

Let us now discuss the correlations between the measured through-etched track density and the appropriate quantities which characterize the various alpha-particle accumula-

ted exposures of cellulose nitrate films to the radon test-chamber atmospheres as described in Section 2. The track detector is capable of recording alpha particles only. They hit the detector surface at all angles in the range $0\text{-}90^{0}$ and at incident energies varying from a few keV up to 7.68 MeV. A number of alpha particles will produce through-etched tracks after etching whose track-density will depend upon the incident particle energy, the angle of entrance and the etching conditions. Since the detection system is unable to discriminate alpha particles coming from their respective alpha emitters Rn, RaA and RaC', and considering that alpha particles emitted by RaC' originate from its precursor RaC, we define the total time-integrated alpha-particle exposure

$$S = t \sum_{i} C_{i} \quad , \qquad i = \text{Rn, RaA, RaC} \tag{8}$$

as a quantity to be related to the observed track density ρ. As usual, the exposure time t and the concentrations C_{i} have been expresses in h and pCi/ℓ, respectively, and their values are listed in Table 1. The correlation between ρ and S is shown in Fig.6. For each group of exposure under consideration, the experimental points result to be well distributed around a straight line. Thus, the dependence of ρ on S can be written as

$$\rho = aS + b \ . \tag{9}$$

The parameter a defines the sensitivity of the detection system while the parameter b measures the background level. The points have been least-squares fitted to give the values of a and b for

each group of exposure (Table 4). Both the sensitivity a and the background b will depend upon the particular choice for the etching conditions. The background includes primitive latent tracks of the films themselves, tracks collected during preexposure time intervals, and tracks which were accumulated from the end of the exposures to the date of film processing. From eqns (8) and (9) we have

$$C_{Rn} + C_A + C_C = \frac{\rho - b}{at} \quad , \tag{10}$$

which means that the total concentration due to Rn, RaA and RaC components of each atmosphere can be obtained from the measured track density and exposure time. For a given atmosphere, good reproducibility for the quantity $C_{Rn} + C_A + C_C$ is to be obtained from prolonged exposure times since, in this case, fluctuations in the observed track density are small and, therefore, they will produce small deviations from the measured $C_{Rn} + C_A + C_C$ values. Calculations have indicated, in fact, that when the observed track densities for 3.5 h of etching are inserted into eqn (10) the sums of the concentrations of Rn, RaA and RaC reported in Table 1 are reproduced within 0.6%, 10% and 7% for the three more prolonged exposure times of groups I, II and III, respectively. Maximum deviations were noted for film number 4 ($\sim$ 7%), film number 5 ($\sim$ 40%) and film number 9 ($\sim$ 15%).

The exposure level to radon itself (and, therefore, the radon concentration) could be evaluated in the following way. Since alpha particles emitted from radon do contribute to a

larger extent tracks to be revealed by etching, let us include the contribution due to radon into the potential alpha-particle energy contained in a given atmosphere. We have

$$E_{\alpha,total} = E_{\alpha,Rn} + E_{\alpha,daughters} \cdot \qquad (11)$$

$E_{\alpha,daughters}$ is given by eqn (2) and

$$E_{\alpha,Rn} = (5.49 + 6.00 + 7.68)\,\frac{C_{Rn}}{\lambda_{Rn}} \times 2.22 \quad \text{MeV}/\ell \quad , \qquad (12)$$

where the radon concentration, C_{Rn}, and decay constant, λ_{Rn}, are expressed in pCi/ℓ and min^{-1}, respectively. Thus eqn(11) transforms into

$$E_{\alpha,total} = 3.38 \times 10^{5}\, C_{Rn} + C_A(134 + 661 r_B + 487 r_C) \quad \text{MeV}/\ell. \qquad (13)$$

The second term in (13) can be safely neglected (for equilibrium air it amounts to $\sim$0.4% of the potential alpha energy due to the radon content) and, therefore, the total time-integrated alpha-particle exposure turns out to be

$$\varepsilon_{Rn} = 2.60\, C_{Rn} t \quad \text{WL-h} \quad . \qquad (14)$$

This quantity is seen to be correlated to the track density as shown in Fig. 7. A linear relationship between ρ and ε_{Rn},

$$\rho = c\, \varepsilon_{Rn} + b \quad , \qquad (15)$$

is the most appropriate one in fitting the points. The parameter b represents the background level as before, and thus the values b = 8.43, 11.59 and 6.15 tracks/mm^2 may be taken *a priory* for Groups I, II and III, respectively. A least-squares procedure gave the corresponding values of the parameter c (Table 4). The correlation expressed by (15) might be considered satisfactory, since the points deviate from the straight lines to less than 7% (group I), 18% (group II) and 10% (Group III), on the average. Finally, eqns (14) and (15) are combined to give the radon concentration C_{Rn} (in pCi/ℓ) as a function of the exposure time t (in h) and the track density ρ (in mm^{-2}):

$$C_{Rn} = \frac{\rho - b}{2.60ct} \quad . \tag{16}$$

Again the observed track densities for 3.5h of etching can be substituted into eqn (16) to give the radon concentrations. For the sake of camparison, we compile in Table 5 all data regarding the radon component of the various atmospheres to which the films were exposed.

As a concluding remark, we point out the feasibility in obtaining reliable estimations (within about 10% uncertainty) of radon concentrations present in contaminated atmospheres by the through-etched track-counting method as described above.

5. RADON-DAUGHTER EXPOSURE DETERMINATION

From the radiation hazard, safety and health point of views, however, the quantity of main interest to be known is

the total radon-daughter accumulated exposure ε_a(WL-h). Once we have obtained both the sum of the concentrations $C_{Rn} + C_A + C_C$ of the alpha-emitter components Rn, RaA and RaC in the atmosphere, and the radon concentration C_{Rn} itself as functions of the track density and exposure time (eqns. (10) and (16), respectively), these were combined to give the RaA concentration. Accordingly,

$$C_A = \left(\frac{1}{a} - \frac{1}{2.60c}\right) \frac{\rho - b}{(1+r_C)t} \quad . \tag{17}$$

By inserting this quantity into the general expression for ε_a (eqn (5)), we obtain

$$\varepsilon_a = k(\rho - b) \qquad \text{WL-h} \quad , \tag{18}$$

where k is given by

$$k = \frac{1.03+5.08r_B+3.75r_C}{1 + r_C} \left(\frac{1}{a} - \frac{1}{2.60c}\right) \times 10^{-3} \quad . \tag{19}$$

Thus, for a given airbone radon daughter mixture, the Working-Level-Hour results to be a linear function of the measured track density. For the mixtures we have worked with the values of k (expressed in WL-h-mm^2 units) are as follows: 0.44± 0.04 for group I, 1.82±0.30 for group II, and 2.08±0.22 for group III.

The Working-Level-Hour has been plotted as a function of the track density for the three exposure groups under consideration (Fig. 8). The straight lines are obtained by using the

values of the parameters k and b already found for each exposure group. Experimental points have been also plotted to allow a comparison. These are drawn from data reported in Tables 1 and 2. Except for film exposure numbers 4 and 5 (which correspond to shorter exposure times in groups I and II) the agreement between calculated and measured Working-Level-Hour might be considered satisfactory (about 9% deviation on the average). Actually, the straight lines shown in Fig. 8 should be thought of as the calibration functions of the detection system to each radon-daughter mixture. They represent the response function of the cellulose nitrate LR-115 (type II) track detector under the specified processing conditions. Similar response functions would be obtained, for instance, from the track counting data corresponding to shorter etching times.

Of course the linear response function of the track detector to radon-daughter exposures for a given mixture would be easily obtained by correlating empirically both the measured Working-Level-Hour and track density. However, since the detector does record alpha particles emitted by the radon component present in the atmosphere, the radon component may contribute to a larger extent to backgroung tracks, mainly for atmosphere in which its concentration dominates. This is the case, for example, of the exposures made in the radon test-chamber atmosphere of group I, for which the radon concentration amounts, on the average, to about 80% of the total concentration of the alpha-emitter components. In such a situation, a correlation between the quantities ρ and S, like those studied in Section 4, can give a reliable indication of the

background level. This is the reason why the response functions have been obtained by first correlating the track density with the quantities S and ε_{Rn} and then subtracting the radon contribution from the total concentration $C_{Rn}+ C_A + C_C$. For atmospheres in which the radon component does not prevail (groups II and III) the above procedure may be disregard, and the correct response function can be obtained by a linear least-squares fitting on the measured quantities ε_a and ρ (eqn (18)).

Finally, we wish to discuss on the parameter k. According to eqn (19), k contains two basic factors. The first is related explicitly to the equilibrium ratios of the radon daughter mixture, and the second depends upon the fitting parameters a and c which in turn are related to the radon content in the atmosphere. This observation suggests us to correlate the parameter k with a quantity appropriate in characterizing the atmosphere under study. As discussed in the introductory part of Section 4, the Working-Level-Ratio (WLR) indicates how far is a given non-equilibrium atmosphere from the equilibrium point. Therefore, we expect k to be correlated to the WLR. In Fig. 9 we show the dependence of k on WLR. It is seen that k increases as the atmosphere approaches the equilibrium point (WLR = 1) . For future application of conservative estimations of the total radon-daughter accumulated exposures received by workers in uranium mine atmospheres, for instance, a value $k \simeq 3.0$ WL-h-mm^2 may be assumed, i.e.,

$$\varepsilon_a \simeq 3.0(\rho - b) \quad \text{WL-h} \quad \text{(conservative value)} \; . \qquad (20)$$

As a final remark, we point out the good performance of the cellulose nitrate LR-115 type II track detector in estimating radon daughter exposures in a wide range of Working-Level-Hour values (up to about 700 WL-h for film exposures to the atmospheres of groups II and III (cf. Fig. 1)).

6. CONCLUSIONS

The use of the cellulose nitrate LR-115 type II (Kodak-Pathē) track detector as a monitor of cumulative radon and radon-daughter exposures was investigated with very encouraging results. The analysis of a series of data obtained from radon test-chamber atmosphere exposures has indicated strong linear correlations between the through-etched track density observed and the time-integrated radon-daughter exposure measured by currents methods. The technique of through-etched track analysis we developed also allows one to estimate the concentration of the radon content in the atmosphere. In addition, it gives reliable indications of the background level which are important to know mainly for those atmospheres where the radon concentration prevails. Under the processing conditions of 4.0 N NaOH solution at $60^{o}C$ during 3.50 h without stirring, the Working-Level-Hour of an individual film exposure can be simply obtained by taking the net through-etched track density $\rho - b$, in tracks/mm^2, times a calibration factor k ($0<k<3$) which depends upon the degree of disequilibrium of the atmosphere. Besides, a number of advantages of the track detector itself (small, light, simple, inexpensive, insensitive to light, beta and gamma radiations, unable of recording alpha particles from radon daugh

ters plated-out on the detector surface) over other monitoring devices strongly recommends the use of the special red cellulose nitrate film as a passive monitor in measuring the cumulative radon daughter exposure received by individual workers of mine atmospheres. Testing of these plastic track detectors under actual operating conditions of mine atmospheres should be conducted in the near future.

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Table 1. Exposure conditions for cellulose nitrate films in the radon test-chamber atmospheres at Denver Research Center.

Group†	Equilibrium ratios of the radon-daughter concentrations			Film number	Exposure time (h)	Radon-daughter exposure (WL-h)	Radon concentration (pCi/ℓ)	Average radon-daughter concentration* (pCi/ℓ)			WLR‡	Room humidity (%)
	RaA	RaB	RaC					RaA	RaB	RaC		
I	1	0.321	0.151	1	28.500	37.36	1981	406	130	61	0.066	2
				2	94.367	102.71	1955	337	108	51	0.056	2
				3	18.133	24.13	1953	412	132	62	0.068	2
				4	3.800	10.48	1960	855	274	129	0.141	3
II	1	0.440	0.230	5	1.867	10.08	2000	1307	575	301	0.270	2
				6	3.750	20.09	2003	1297	571	298	0.267	3
				7	7.500	40.80	2009	1317	579	303	0.271	3
				8	18.500	108.38	2008	1419	624	326	0.292	4
III	1	0.640	0.428	9	0.433	10.19	5313	3997	2558	1711	0.443	28
				10	0.900	21.31	5305	4021	2573	1721	0.446	28
				11	1.750	41.78	5308	4055	2595	1736	0.450	28
				12	4.383	106.41	5326	4123	2639	1765	0.456	28

†Group I was exposed using the room humidity and condensation nuclei generated in the room. Group II was exposed using room humidity with the bulk of the condensation nuclei coming from a nebulizer using diesel fuel. In Group III, the humidity level was raised with a humidifier and the condensation nuclei source was the same as Group II. Two mixing fans kept the chamber air in a turbulent condition.

*Deduced values by using eqn (5).

‡The working level ratio, WLR, is defined as 100 times the WL divided by the radon concentration in pCi/ℓ. The quoted values were deduced by using eqn (7).

Table 2. Alpha-particle through-etched track densities (expressed as number of tracks per square centimetre), measured in cellulose nitrate films exposed to different test-chamber atmospheres.

Group	Film number	Etching time†, h				
		1.50	2.25	2.50	3.00	3.50
I	1	880 ± 90‡	3450 ± 180	4800 ± 200	8350 ± 340	8330 ± 380
	2	3150 ± 170	9030 ± 350	13350 ± 500	23300 ± 930	24300 ± 950
	3	720 ± 100	2390 ± 160	3000 ± 150	4760 ± 220	5540 ± 260
	4	220 ± 50	790 ± 100	1190 ± 140	1680 ± 140	1950 ± 140
II	5	190 ± 50	510 ± 60	680 ± 110	1490 ± 120	1500 ± 130
	6	350 ± 60	1010 ± 110	1400 ± 120	-	2620 ± 130
	7	530 ± 70	-	2360 ± 150	2880 ± 170	3420 ± 170
	8	990 ± 100	3060 ± 150	3780 ± 190	6210 ± 260	7060 ± 300
III	9	100 ± 30	350 ± 60	-	800 ± 90	1040 ± 110
	10	220 ± 60	-	900 ± 90	-	1570 ± 130
	11	-	1080 ± 100	1360 ± 150	2270 ± 190	2870 ± 140
	12	970 ± 140	2030 ± 150	2580 ± 150	4620 ± 240	5720 ± 230

†Etching in 4.0N NaOH solution at 60°C without stirring.

‡The quoted errors are statistical only.

Table 3. Decay data for the radionuclides of interest in contaminated atmosphere studies.

Radioactive species	Nuclide	Mode of decay	Particle energy (MeV)	Half-life	Decay constant (min^{-1})
Rn	$^{222}_{86}Rn$	α	5.49	3.83 d	1.26×10^{-4}
RaA	$^{218}_{84}Po$	α	6.00	3.05 min	2.27×10^{-1}
RaB	$^{214}_{82}Pb$	β^-	-	26.8 min	2.58×10^{-2}
RaC	$^{214}_{83}Bi$	β^-	-	19.8 min	3.50×10^{-2}
RaC'	$^{214}_{84}Po$	α	7.68	162 μs	2.57×10^{5}

Table 4. Best values for the parameters a, b and c from least-squares fitting (eqns (9) and (15); Figs. 6-7).

Group	Parameter a (tracks/(mm^2-h-pCi/ℓ))	Parameter b (tracks/mm^2)	Parameter c (tracks/(mm^2-WL-h))
I	$(1.062 \pm 0.005) \times 10^{-3}$	8.43 ± 0.60	$(0.491 \pm 0.008) \times 10^{-3}$
II	$(0.853 \pm 0.057) \times 10^{-3}$	11.59 ± 2.16	$(0.609 \pm 0.041) \times 10^{-3}$
III	$(1.045 \pm 0.053) \times 10^{-3}$	6.15 ± 1.42	$(0.849 \pm 0.040) \times 10^{-3}$

Table 5. Comparison between measured and calculated quantities for the radon component in various atmospheres.

Group	Film number	Exposure time t(h)	Measured track-density ρ(mm^{-2})	Radon concentration, C_{Rn}(pCi/ℓ)		Time-integrated radon exposure ε_{Rn} (10^5 WL-h)	
				Measured	Calculated (Eqn (16))	Measured†	Calculated (Eqn (15))
I	1	28.500	83.3 ± 3.8	1981	2058	1.47	1.52
	2	94.367	243 ± 10	1955	1947	4.80	4.78
	3	18.133	55.4 ± 2.6	1953	2029	0.92	0.96
	4	3.800	19.5 ± 1.4	1960	2282	0.19	0.23
II	5	1.867	15.0 ± 1.3	2000	1153	0.10	0.06
	6	3.750	26.2 ± 1.3	2003	2460	0.19	0.24
	7	7.500	34.2 ± 1.7	2009	1904	0.39	0.37
	8	18.500	70.6 ± 3.0	2008	2014	0.97	0.97
III	9	0.433	10.4 ± 1.1	5313	4447	0.06	0.05
	10	0.900	15.7 ± 1.3	5305	4807	0.12	0.11
	11	1.750	28.7 ± 1.4	5308	5838	0.24	0.27
	12	4.383	57.2 ± 2.3	5326	5276	0.61	0.60

†Deduced values by using eqn (14).

FIGURE CAPTIONS

Fig.1 - A triangular plot showing the three RaA, RaB and RaC mixtures used during the exposures (filled circles). These mixtures are indicated as group I, II and III in Table I. Equilibrium air is represented by the point cross. Open triangle locates the average experimental mixture of all uranium mine atmosphere data compiled by Holub and Droullard (Ho78). Open square indicates the location of the disequilibrium 1.00:0.60: 0.40 (Bu80). To compare these data with those measured in mine atmospheres see Fig. 2 of (Ho78).

Fig.2 - Observed through-etched track density plotted against etching time for the cellulose nitrate films of group I. The numbers near the curves indicate the different exposure conditions as reported in Table I. All curves are eye-fits through the experimental points.

Fig.3 - The same as in Fig.2 for group II.

Fig.4 - The same as in Fig.2 for group III.

Fig.5 - Through-etched track-diameter distributions obtained for two film exposures. The corresponding exposures conditions are indicated in Table 1. Alpha-particle incident energies can be deduced from the upper scale which was constructed from calibration data by Knöfel *et al.* (Kn80).

Fig.6 - Dependence of through-etched track density ρ on total alpha-particle exposure S. The quantity S is defined by the sum of the concentrations of the alpha-emitter components in the air (Rn, RaA and RaC) times the ex

posure time (eqn (8)). The points represent the track densities measured under the specified etching conditions (last column of Table 2). The straight lines are least-squares fits through the points.

Fig.7 - Through-etched track density ρ plotted against total alpha-particle exposure ε_{Rn} due to the radon component only. The quantity ε_{Rn}, in WL-h, equals to 2.60 times the radon concentration, in pCi/ℓ, times the exposure time in h (eqn (14)). The points represent the track densities measured under the specified etching conditions (last column of Table 2). The straight lines are least-squares fits through the points.

Fig.8 - Dependence of radon-daughter accumulated exposure ε_a (WL-h) on through-etched track density ρ for cellulose nitrate LR-115 (type II) films. All data have been obtained from radon test-chamber atmosphere exposures (for details see text).

Fig.9 - Illustrating the trend of the parameter k with the degree of disequilibrium of contaminated atmospheres (WLR). From the lower left corner up are plotted the points being refered to as group I, II and III. The k values have been obtained under the specified processing conditions. The curve is a least-squares fit through the points.

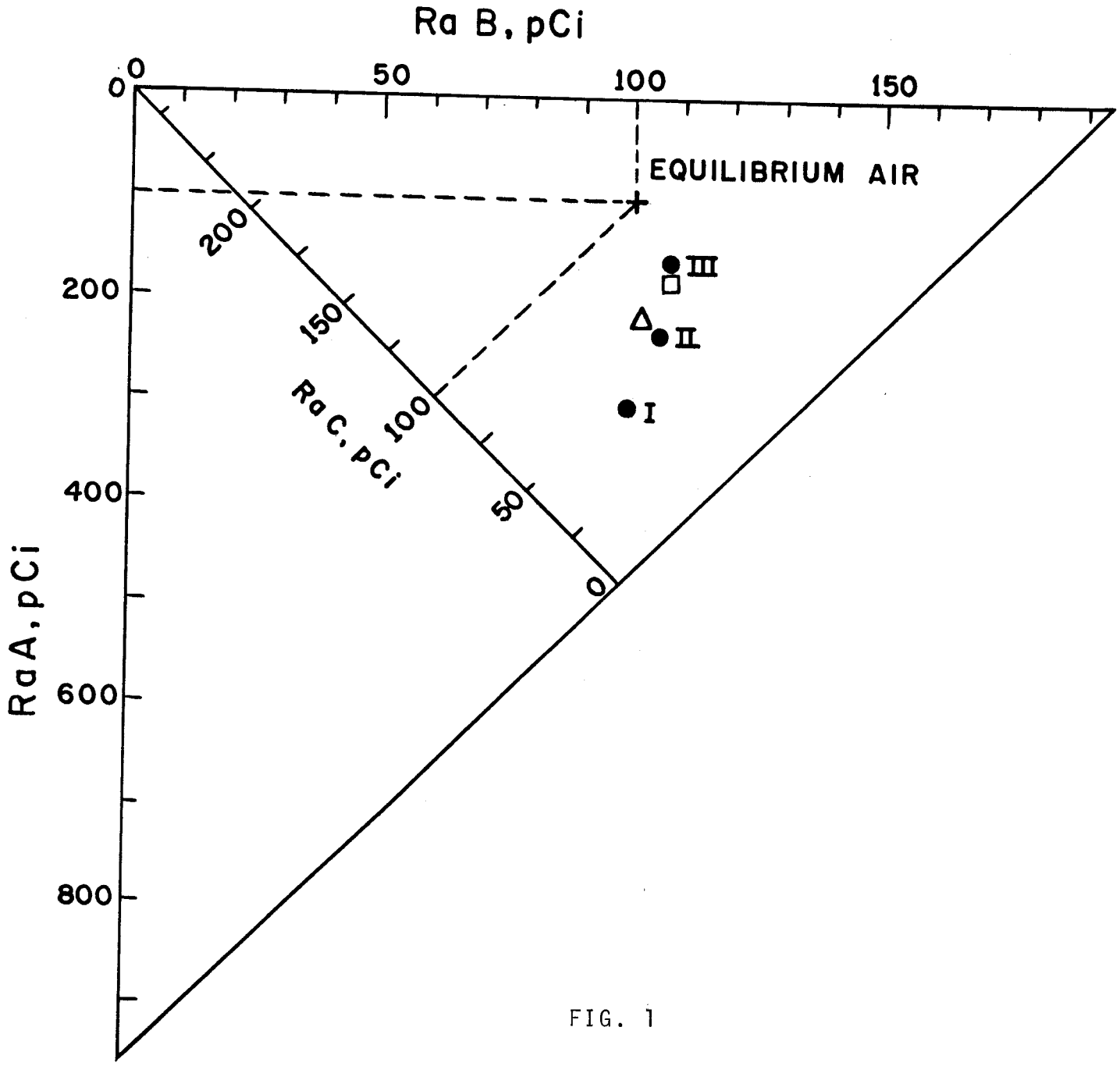
Ra B, pCi
0
50
100
150
EQUILIBRIUM AIR
200
150
100
50
0
Ra C, pCi
III
II
I
200
400
600
800
Ra A, pCi

FIG. 1

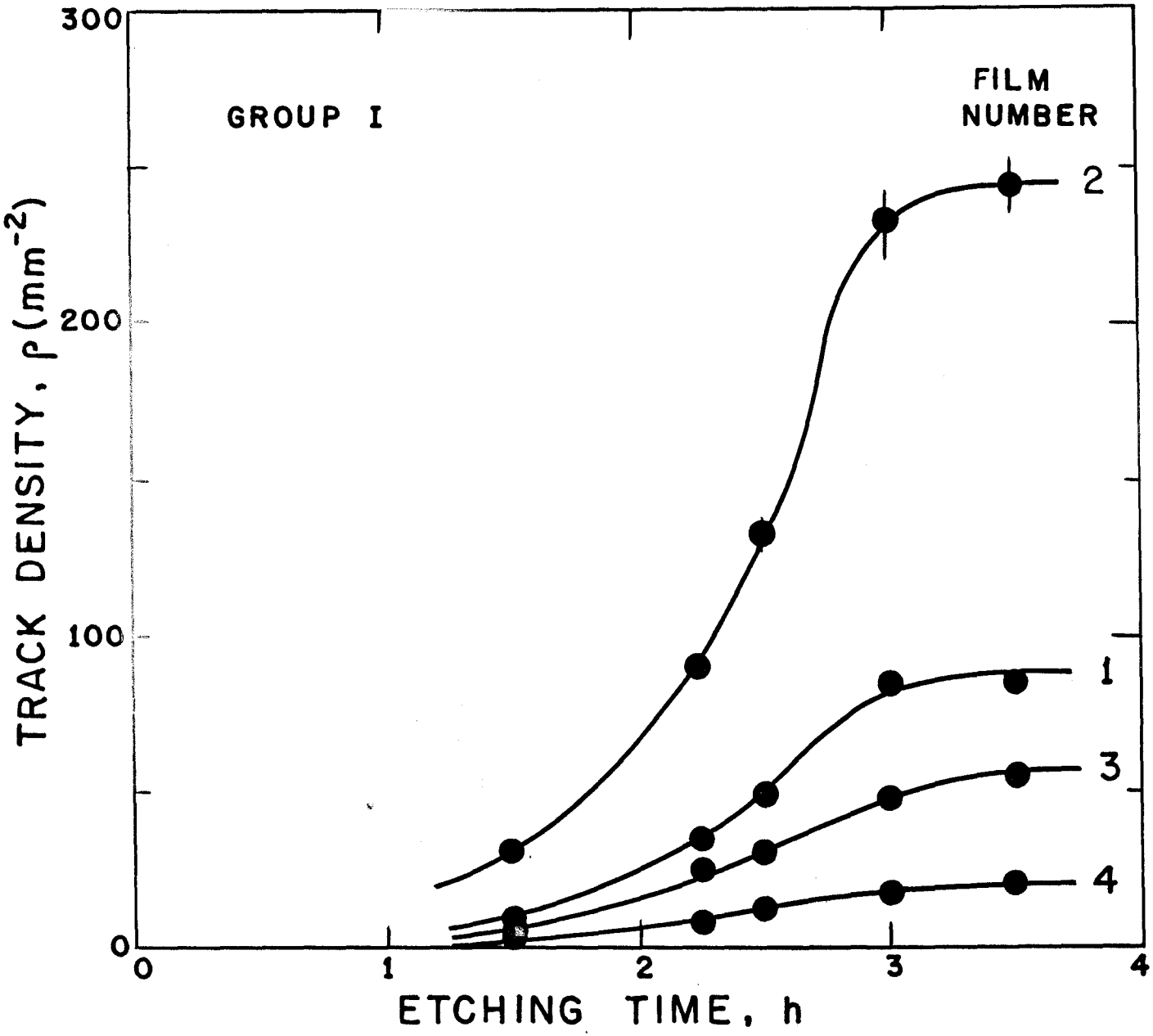
GROUP I
FILM NUMBER
2
1
3
4
TRACK DENSITY, ρ (mm⁻²)
ETCHING TIME, h
0
100
200
300
0
1
2
3
4

FIG. 2

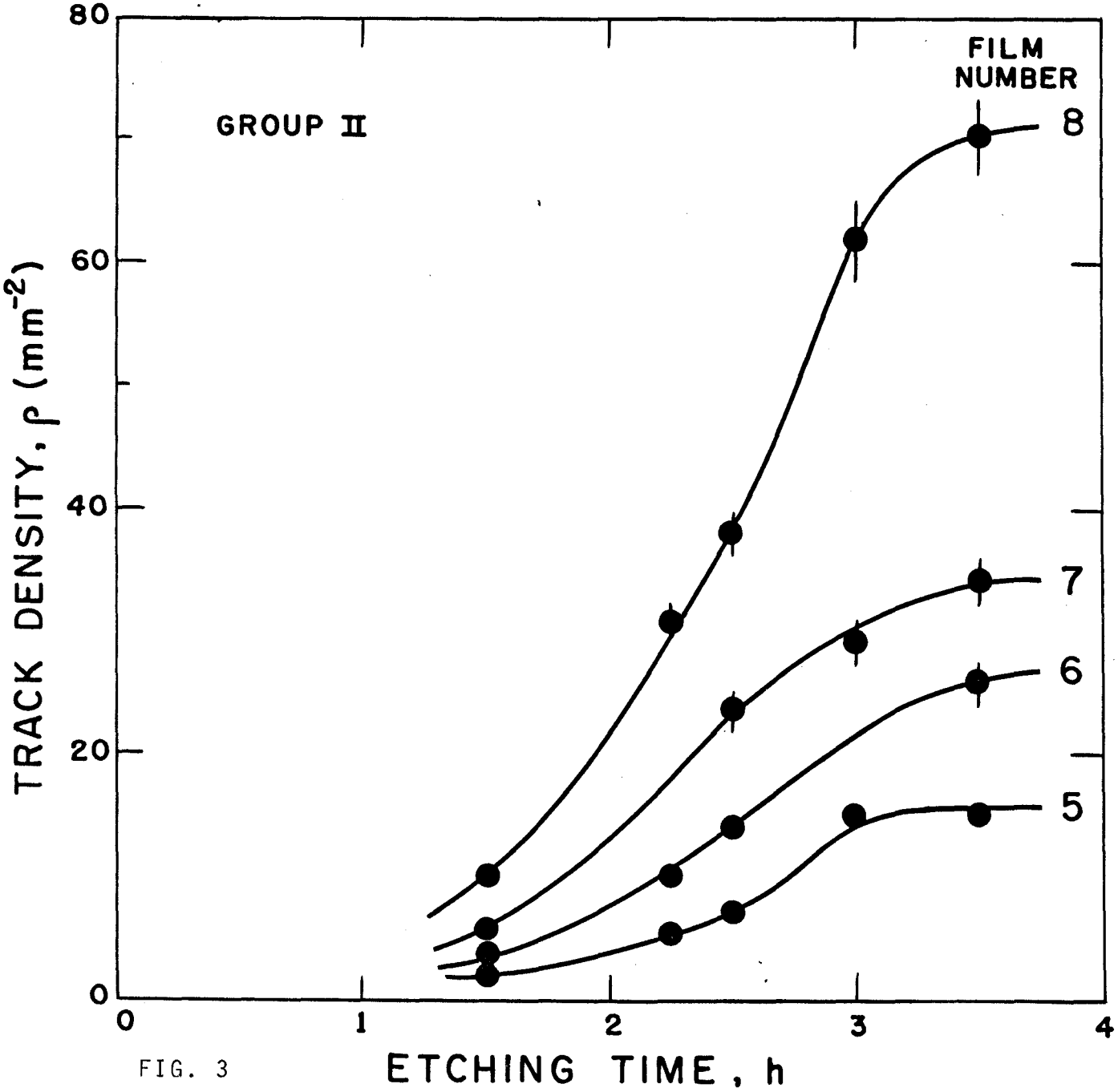
80
60
40
20
0
0
1
2
3
4
GROUP II
FILM NUMBER
8
7
6
5
TRACK DENSITY, ρ (mm⁻²)
ETCHING TIME, h

FIG. 3

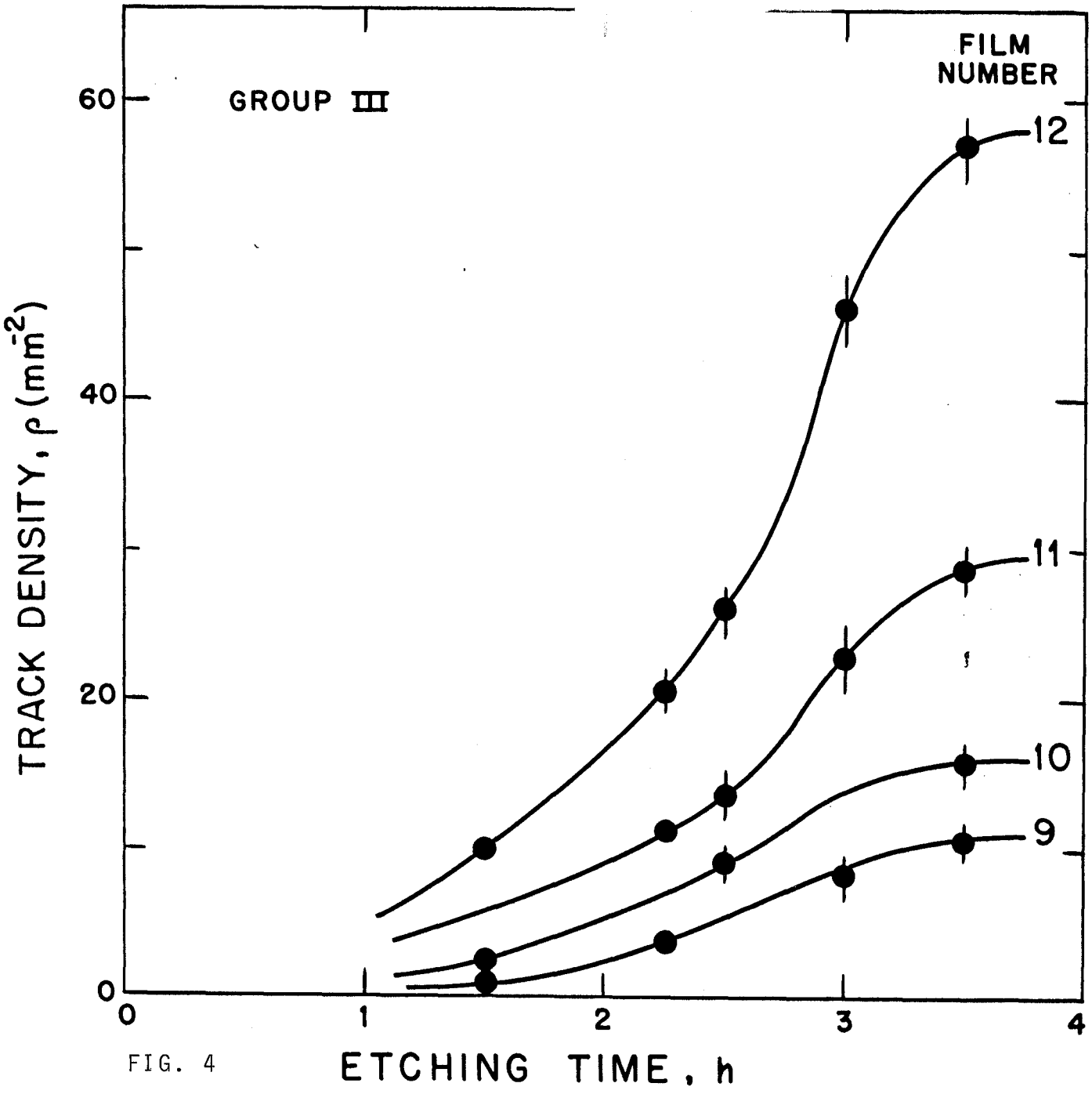
GROUP III
FILM NUMBER
12
11
10
9
60
40
20
0
0
1
2
3
4
TRACK DENSITY, ρ (mm⁻²)
ETCHING TIME, h

FIG. 4

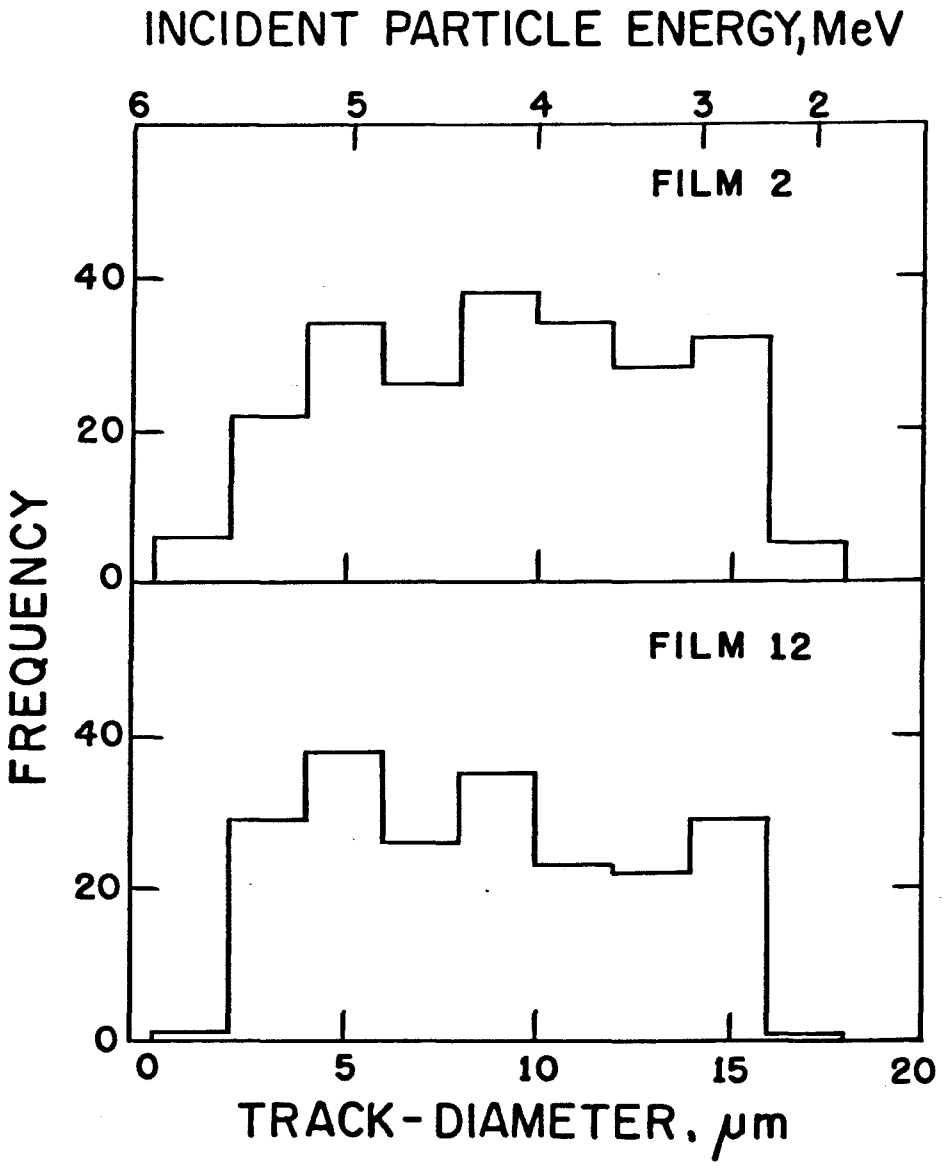
INCIDENT PARTICLE ENERGY, MeV
6
5
4
3
2
FILM 2
40
20
0
FILM 12
40
20
0
FREQUENCY
0
5
10
15
20
TRACK-DIAMETER, μm

FIG. 5

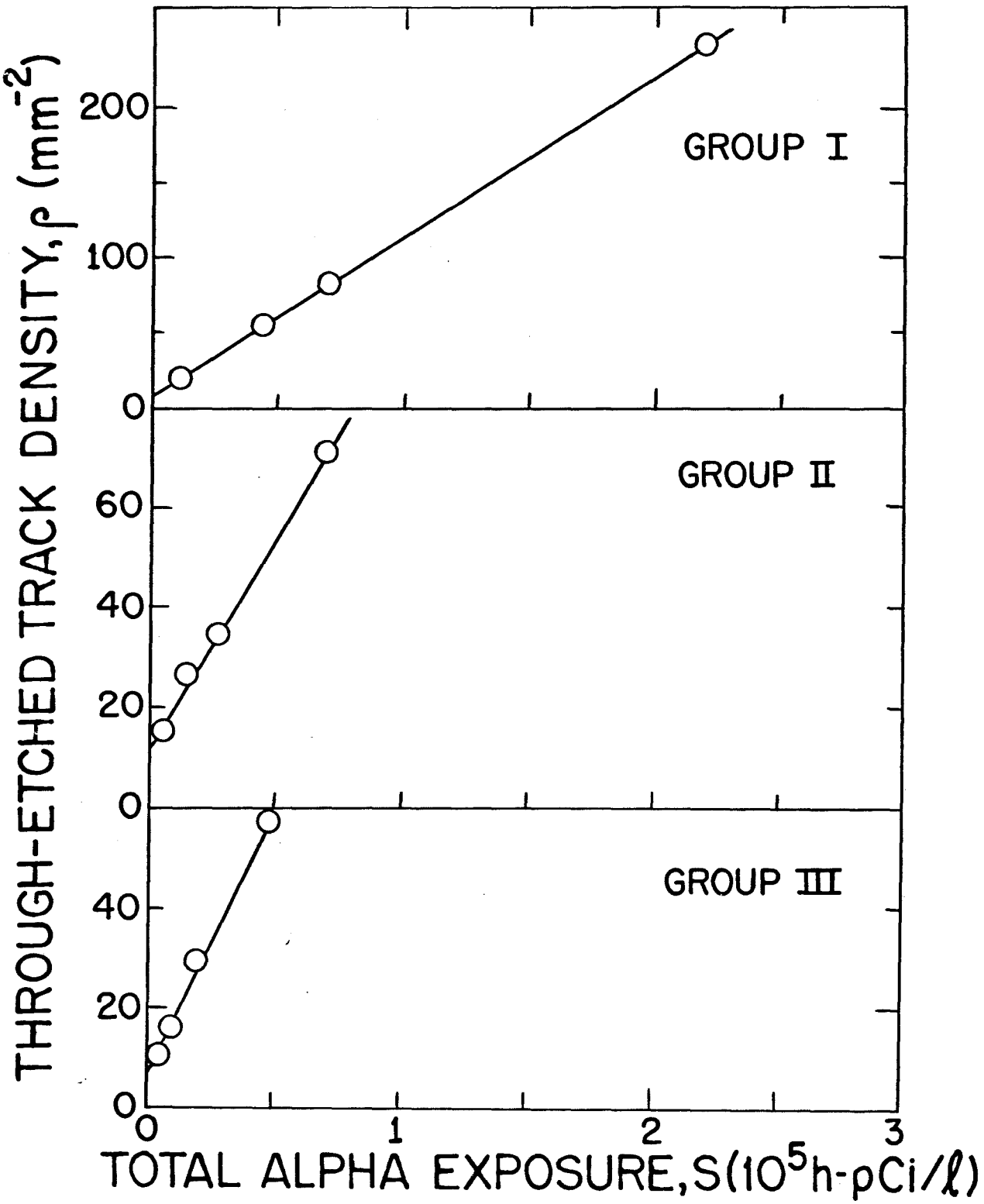
THROUGH-ETCHED TRACK DENSITY, ρ (mm⁻²)
200
100
0
GROUP I
60
40
20
0
GROUP II
40
20
0
GROUP III
0
1
2
3
TOTAL ALPHA EXPOSURE, S(10⁵h-pCi/ℓ)

FIG. 6

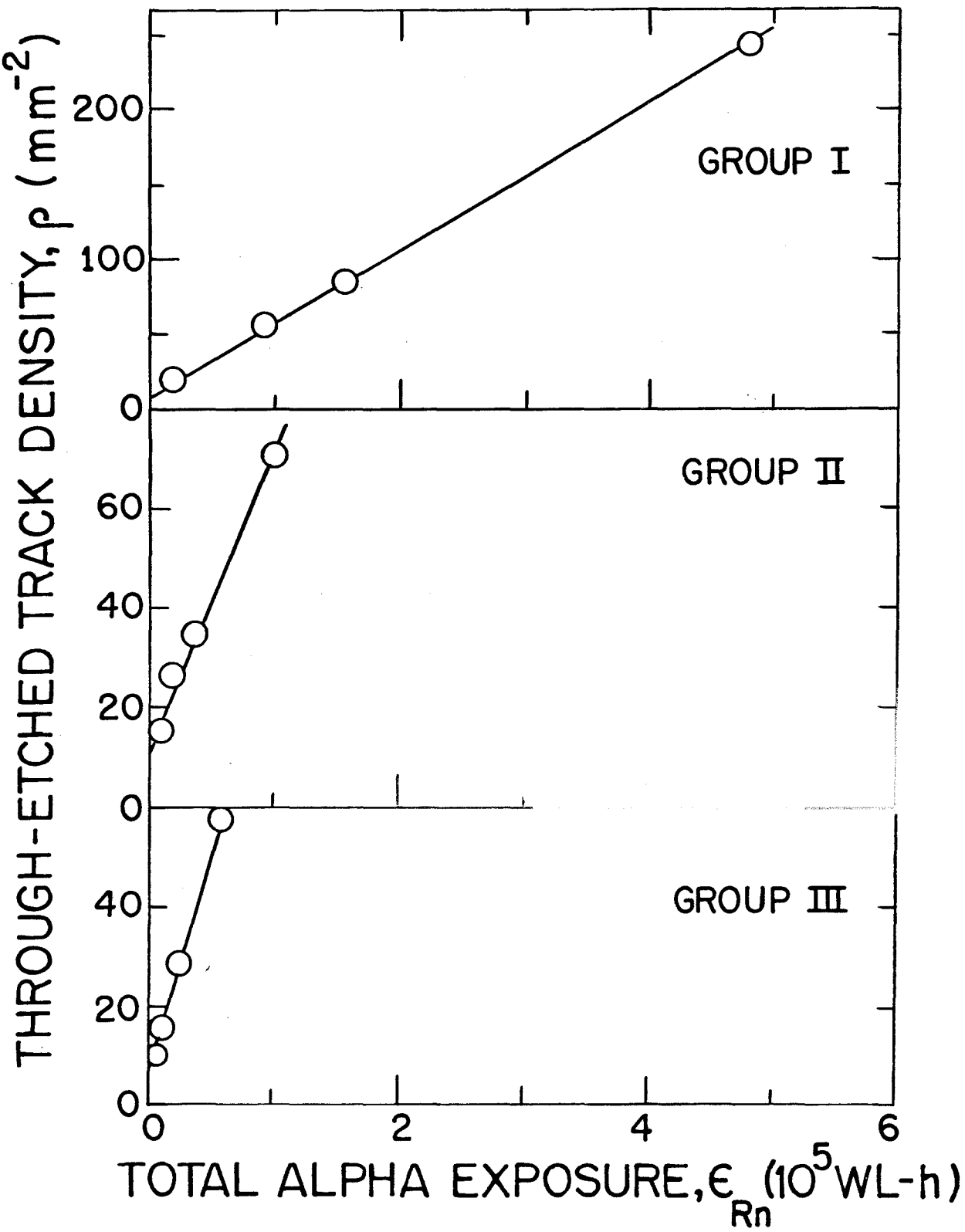
THROUGH-ETCHED TRACK DENSITY, ρ (mm⁻²)
200
100
0
GROUP I
GROUP II
60
40
20
0
GROUP III
40
20
0
0
2
4
6
TOTAL ALPHA EXPOSURE, ε_Rn (10⁵ WL-h)

FIG. 7

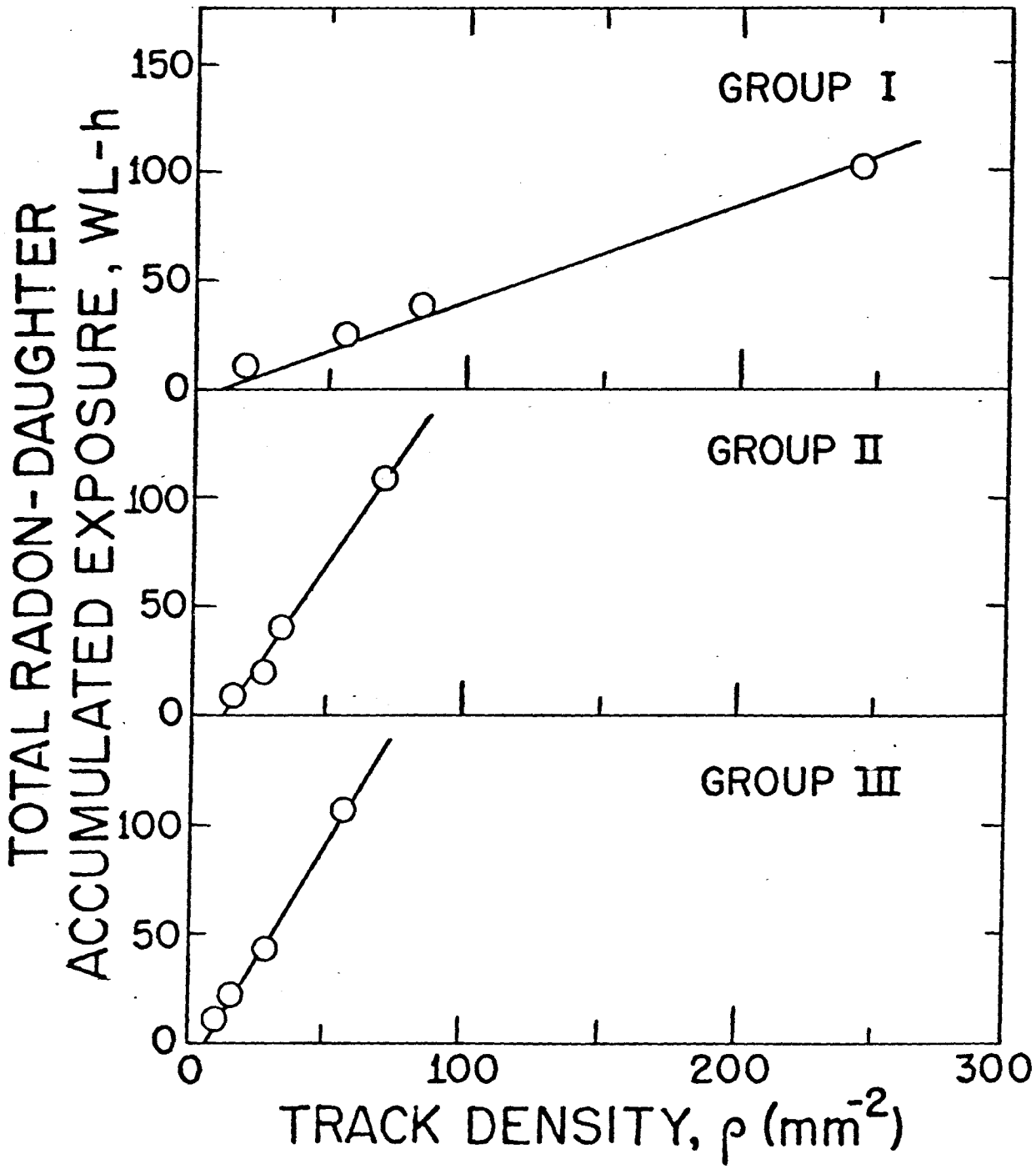
TOTAL RADON-DAUGHTER
ACCUMULATED EXPOSURE, WL-h
150
100
50
0
GROUP I
GROUP II
GROUP III
0
100
200
300
TRACK DENSITY, ρ (mm⁻²)

FIG. 8

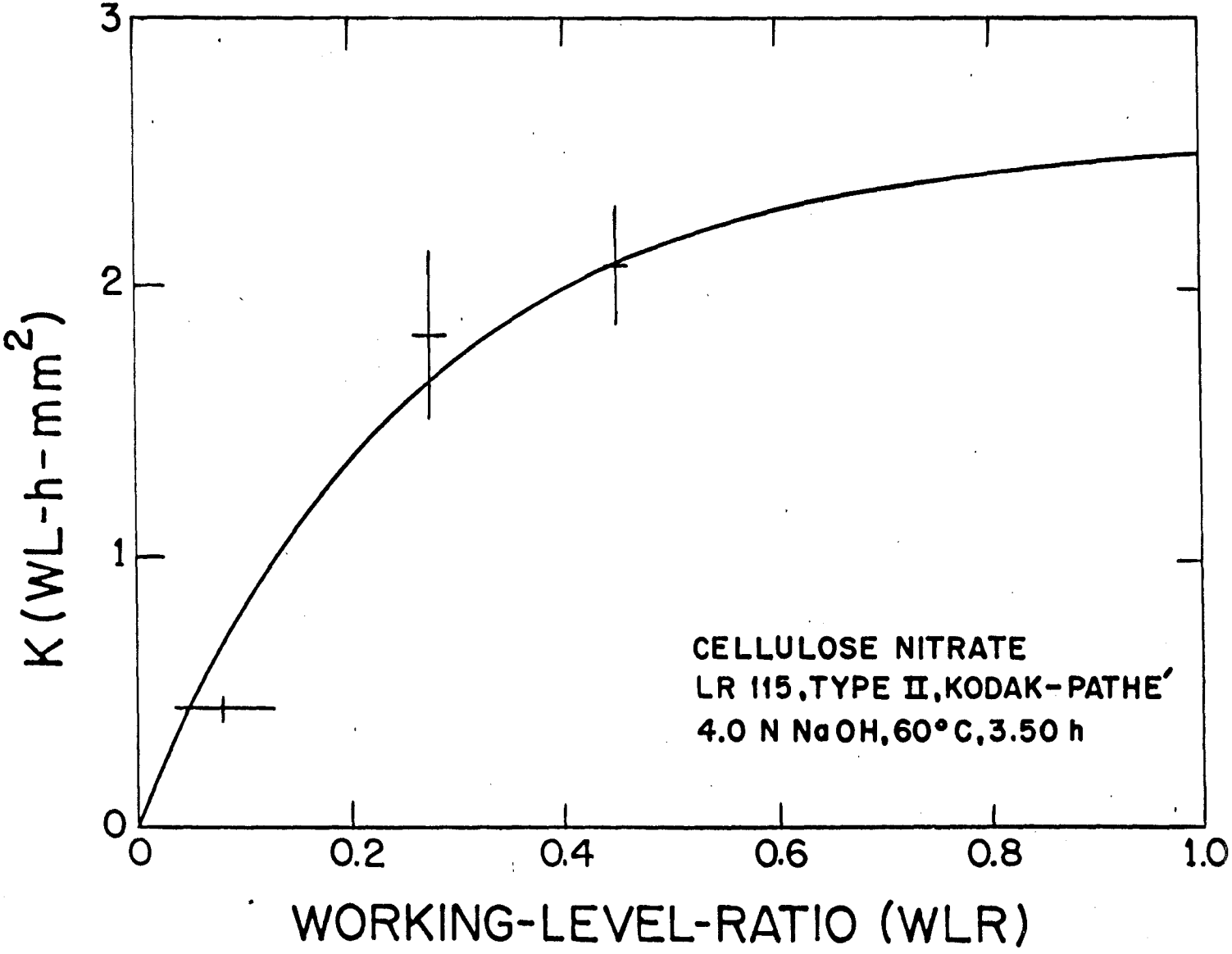
3
2
1
0
0
0.2
0.4
0.6
0.8
1.0
K (WL-h-mm^2)
WORKING-LEVEL-RATIO (WLR)
CELLULOSE NITRATE
LR 115, TYPE II, KODAK-PATHE'
4.0 N NaOH, 60° C, 3.50 h

FIG. 9