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Particle Tracks in Supralinear Nuclear Research Emulsions*

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The tracks of isolated particles in the grain-count regime in emulsion is described by a theory extended from 1-hit detectors to c-or-more hit detectors, for study of the tracks of ^{12}C, ^{16}O, and ^{20}Ne ions in a series of desensitized Ilford K-minus emulsions, as processed in a wet hot-stage (Bristol formula) developer. These emulsions represent a class of many-hit nuclear track detectors able to discriminate against low LET radiations, with a threshold that can be varied by processing, and which are able to mimic some of the aspects of the response of biological cells to radiations of different quality.

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DISCLAIMER

I. Introduction

Our identification of Ilford K-2 emulsion as as a supralinear, 8-or-more hit detector under certain developing conditions[1] has stimulated further investigation into the properties of emulsion and into the process of track formation. We are interested in the range of hittedness and the range of radiosensitivity that can be achieved with different emulsion-processing conditions, in the effect of pre- and post-irradiation conditions, and on the relation of these parameters to radiobiology and to the nature of the photographic process.

The discovery of supralinear emulsions is especially interesting for radiobiology, to radiation therapy with high LET radiations, and to the evaluation of radiation hazards, for biological cells and tissues also exhibit supralinear response[2] to gamma-rays, and consequently display a variation in Relative Biological Effectiveness (RBE) with Linear Energy Transfer (LET) of the incident radiation. An emulsion whose response to these radiations resembles the response of human tissues would be of great value.

The problem has been approached through both theory and experiment. Emulsions have been exposed to the particle beams available at Berkeley, and developed there by use of a wet hot-stage (Bristol) developer. Tracks have been photographed, and analyzed by grain counting. Coupled with these experiments, the theory of particle tracks in supralinear emulsions has been extended to c-or-more hit detectors, paralleling the multi-target single-hit-per-target theory that was developed earlier for biological cells[2], so that we can now describe grain counts, track-width, ion-kill and gamma-kill for the c-hit detector.

In a companion paper the work on sensitometry of emulsions exposed to x-rays, and to isolated alpha-particles and fission fragments will be described. There we have evidence of the gamma-kill process in supralinear emulsion, in that emulsions so insensitive that they do not show tracks of electrons, alpha-particles, and even fission-fragments, can be blackened by x-rays. Beams of heavy ions of sufficient intensity for sensitometry at different LET values have not been available, so that the work with particle beams is restricted to the study of the tracks of individual particles, to be described here.

II. Track Theory

Track theory assumes that secondary electrons, or delta-rays, are essentially responsible for the effects observed in many detectors, after irradiation with gamma-rays, or electron beams, or heavy ions. To build a theory of track structure we must first calculate the radial distribution of "local dose" from delta-rays ejected from the region along the path of the heavy ion. Into this we must fold the properties of the detector.

We take the point distribution of local dose[1] to be given as

$$E(t) = N(e^4/mc^2)(z^2/\beta^2)(1/t^2 - 1/t\tau) \qquad (1)$$

where N is the number of electrons per $cm^3 = 1.024 \times 10^{24}$, in emulsion,

e and m are the electron charge and mass ,

c is the speed of light,

z is the effective charge number of an atom of atomic number Z , as $z = Z(1 - e^{-125\beta Z^{-2/3}})$,

β is the relative speed of the ion,

t is the radial distance from the ion's path

τ is the maximum radial projection of the fastest delta ray permitted by collision kinematics, taken here to be of energy $w_{max} = 2mc^2\beta^2/(1-\beta^2)$, and projected normally a distance $\tau = (3015 \text{ cm/erg})w_{max}$.

We take the probability that an emulsion grain will be developed to be determined by a characteristic dose E_o, at which there is an average of 1 hit per grain, the minimum number of hits per grain, c, (not the speed of light), to produce a developable latent image, and the average local dose per grain delivered in a sensitive element of radius a_o (here taken to be approximately 0.12 μm for Ilford K emulsions). We calculate $\overline{E}(t)$ by averaging $E(t)$ in near circular cylinders of radius a_o whose axis is parallel to and at radial distance t from the ion's path. So long as $\tau/a_o > 4$, we find that the value of $\overline{E}\beta^2 a_o^2/z^2$ has a value here found to be 7×10^7 erg/cm out to t approaching a_o, then declining as t^{-2} until we approach τ, as shown in Figure 1.

From Fig. 1 we see that, for $t/a_\circ < 1$

$$\bar{E}\beta^2 a_\circ^2/z^2 = 7 \times 10^{-7} \text{ erg/cm} \qquad (2)$$

and

$$\bar{E}/E_\circ = z^2/\kappa\beta^2 \quad , \qquad (3)$$

where

$$\kappa = E_\circ a_\circ^2/7\text{x}10^{-7}\text{erg/cm} \ . \qquad (4)$$

We expect the quantity κ to be a good parameter with which to describe particle tracks, in the grain-count regime, at sufficiently high particle speeds that $\tau/a_\circ > 4$, so that emulsion grains along the ion's path experience a local dose field that falls off as t^{-2}, not constrained by τ.

We take the emulsion response to be given by the cumulative Poisson distribution, where the probability $P(c,A)$ for activating an emulsion grain is

$$P(c,A) = \sum_{c=0}^{\infty} \frac{A^x e^{-A}}{x!} = 1 - e^{-A} \sum_{x=0}^{c-1} \frac{A^x}{x!} \quad , \qquad (5)$$

with

$$A = \bar{E}/E_\circ \quad . \qquad (6)$$

The cumulative Poisson Distribution is shown in Fig. 2.

When the linear density of developed grains along the path of a particle is measured, one makes an arbitrary decision as to whether a grain slightly off to the side is really part of the track. We have here chosen to include in our calculations all grains whose centers lie within an undeveloped grain diameter of the ion's path as part of the track, noting that the developed grain is several times larger than the undeveloped grain. This implies that our grain-count calculations will exhibit some track width. To illustrate, Fig. 3 shows the result of grain-count calculations for Z = 2,3,4,6,10,36 in a hypothetical emulsion for which c=4 and $E_\circ = 10^7$ erg/cm^3 and

a_o = 0.12 microns. The grain-count (per micron) is plotted as a function of z^2/β^2. Several positions, at $\beta = 0.02$, .03, .04, and .05 are flagged. Here we see the effect of the failure of scaling at low β, where $\tau/a_o < 4$; we see the appearance of the track-width regime in the heaviest particle. So long as κ is a good parameter, the grain-count is well described by Eq.5, where we now must take

$$A = z^2/\kappa\beta^2 \quad . \qquad (7)$$

Thus we may extract emulsion parameters, c and κ, by comparing a log-log plot of grain count vs z^2/β^2 to a similar plot of P(c,A), as shown in Fig. 2. We see that c is never less than the steepest slope displayed experimentally, whether or not we can see saturation in the grain-count, or whether we have fulfilled the particle speed condition.

While our present emphasis is on the relationship of the supralinear emulsion to biology, their discrimination is of potential interest for problems in physics. For example we show in Fig. 4 a computer simulation of tracks of very heavy particles in a hypothetical emulsion whose parameters are not very distant from those already available, say, with K-3 emulsion.

III. Experimental Procedures and Results

Desensitized Ilford K-minus emulsions, designated as K-1, K-1.5, K-2, and K-3, in the form of 100 and 600 micron plates, were exposed to ^{12}C and ^{16}O particles, and these plates and 600 micron pellicles were exposed to ^{20}Ne particles, in the beams available at Berkeley, and were developed there following the procedures of Table I.

Tracks of a number of stopping ^{20}Ne particles in a 100 micron plate of K-3 emulsion are shown in Fig.5. Note the variation in grain size along the track, presumably associated with the fluctuation in latent image size.

Tracks of stopping ^{20}Ne particles in a) 100 micron plates, b) 600 micron plates, and c) 600 micron pellicles of K-1, K-1.5, and K-2 emulsion are shown in Fig. 6. Although plates and pellicles of a single emulsion type were made from the same batch of sensitized material, at the same time, and were processed by the same formula (except for a longer presoak time for 600 micron emulsions) there are clear differences in the appearance of the tracks in the different media. In all cases the 100 micron plates give the slowly changing track appearance with range of a 1-hit detector (a). So also for K-1(b) and K-1.5(b), the 600 micron plates of these materials. The steep gradients in linear grain density in K-2(b) and (c) indicate high hittedness. Tracks K-1(c) and K-1.5(c) seem to be a combination of high hittedness response at the slowing end, at left, of high grain density, and of 1-hit response at right where the particle is fast and the grain density is low. This observation is confirmed by actual grain count in these two plates, as shown in Fig. 7, where the mixed hittedness is evident in the disjoint character of the plot of grain-count vs z^2/β^2, though we are less certain of the hittedness assignment in the slowing portion of the track. For comparison, some data for singly charged particles in K.5 emulsion, from Patrick and Barkas[4], are plotted in Fig. 8, with the superimposed line being drawn from

Fig. 2. We infer that the differences in track appearance in the same emulsion, of different thickness, and from the high speed, dispersed grain region to the low speed region of concentrated grains, arise from "developer starvation". The consistent attainment of high hittedness and uniform radiosensitivity parameters will require careful control of both manufacture and processing. Similar effects are found in sensitometric exposures with x-rays.[7] Grain counts of ^{20}Ne tracks in K-2 emulsion (see Fig. 6) are plotted in Fig. 9.

A preliminary analysis of some grain count measurements in different emulsions yields parameters displayed in Table II. Here we have attempted to estimate values of c, κ, and the "saturation value of the grain-count", g_0, from a few tracks in a few plates, to obtain an estimate of the range of parameters achieved in this wet hot stage development. Values in the table may be compared to the parameters of K.5 emulsion from the data of Patrick and Barkas, Fig.8, where we find c=1, κ=55, and g_0 = 700 grains/100 microns. The increased value of κ in the table reflect the decrease in emulsion sensitivity. Especially in K-2 emulsion, there is clear evidence of higher hittedness, from the Bristol processing. In all cases there is a much lower value of the saturation grain density as compared to the value from Patrick and Barkas of 700 for K.5 emulsion. For this we have no ready explanation.

IV. A Preliminary Comparison with Biology

Although there are obvious characteristic differences between biological cells and nuclear emulsions, it is of interest to compare the radiosensitivity parameters estimated here from grain-count measurements with a specific biological system for which we have extracted radiosensitivity parameters in earlier work[2,5], namely for bacterial spores irradiated in a nitrogen environment, from the work of Powers, Lyman, and Tobias[6].

There we have taken the response of the spore to be described by the multi-target model, in which the probability of inactivation is given as

$$P(m,A) = (1 - e^{-A})^m \qquad (8)$$

where m is thought to be the number of internal targets which must be hit to inactivate the spore. In these calculations cells are taken to have the energy absorption properties of water (for ionizing radiations), and Eq. 4 is written as

$$\kappa = E_0 a_0^2/2 \times 10^{-7} \text{ erg/cm.} \qquad (9)$$

Although plots of P(m,A) and P(c,A) look alike, at c=m, there is not an exact numerical coincidence. Thus if we compare the values of A for which P=0.01, we find that this is achieved at A = 0.4 for the 4-target case, and at A = 0.8 for the 4-hit case.

In the biological system we find E_0 and m from survival curves after gamma-irradiation, then find the parameters σ_0 and κ from survival curves at appropriate high LET radiations. We use Eq. 9 to infer a hypothetical value for the size of the hypothetical internal targets, from E_0 and κ. The quantity σ_0 reflects the cross-sectional area of the spore, or of the cell nucleus. In the present case the biological parameters are better and more reliable than the physical measurements.

For the emulsion we find c and κ from the measured grain-count, and use Eq. 4 to estimate E_0 from knowledge of the radius a_0 of the undeveloped grain. The cross-sectional area of the developed grain is represented by A.

For the anoxic irradiation of bacterial spores we have

$E_\circ = 5.9 \times 10^6$ erg/cm^3 (in water of density 1)

$m = 4$

$\kappa = 950$

$\sigma_\circ = 0.2\ \mu m^2$,

and calculate that the sensitive target radius is $a_\circ = 0.18\mu m$.

For 600 micron plates of K-2 emulsion bombarded with O ions (Table II), we have

$c = 4$

$\kappa = 5000$

$a_\circ = 0.12\ \mu m$

from which we estimate, from Eq. 4,

$E_\circ = 2 \times 10^7$ erg/cm^3 (in emulsion, of density 3.8)

and take the cross sectional area of the developed grain to be $A = 0.35\mu m^2$.

We find these parallels between biology and emulsion to be stimulating and encouraging. A silver bromide emulsion cannot be expected to match radiobiology in its response to arbitrary radiation fields, for it cannot be tissue equivalent. But we find the extent to which these emulsion parameters mimic biology to be quite remarkable.

V. Acknowledgements

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Captions

Fig. 1 Radial distribution of local dose $\overline{E}(t)$, deposited in a near circular cylinder of radius a_0 whose axis is parallel to and at radial distance t from the path of an ion of effective charge number z moving at relative speed β through emulsion.

Fig. 2 The cumulative Poisson distribution P(c,A) vs the number of trials A, representing the probability that c or more hits are scored when there is an average of A trials per target.

Fig. 3 Calculated values of the grain density in a 4-hit emulsion with $E_0 = 10^7$ erg/cm^3. The vertical bar at the end of each curve, at right, is at $\beta = 0.02$, while other bars are at 0.03, 0.04, and 0.05. The departure of the curves from a common functional form, when plotted as a function of z^2/β^2 is from the failure of Fig. 1 to scale properly at low β, where $\tau/a_0 < 4$.

Fig. 4 Computer simulation of the tracks of stopping heavy ions for which the atomic number and atomic weight are (54,132),(86,222), (92,238), and (114,298), in an emulsion for which c = 10 and $E_0 = 10^8$ erg/cm^3. Since the lowest calculated point is at β=0.15, the range scale should be displaced to the left about 7 microns for the Z=54 ion and about 15 microns for the Z=114 ion, and a corresponding displacement for the energy scale. It may be possible to design interesting experiments around the discriminating properties of these emulsions.

Fig. 5 Tracks of stopping Ne ions in K-3 plates, 100 microns thick.

Fig. 6 Tracks of Ne ions in K-1, K-1.5, and K-2 emulsion plates and pellicles developed with a wet hot-stage (Bristol) developer. Tracks have strikingly different appearance in (a) 100 micron plates, (b) 600 micron plates, and (c) 600 micron pellicles, even though all K-1 plates and pellicles were prepared at the same time from a single batch of emulsion, and all were developed in the same processing regimen, and so for K-1.5 and K-2. In all cases the tracks (a) are 1-hit, for the 100 micron plates. Higher hittedness in other cases may arise, in part, from developer starvation.

Fig. 7 Grain count data for tracks of ^{20}Ne ions in 600 micron pellicles of K-1 and K-1.5 emulsion are plotted against z^2/β^2, from plates in which the illustrations of Fig. 6 were taken. Note the disjoint character of the grain formation, at low speeds, at right. The grain count displays 1-hit character at high speeds, and shows higher hittedness near the stopping end.

Fig. 8 Grain counts for singly charged particles in Ilford K.5 emulsion, plotted against z^2/β^2. From Patrick and Barkas[4]. A 1-or-more hit Poisson curve is superimposed on the data points.

Fig. 9 Grain counts in K-2 emulsion of tracks of ^{20}Ne ions. See Fig. 6.

Table I

Type of Material	Water Presoak at 5°C	#Developer Presoak at 5°C	Wet Hot Stage at 22°C	†Short Stop at 5°C	Fix at 5°C	Wash at 5°C	Dry (1) at 5-10°C	Dry (2) at 10-20°C
		3.25 g/l amidol	40% of 3.25 g/l amidol	acetic acid			water 45% glycerin 5% ethanol 50%	20% by vol 5% by vol 75% by vol
100 μm plate	25 min	25 min	50 min	25 min	until clear +50% 3-6 hr	until OK by permanganate test	-	30 min
600 μm plate	150 min	150 min	50 min	150 min	until clear +50% 70-90 hr	"	150 min	150 min
600 μm * pellicle	"	"	"	"	"	"	"	"

* pellicle presoak in distilled water, mounting on glass, refrigerator drying, approx 5 hrs.

#		
distilled water	1	L
sodium sulfite anhy	7.2	g
sodium bisulfite meta	1	g
potassium bromide	0.87	g
amidol	3.25	g

†	
distilled water	1 L
acetic acid glacial	2 cc

Emulsion	K-1				K-1.5				K-2				K-3			
	ion	c	κ	g_o	ion	c	κ	g_o	ion	c	κ	g_o	ion	c	κ	g_o
100 micron plate	Ne	1	270	190	Ne	1	170	140	Ne	1	320	130	Ne	4	7000	170
	O	1	290	130	O	1	260	130	O	1	600	80	O	(no tracks)		
	C	1	310	110	C	1	470	120	C	1	1050	70	C	(no tracks)		
600 micron plate	Ne	1	480	130	*Ne	1	320	110	Ne	4	3800	200	Ne	(no tracks)		
	O	1	660	75	*O	1	900	70	O	4	5000	160	O	(no tracks)		
	*C	1	820	50	*C	1	1300	60	C	3	3200	40	C	(no tracks)		
600 micron pellicle	#Ne	1	450	50	#Ne	1	320	40	[4]Ne	4	4400	220	Ne	(no tracks)		
		+2	5800	90		+4	4000	140	[5]Ne	6	3300	140				
									[6]Ne	6	3900	180				

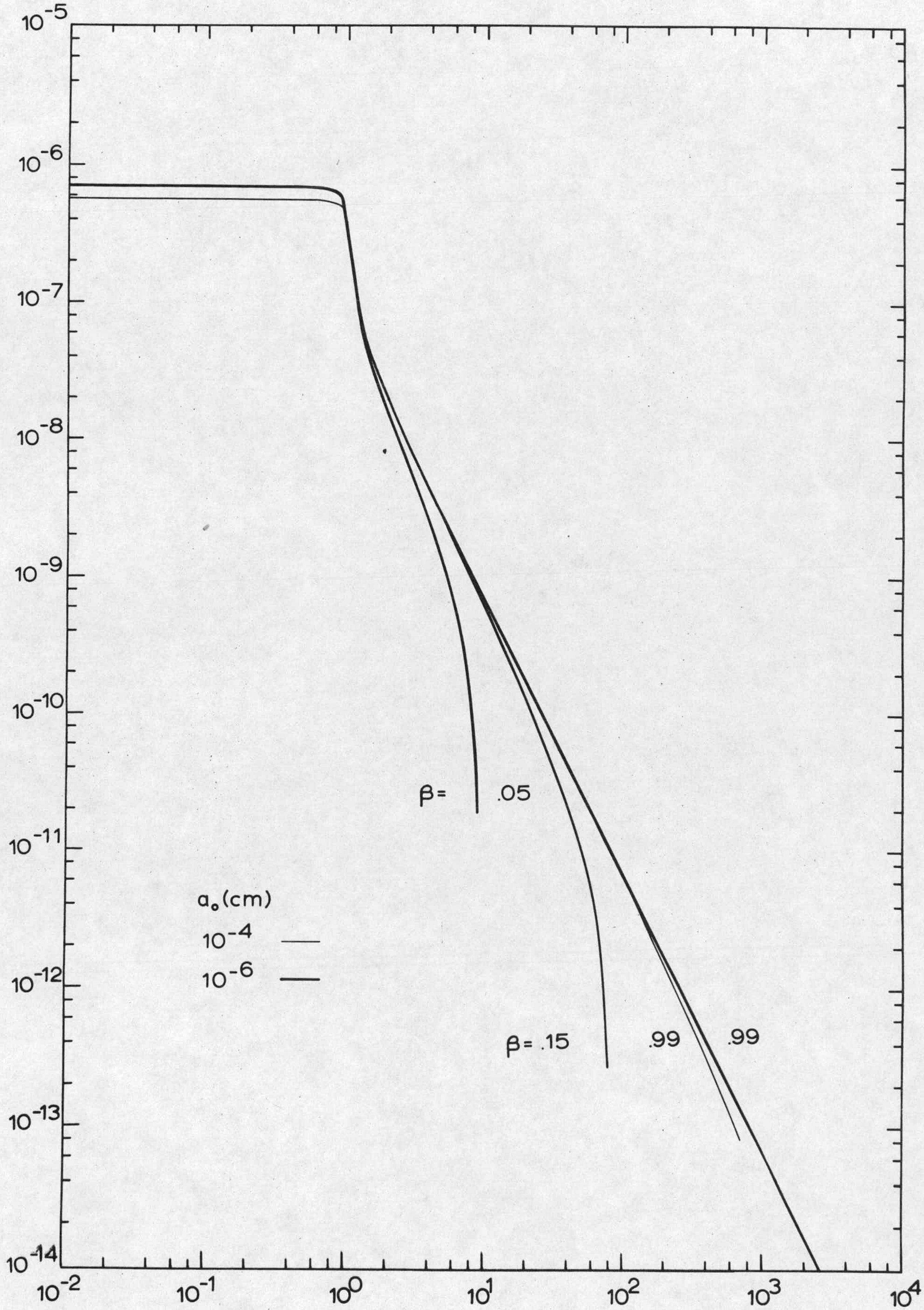
* indicates the presence of a component of higher hittedness at low β whose parameters cannot be evaluated.

higher hittedness component at low β . See Figs. 6 and 7.

[4]Ne. Tracks measured in a plate exposed to a fluence of 10^4 ions/cm^2
[5]Ne. " 10^5 "
[6]Ne. " 10^6 "

Table II

Fig 1


10^{-5}
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
10^{-11}
10^{-12}
10^{-13}
10^{-14}
$\bar{E}\beta^2 a_o^2 / z^2$ (erg / cm)
β= .05
a_o(cm)
10^{-4}
10^{-6}
β= .15
.99
.99
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
10^{3}
10^{4}
t / a_o

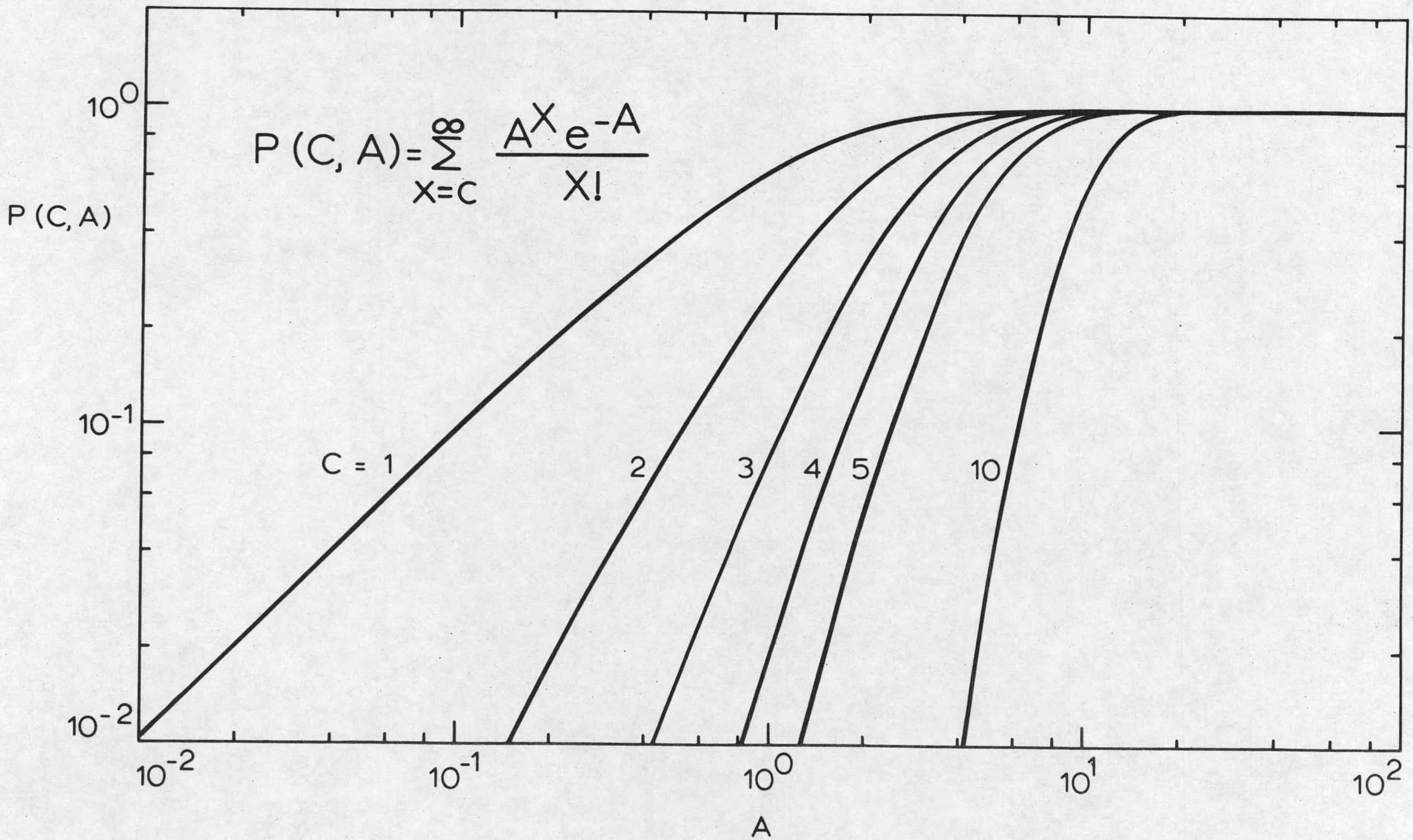
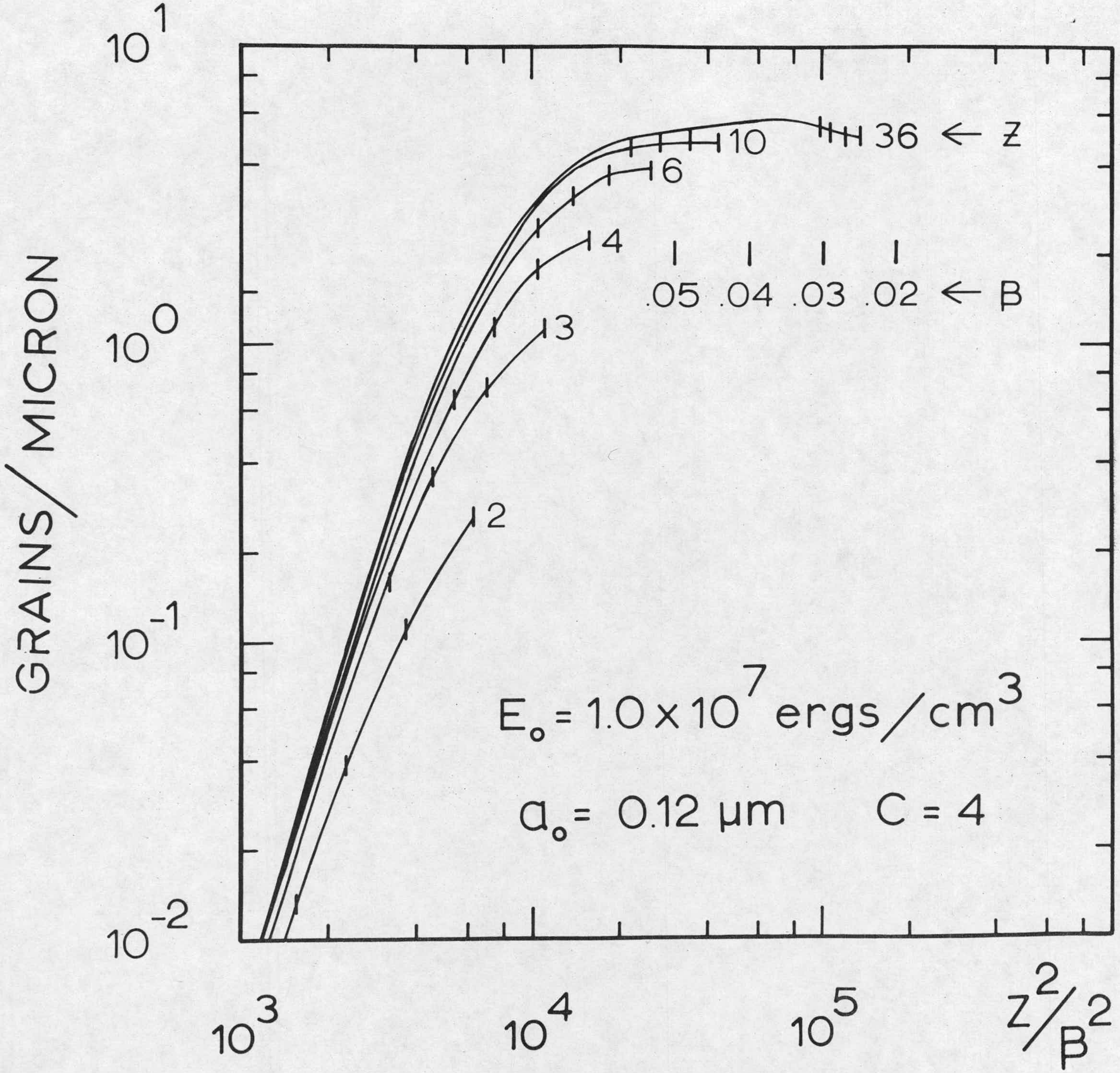

$P(C, A) = \sum_{X=C}^{\infty} \frac{A^X e^{-A}}{X!}$
P (C, A)
10^0
10^{-1}
10^{-2}
C = 1
2
3
4
5
10
10^{-2}
10^{-1}
10^0
10^1
10^2
A

Fig 2



GRAINS / MICRON
10^1
10^0
10^-1
10^-2
10^3
10^4
10^5
Z²/β²
36 ← Z
10
6
4
3
2
.05 .04 .03 .02 ← β
E₀ = 1.0 x 10⁷ ergs/cm³
a₀ = 0.12 μm C = 4

Fig 3

SIMULATED PARTICLE TRACKS

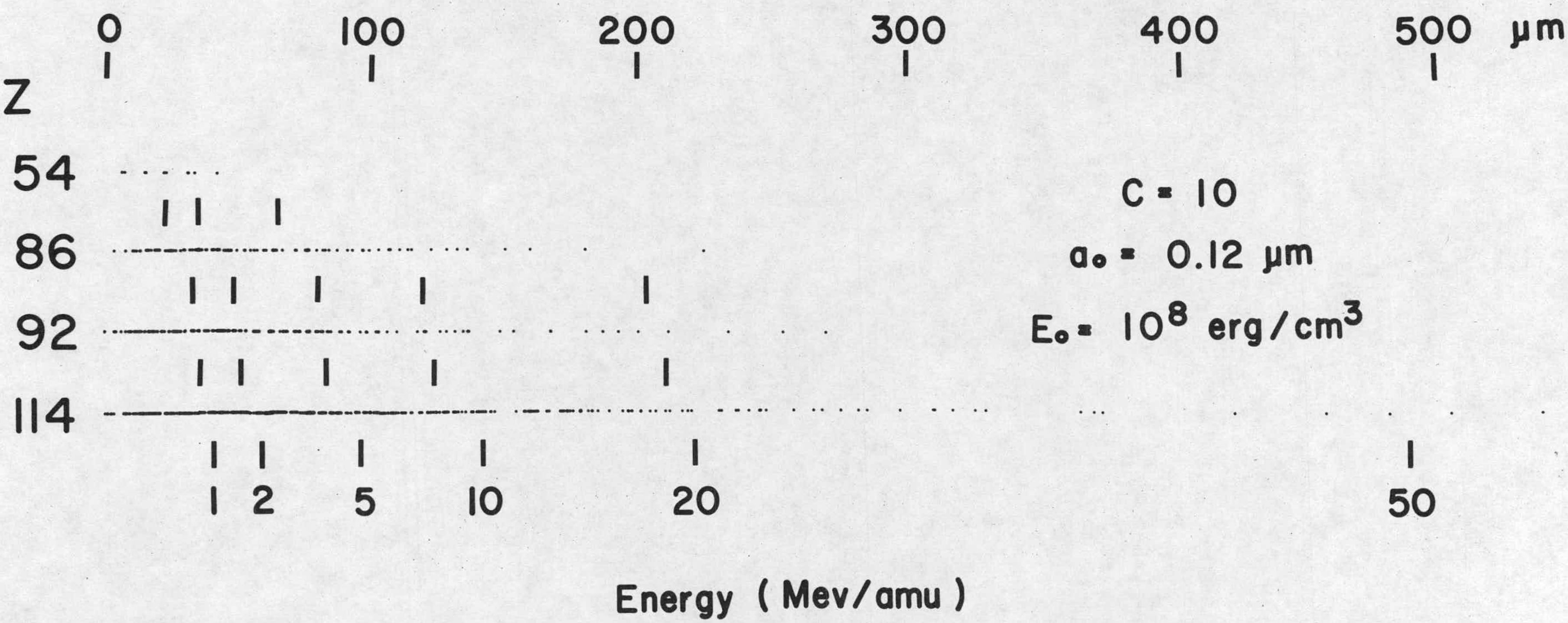
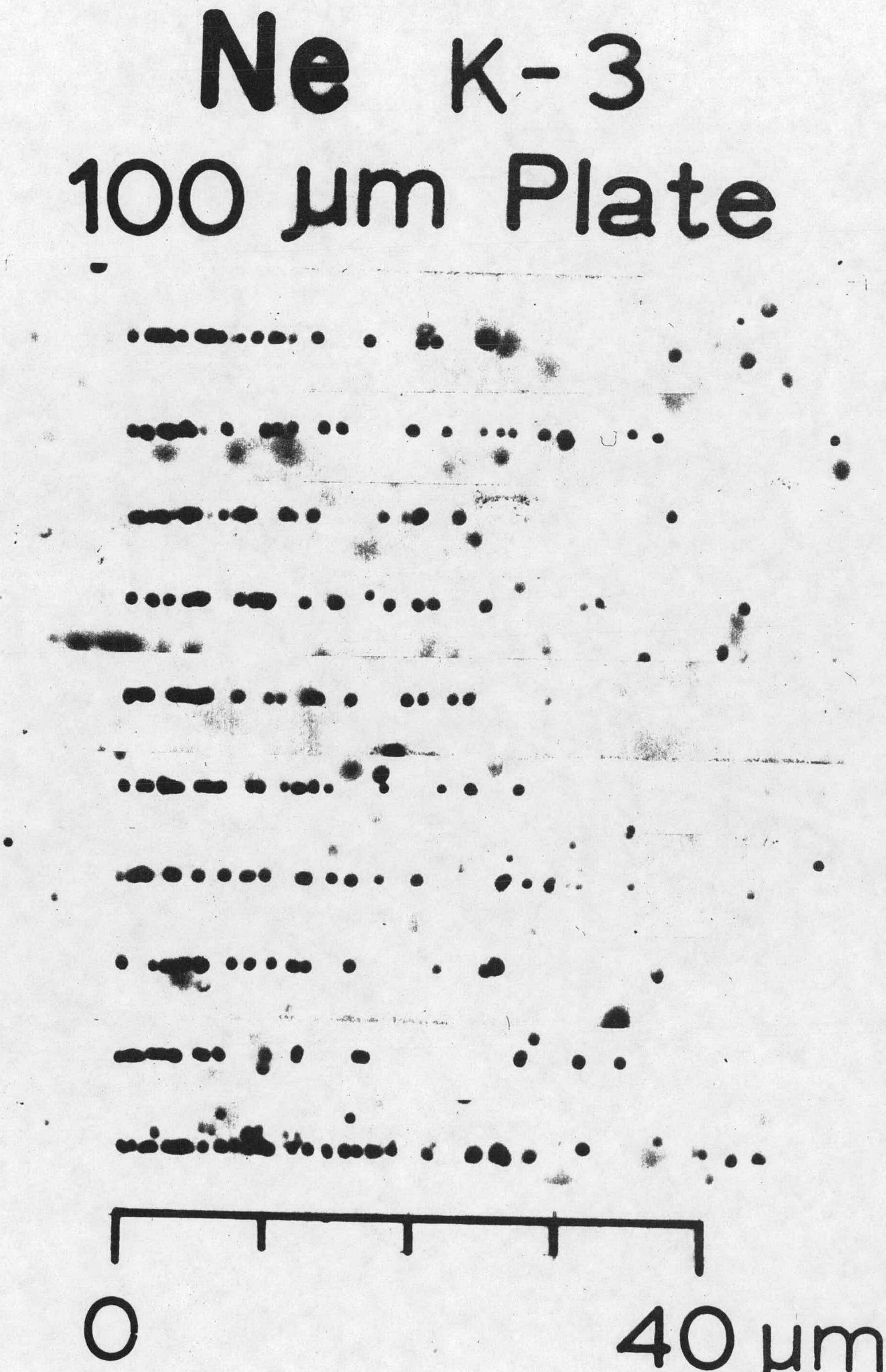


Fig. 4


Ne K-3
100 µm Plate
0
40 µm

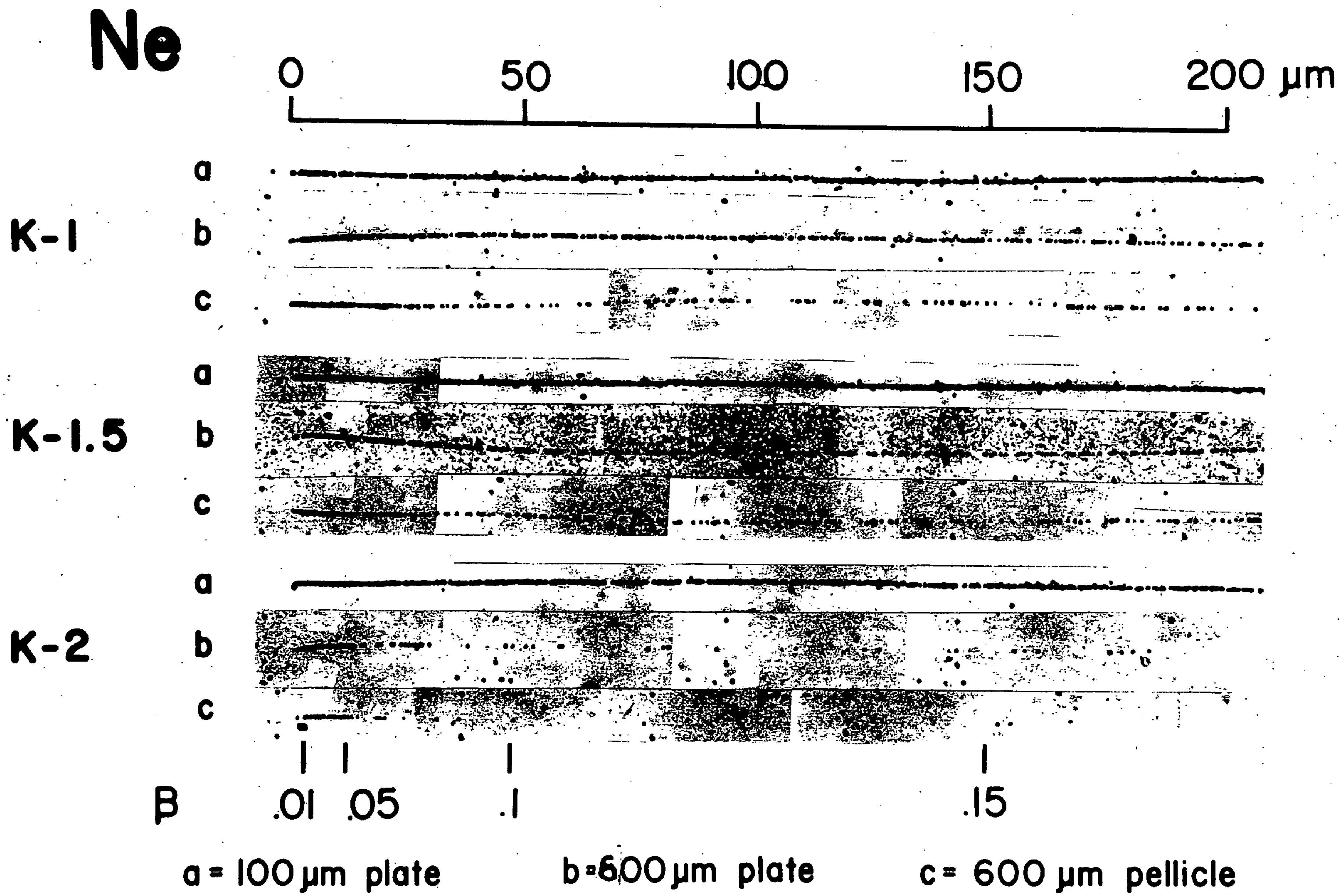
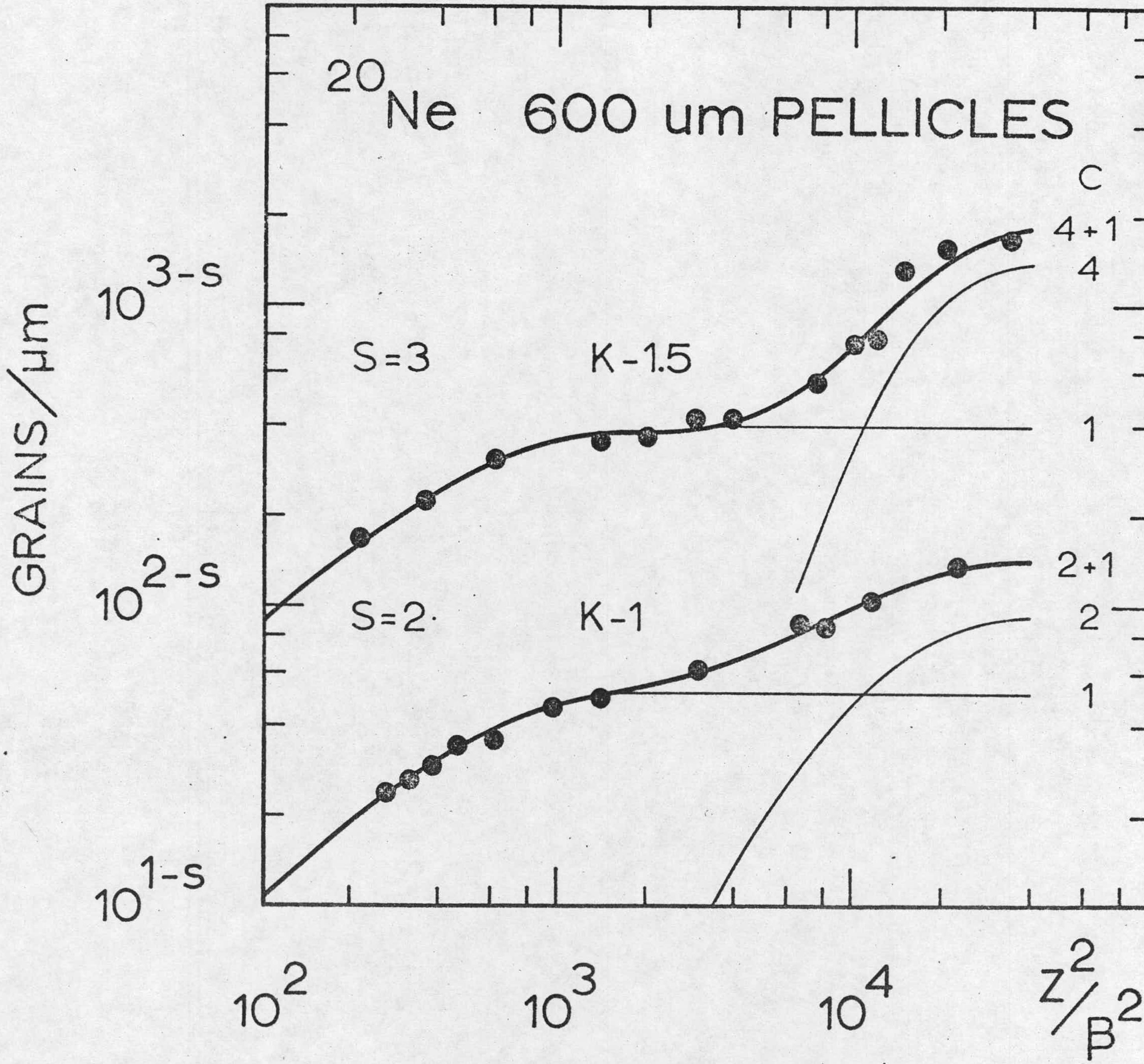

Ne
0
50
100
150
200 μm
K-1
K-1.5
K-2
a
b
c
β
.01
.05
.1
.15
a = 100 μm plate
b = 600 μm plate
c = 600 μm pellicle

Fig 6



^{20}Ne 600 um PELLICLES
GRAINS/μm
10^{3-s}
10^{2-s}
10^{1-s}
S=3
K-1.5
S=2
K-1
C
4+1
4
1
2+1
2
1
10^2
10^3
10^4
Z^2/β^2

Fig. 7

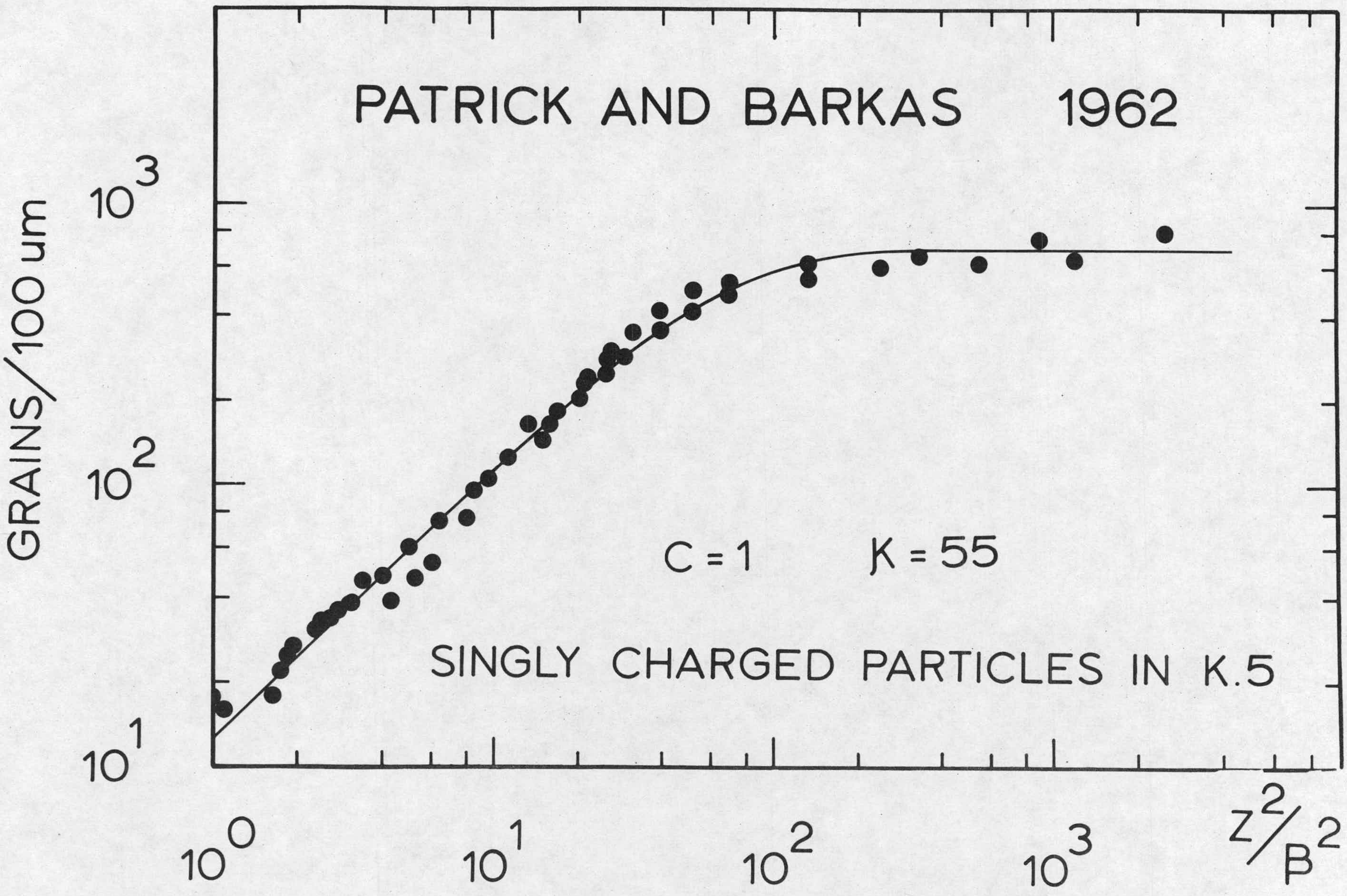

PATRICK AND BARKAS 1962
GRAINS/100 um
C = 1 K = 55
SINGLY CHARGED PARTICLES IN K.5
Z^2/β^2
10^0 10^1 10^2 10^3
10^1 10^2 10^3

Fig 8

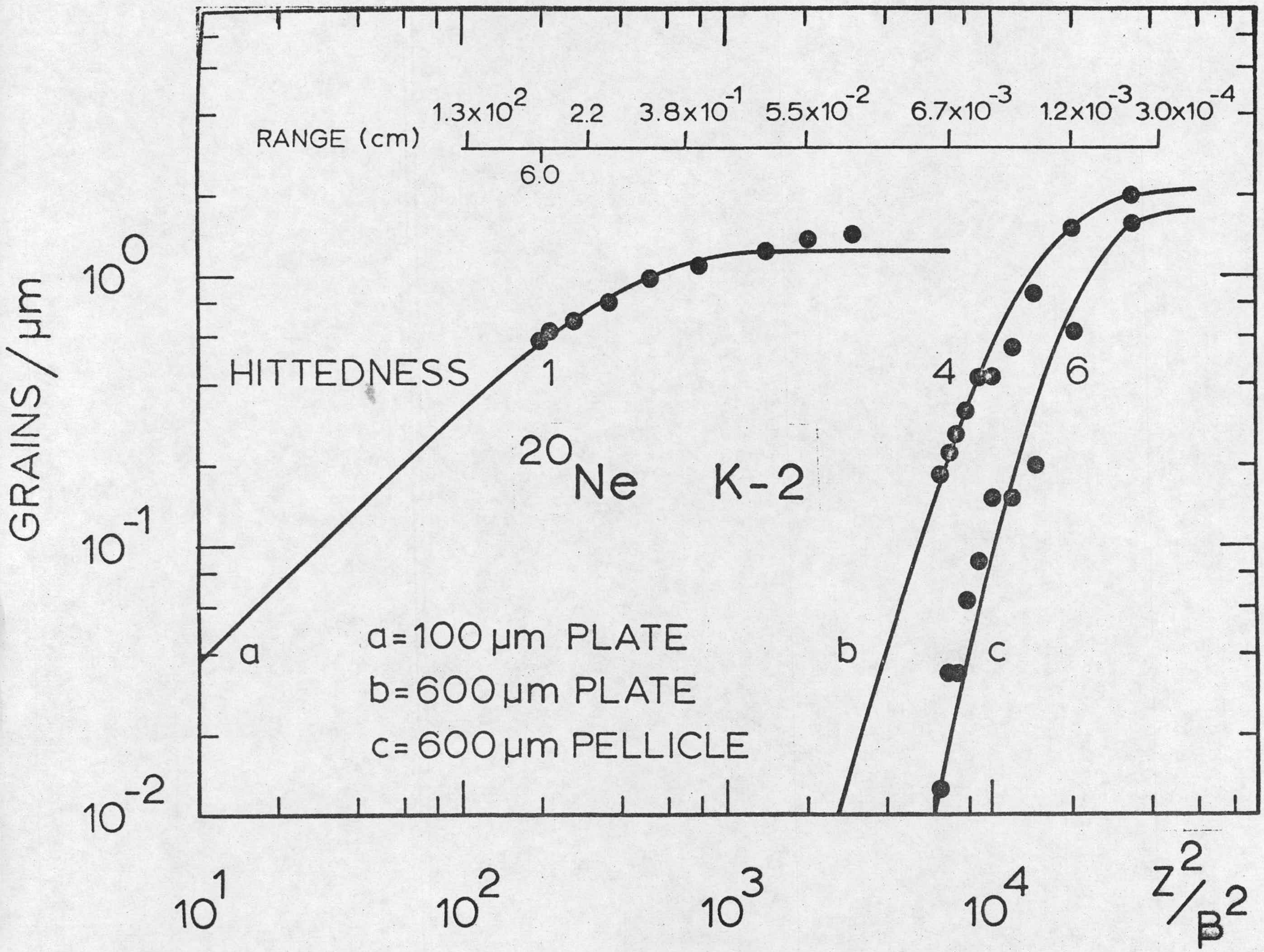

RANGE (cm)
1.3x10^2
6.0
2.2
3.8x10^-1
5.5x10^-2
6.7x10^-3
1.2x10^-3
3.0x10^-4
HITTEDNESS
1
4
6
20Ne K-2
a
b
c
a=100 μm PLATE
b=600 μm PLATE
c=600 μm PELLICLE
GRAINS / μm
10^0
10^-1
10^-2
10^1
10^2
10^3
10^4
Z^2/B^2

Fig 9