GENERAL ELECTRIC
RESEARCH AND DEVELOPMENT CENTER
SCHENECTADY, NEW YORK

STUDY OF HIGH RESOLUTION PLASTIC TRACK DETECTORS OF HEAVY COSMIC RAYS

P.B. Price, R.L. Fleischer, and A.A. Miller

Final Report
Contract No. NAS 9-8868
August, 1969

Prepared by

General Electric Research and Development Center
Schenectady, New York

for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
Manned Spacecraft Center
Space Sciences Procurement Branch
Houston, Texas

GENERAL ELECTRIC COMPANY

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CONTENTS

PREFACE

The goals of this contract have been achieved. The major scientific results have been reported in three papers attached to this report; in one patent application by A. A. Miller; and in invited talks given by P. B. Price at an International Conference on Nuclear Tracks in Solids held in May at Clermont-Ferrand, France, and the the Summer Meeting of the American Physical Society in Rochester, and by R. L. Fleischer at the Gordon Research Conference on Nuclear Chemistry.

Section I of this report is a detailed discussion of the effects of ionizing radiation on the chemistry of Lexan polycarbonate and other track-recording plastics. Section II is the text of an invited talk on particle identification by dielectric track detectors given at Clermont-Ferrand. Sections III and IV are papers to be given in Budapest and to appear in a special issue of Acta Physica Hungarica containing the proceedings of the Eleventh International Conference on Cosmic Rays. Section V briefly summarizes the results of our studies and makes certain recommendations for future work.

We wish to thank the NASA Manned Spacecraft Center for its support during the contract period. We also wish to acknowledge the excellent cooperation and continued encouragement of the contract monitor, Dr. D. E. Hagge.

I. EFFECT OF IONIZING RADIATION ON SURFACE ETCH RATES OF PLASTICS

I. Effect of Ionizing Radiation on Surface Etch Rates of Plastics

It has been pointed out[1] that there is a qualitative correlation between the sensitivity of plastic track detectors toward heavily ionizing particles and their susceptibility to general radiation damage by lightly ionizing radiation (i.e., γ, X-rays, and high-energy electrons). For example, cellulose polymers are much more sensitive in track detection toward light ions than is Lexan polycarbonate. Correspondingly, the radiation yields for chain-scissioning by γ- or high-energy electron irradiation are ~5 to 11 scissions/100 ev for cellulose polymers[2] and only ~0.1 for polycarbonate[3]. If the plastic film is uniformly irradiated with high-energy electrons, the surface etch-rate as a function of irradiation dose should be related to the etch-rate in a particle track at the same "average" energy deposition by the secondary electrons.

In this phase of our study, various plastic films (2.0 x 3.5 cm, 5 to 11 mils thick) were irradiated with 1.5 Mev electrons from the G.E. Resonant Transformer at a dose rate of 20 Mr/minute (Mr=10^6r). The irradiations were done at about 25°C on a water-cooled block and in an atmosphere of N_2. The samples were stored in N_2 until weighed and then immersed in 6N aqueous KOH at room temperature (26 ± 1°C). After the appropriate etch-time, the films were rinsed in distilled water, dried in a vacuum oven at 50-60° for at least 1 hour and reweighed. The etch rates are reported as the weight loss per unit area per hour, considering the total area of the two surfaces (14 cm^2). The etch rates will be designated as V_D for irradiated samples and V_o for the unirradiated controls.

A. Daicell Cellulose Nitrate (CN)

This film contained camphor as a plasticizer, the amount of

which was not specified by the supplier. A sample was dissolved in acetonitrile and the infrared absorbance at 5.75μ, characteristic of the carbonyl group in camphor, was compared with that of a known concentration of camphor in the same solvent. The camphor content of the Daicell CN was thus found to be about 23% by weight.

The etch-times for Daicell CN were 1 hour or less, depending upon the irradiation dose. Fig. 1 is a semi-log plot of V_D/V_o against dose. The points at 300 and 400 Mr are based on times for complete disintegration of the samples (4.5 and 2.5 minutes, respectively) in the etching solution. Up to about 50 Mr, an exponential relationship, $V_D/V_o = e^{kD}$, applies, where $k \simeq 0.046$/Mr and D is the dose in Mr.

B. Cellulose Triacetate (CTA)

Unplasticized 5 mil film, Kodacel (Experimental), was supplied by the Eastman Chemical Company. Fig. 2, Curve A, shows the results based on 20 hour etch-times (0-20 Mr) or less at the higher doses. As before, the values at the highest doses, 200 and 400 Mr, are based on times for complete disintegration of the films (22 and 5 minutes, respectively). For the linear portion up to about 50 Mr, we find $k \simeq 0.05$, very similar to the value for Daicell CN.

C. Plasticized Cellulose Triacetate (CTA + P)

The nature and amount of plasticizer in this film (Kodacel TA-401) were not divulged by the supplier. The results of the etch measurements are shown in Fig. 2, Curve B. Up to 50 Mr, $k \simeq 0.031$/Mr, significantly lower than for either CN or CTA itself, suggesting that the plasticizer was providing some protection against radiation damage of the CTA.

Lower aliphatic esters of phthalic acid are commonly used as

plasticizers for cellulose triacetate[4]. The present film gave, upon burning, a dense black smoke, typical of aromatic compounds (i.e., benzene) and absent in the unplasticized CTA. Nuclear magnetic resonance measurements on methylene chloride solutions of this film confirmed the presence of an aromatic component.

Films were cast from methylene chloride solutions of unplasticized CTA, one containing 15 weight percent of dimethyl phthalate (DMP) and a second containing the same concentration of glycerol triacetate (TA). Both were dried simultaneously at 50° in a vacuum oven for 2 hours. The etch rates are compared below.

Table 1. Effect of Plasticizer Type on Etch Rates of CTA

	CTA+DMP	CTA+TA
V_o, mg/cm^2/hr	0.072	0.33
$V(20\ Mr)/V_o$	1.3	2.6
$V(50\ Mr)/V_o$	3.7	5.6

The V_D/V_o ratios for CTA + DMP are very close to those for the plasticized Kodacel TA-401 at the same doses. Also, the higher values for CTA + TA indicate that the sensitivity of plasticized CTA in particle track detection should be greater if the plasticizer is aliphatic (i.e., TA) rather than aromatic (i.e., DMP).

D. Polycarbonates

Ten mil films cast from 10% methylene chloride solutions of Lexan bisphenol-A polycarbonate (PC) were used. In an effort to increase the sensitivity of PC in particle track detection, a halogenated additive, pentachlorobiphenyl (Aroclor 1254, Monsanto) was incorporated at a 10 weight percent concentration. The film

was dried at 50°C for 3 hours in a vacuum oven. Although a very high V_D/V_o ratio was initially observed and reported[5], it was subsequently found that this enhancement may have been largely due to residual methylene chloride solvent, amounting to as much as 6 to 7 weight percent.

Blends of an equi-molar copolymer of tetrachlorobisphenol-A and bisphenol-A polycarbonate (Cl_4PC-PC) with PC itself, covering a range of 5 to 10 weight percent chlorine and more thoroughly dried of solvent, showed only a small increase in etch rates, $V_D/V_o \simeq 2\text{-}3$ compared to $V_D/V_o \simeq 1.2\text{-}1.3$ for PC itself at a dose of 200 Mr. Analogous bromine-containing polycarbonates showed an even smaller enhancement of the V_D/V_o ratio. For Lexan polycarbonate itself, $V_D/V_o = 1.2\text{-}1.3$ at 200 Mr and 1.9 at 400 Mr giving, approximately, $k \simeq 0.0016$, about 1/30 the values for CN and unplasticized CTA.

The following etch-rates for unirradiated films of chlorinated Lexan compositions in 6N aqueous KOH and 3N methanolic KOH were measured.

Table 2. Etch Rates for Chlorinated Lexan Compositions (Unirradiated)

		Etch Rate, mg/cm^2/hr.	
Composition	%Cl	6N KOH(aq.)	3N KOH(CH_3OH)
PC	0	~0.008	12.8
PC+Cl_4PC-PC	5	-	7.1
" "	8	~0.005	6.3
Cl_4PC-PC	22	~0.003	1.2
Cl_4PC	36	~0.0014	0.03

There is an obvious trend of decreasing etch-rate (i.e., increasing hydrolytic stability) with increasing chlorine content,

the effect being much more pronounced for the methanolic KOH etchant. Apparently, the chlorine substituents, adjacent to the carbonate linkage being hydrolyzed, lower the hydrolysis rate through steric hindrance.

Finally, a new and unexpected effect of ionizing radiation on halogenated bisphenol-A polycarbonate compositions was discovered. Following the KOH etching measurements on the irradiated samples, they were tested for solubility in methylene chloride solvent and in some cases crosslinking was evident. The various compositions were then systematically examined for radiation-crosslinking and the results are described in the appended Patent Letter for the chlorinated materials. Subsequent tests on brominated compositions were made with similar results, as shown in the following table.

Table 3. Radiation-Crosslinking of Brominated Lexan Compositions: Area Swelling Ratios

Composition, Parts by Weight		%Br	Dose, Mr			
			20	50	100	200
Br_4PC		~51	*	*	*	*
PC+Br_4PC-PC						
0	1	37	*	(gel)	8.0	~7
2.7	1	10	(gel)	9.6	6.6	5.3
6.3	1	5	*	9.4	8.2	5.8

Note: *indicates complete solubility in methylene chloride - no gel.
(gel) indicates highly swollen, formless gel whose dimensions could not be measured.

One of the objectives of this portion of the program was to increase the radiation damage (i.e., chain-scissioning) and hence

etch rates of Lexan polycarbonate by incorporating the halogenated polymers, to approach the sensitivities of cellulose nitrate or triacetate. Actually, uniform electron-irradiation gives the inverse effect (i.e., crosslinking) with relatively little increase in etch rates! The response of these halogenated polycarbonates toward more highly ionizing particles (i.e., helium ions) remains to be investigated.

References

(1) See, for example, R. Katz and E.J. Kobetich, Phys. Rev. 170, 401 (1968).

(2) T.A. Chamberlin and G.L. Kochanny,Jr., Macromolecules 2, 88 (1969).

(3) J.H. Golden and E.A. Hazell, J. Poly. Sci. Part A 1, 1671 (1963).

(4) See W.D. Paist, "Cellulosics" Reinhold Pub. Co., N.Y. (1958) p. 29.

(5) Monthly Progress Report No. 3.

FIG. 1 ETCH RATES FOR CN (DAICELL®)

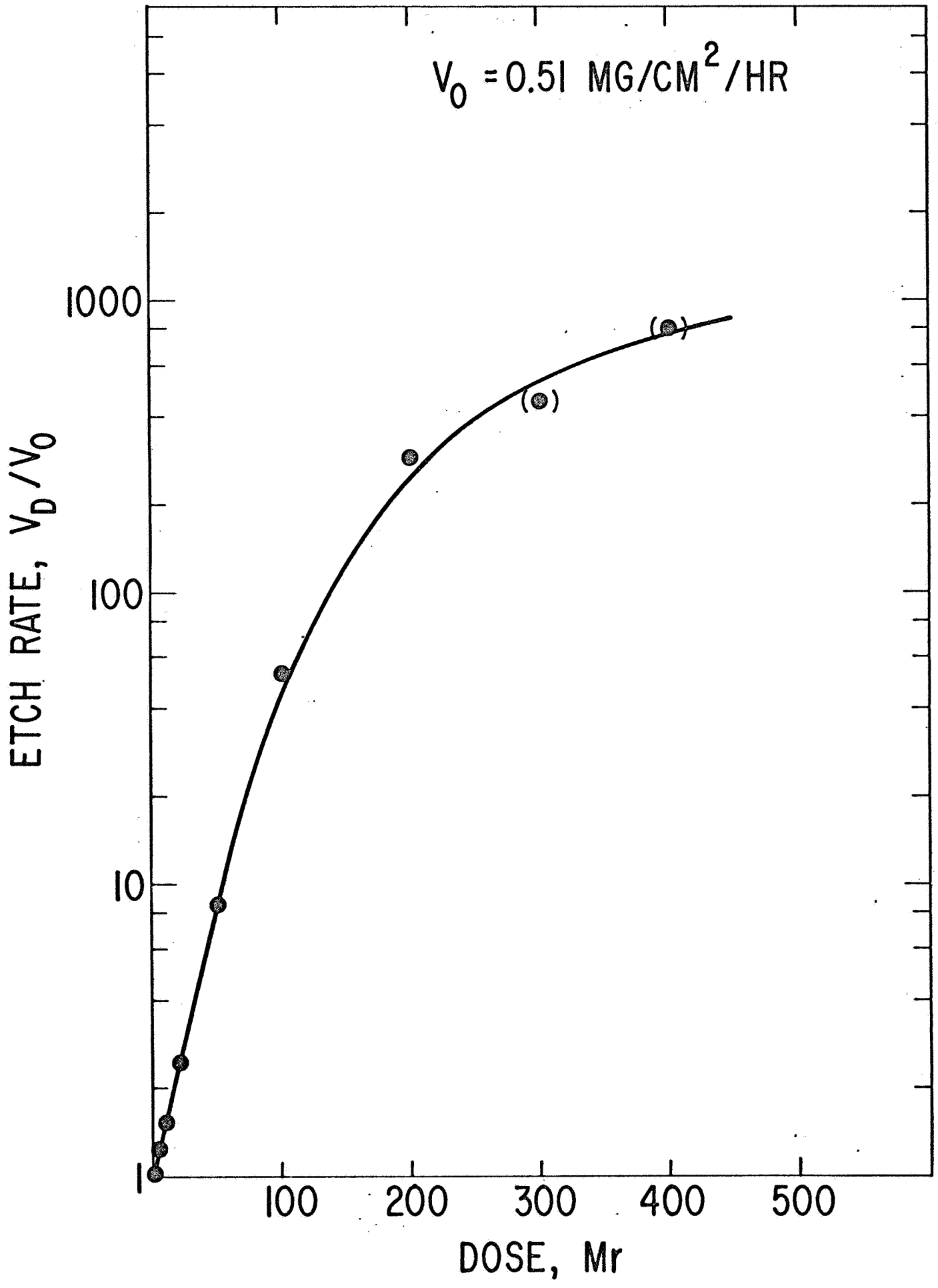
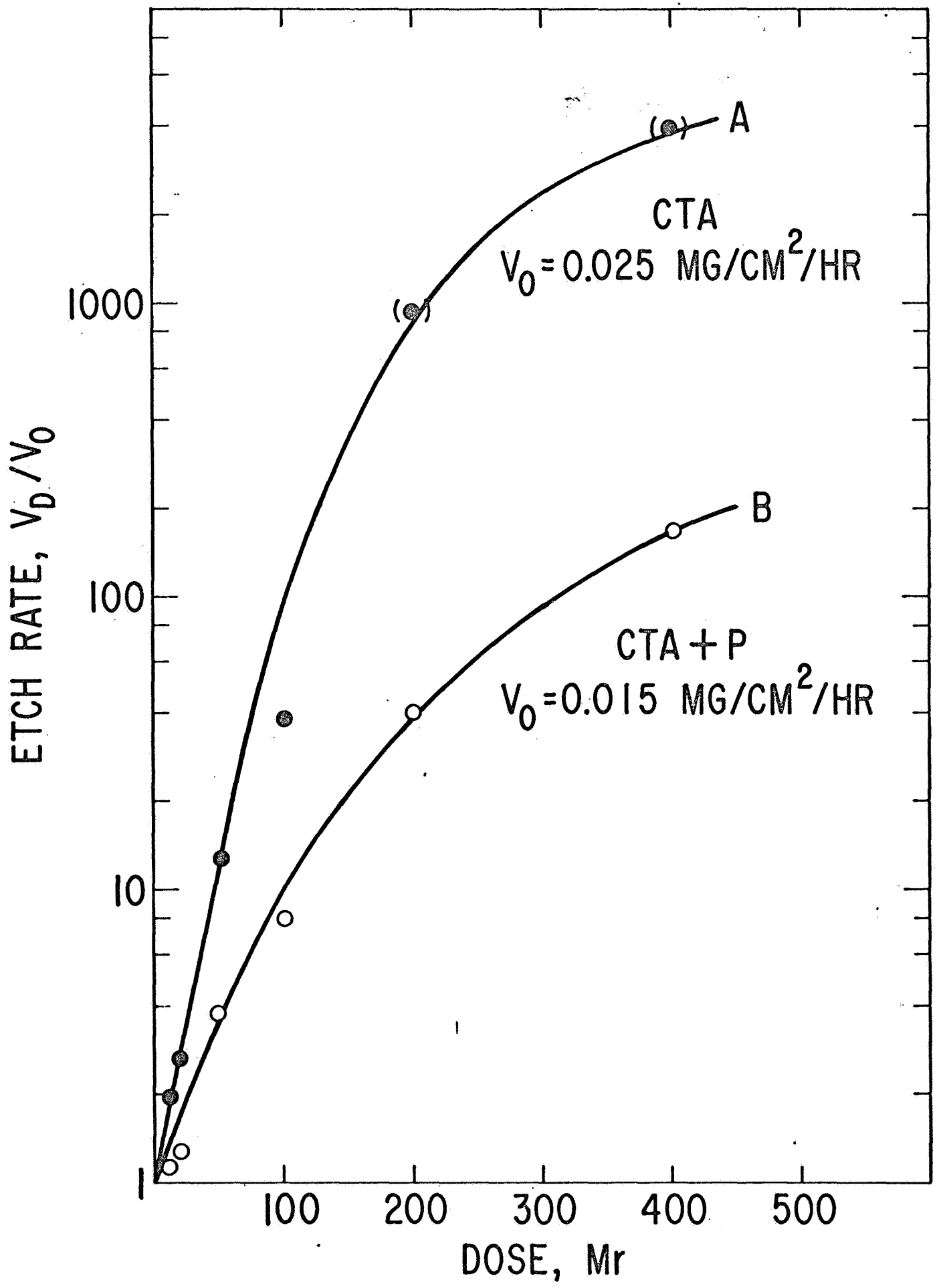


FIG. 2 ETCH RATES FOR CTA (KODACEL®)



PATENT DISCLOSURE LETTER ON RADIATION-CROSSLINKING OF POLYCARBONATES

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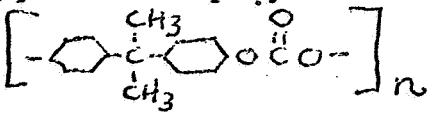
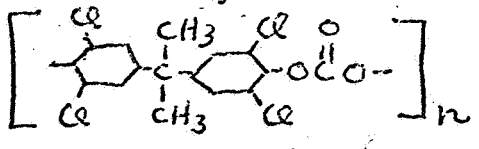
SUBJECT: PATENT DISCLOSURE LETTER ON RADIATION-CROSSLINKING OF POLYCARBONATES

This discovery was made during work under NASA-Houston Contract No. NAS9-8868, negotiated by the R&D Center.

It is known that "bisphenol A" polycarbonate....



henceforth referred to as PC, undergoes chain scission (i.e., degradation) rather than crosslinking when exposed to ionizing radiation[1]. Similarly, I have shown that the symmetrical tetrachlorobisphenol A polycarbonate ....



henceforth called Cl_4PC, shows no evidence of crosslinking over a wide range of radiation doses.

I have discovered that, unexpectedly, a Cl_4PC-PC copolymer (1:1 mole ratio), mixtures of Cl_4PC and PC, and mixtures of Cl_4PC-PC (copolymer) and PC are all crosslinked by ionizing radiation. The materials used were GE Lexan® polycarbonates, characterized by the following intrinsic viscosities (deciliters per gram): 0.81 for PC, ~0.5 for Cl_4PC, and 0.59 for Cl_4PC-PC. Films (10 mils) of various compositions were made from 10% solutions in methylene chloride, vacuum-dried for three hours at 50°C. Rectangular pieces of measured dimensions were irradiated with 1.5 Mev electrons from the GE Resonant Transformer Unit, at R.T. under a nitrogen atmosphere, and at dose rate of 20 Mr/min. (1 Mr = 10^6r). The samples were placed in methylene

chloride for one hour at R.T. and the area swelling ratio, A_s/A_o, determined (A_s is the swollen area, A_o is the original area). The data are given in the table below, arranged on the basis of the chlorine content (weight %) of the Cl_4PC + PC or Cl_4PC - PC (copolymer) + PC compositions.

AREA SWELLING RATIOS OF IRRADIATED POLYCARBONATES

Composition, Parts by Weight		% Cl	Dose, Mr 20	50	100	200	500
PC + Cl_4PC							
1	0	0	*	*	*	*	*
17	1	2	*	(gel)	9.6	6.8	-
6	1	5	*	(gel)	6.2	4.8	-
1	1	18	(gel)	5.6	4.0	3.3	-
0	1	36	*	*	*	*	*
PC + Cl_4PC-PC							
10	1	2	*	9.6	9.3	7.6	-
1	1	11	*	6.3	5.0	3.7	-
0	1	22	*	7.5	5.0	3.0	-

NOTE: * indicates complete solubility; no gel.

(gel) indicates highly swollen, formless gel whose dimensions could not be measured.

The data above show that PC or Cl_4PC alone gave no crosslinked gel even at a dose of 500 Mr. Where gel was observed, the area swelling ratio decreased with increasing dose (i.e., increasing degree of crosslinking), the normal behavior for crosslinked systems. Broadly, the degree of crosslinking for a given irradiation dose increases with the chlorine content in both the Cl_4PC + PC and the Cl_4PC - PC + PC combinations.

The physical properties (i.e., impact and tensile strengths, fracture energy, craze resistance, etc.) of these crosslinked systems remain to be examined. Also, I plan to do analogous experiments with tetrabromobisphenol polycarbonate and Br_4PC - PC copolymer.

This work was done in the period April 29 to May 19, 1969 and is recorded in Notebook No. 8741, pgs. 121 to 129.

Reference (1) J.H. Golden and E.A. Hazell, J. Polymer Sci. Part A 1 1671 (1963).

INVENTOR: A.A. Miller

A.A. Miller

DATE: June 4, 1969

WITNESS: C. Wm Wright

DATE: June 4, 1969

WITNESS: James Stevenson Humphrey Jr.

DATE: 4 June 1969

AAM:DTP

cc: JT Cohen
MD Fiske
AR Gilbert
SW Kantor
AN Wright
PB Price

II. PARTICLE IDENTIFICATION BY DIELECTRIC TRACK DETECTORS

II. PARTICLE IDENTIFICATION BY DIELECTRIC TRACK DETECTORS*

P. B. Price and R. L. Fleischer

INTRODUCTION

Five years ago I believe none of us was optimistic enough to expect that dielectric track detectors would soon be able to resolve individual elements or isotopes of energetic particles. Today I am pleased to be acting as rapporteur of a session devoted to this very topic. Of course, remarkable advances have also been made during these five years in particle identification by electronic methods using semiconductor detectors. We must therefore be careful to ask ourselves whether dielectric track identifiers have any unique advantages over electronic particle identifiers. At the moment I can think of four situations in which dielectric track identifiers are uniquely useful. Perhaps other ways will be brought up during this session.

1. Identification of energetic particles recorded during the ancient past.

2. Identification of heavy particles in a high background of less heavily ionizing particles.

3. Identification of rare particles emitted at extremely low rates such as 1 per cm² per year.

4. Identification of particles of very high ionization rate for which resolution by dielectric detectors exceeds that by other detectors.

THE CONCEPT OF A CRITICAL RADIATION DAMAGE DENSITY

Particle identification by track etching must of necessity be based on the way in which chemical reactivity along the trajectory depends on radiation damage density. Careful work over several years has shown that there exists a well-defined threshold damage density above which the etching rate along a track, V_T, exceeds the general rate of attack, V_G, and below which no preferential etching occurs.(1) Different authors have used different models of track formation to derive expressions for damage density (Refs. 2-4). Each of these models contains an adjustable parameter which is related to the radial extent of the intensely damaged "core" of the track and which differs from solid to solid. We have found that the following expression gives an adequate empirical representation of damage density:

$$J = (aZ_{eff}/\beta^2)\{\ln(\beta^2\gamma^2) + K - \beta^2\}. \qquad (1)$$

Figure 1 displays curves of J vs β for nuclei of effective charge $Z_{eff} = Z[1-\exp(-125\beta/Z^{2/3})]$.

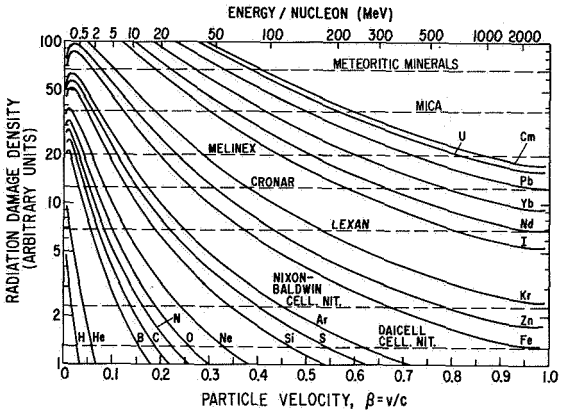


Fig. 1 Damage densities in a typical dielectric solid as a function of velocity for various bombarding nuclei, calculated from Eq. (1). Measured thresholds for track recording by several solids are indicated by horizontal lines.

The equation is of the same form as Bethe's primary ionization equation(5) and was first applied to track formation in crystalline solids, to which the ion explosion spike model seems to apply.(2) We have found that this same equation is useful for plastics and other noncrystalline solids, provided K is chosen for each solid to make the response V_T depend only on J. Thus, although for convenience threshold values J_c for several solids have been indicated on the same graph in Fig. 1, in practice the exact shapes of the curves will differ somewhat for each solid studied. In order that J_c not depend on the angle of entry, we define J_c for a particle at normal incidence and correct each measurement of observed track length by adding to it the amount $V_G t \csc\delta$ that was etched away during the time t for a dip angle δ.

TOTAL ETCHABLE TRACK LENGTH AS A MEASURE OF Z

The first method proposed for particle identification assumed that after a long etching time t_{max}, the entire portion of the trajectory along which $J \geq J_c$, would be etched out. On this assumption, using appropriate curves of radiation damage density, a relationship between Z and t_{max} could be calculated. In their application of this method to fossil cosmic ray tracks in meteorites, Fleischer et al.,(6) Maurette (Ref. 7), and Price et al.(8) measured only that portion of t_{max} in one of the two fragments of a cleaved crystal, so that at best a distribution of delta functions in length would have been smeared into a set of superimposed, overlapping step functions. As crude as it was, this method yielded the first definitive observations of nuclei heavier than iron in the cosmic

*Talk given by P. B. Price based on joint work with R. L. Fleischer, May 1969 @ Conf. on Nuclear Track Registration ar Clermont-Ferrand, France.

radiation. Each of the papers reported similar values of the abundance of nuclei with Z > 30 relative to the Fe-group, namely ~1/3000.

Since then, Lal et al.[9] and Maurette et al.[10] have tried more elaborate schemes for observing the entire etchable lengths, ℓ_{max}, which show considerable promise. The "track in track" method of Lal et al. is illustrated in Fig. 2. See also the review paper by Walker in this volume.

Another approach utilizes the fact that the resistance to "track-fading" increases with damage density. Several years ago Perelygin and his co-workers[23] found that by a suitable annealing treatment they could greatly reduce the lengths of tracks of 10MeV/nucleon argon ions in mica without appreciably affecting the appearance of tracks of the more heavily damaging fission fragments. In this session Maurette discusses differential annealing experiments in which the charge distribution of tracks in crystals could be inferred from the temperature at which they anneal out.

In my opinion rapid progress in identification of particles with $Z \gtrsim 24$ in meteorites must await the development of beams of heavy ions such as Fe, Co, Ni, Zn, and Kr in accelerators. Only then can we determine experimentally the relation between ℓ_{max} and Z as well as the degree of dispersion of values of ℓ_{max} for ions of the same Z. Two factors can contribute to a dispersion of ℓ_{max} values: (1) the threshold is certainly not mathematically sharp; and (2) the etching rate V_T depends on $J-J_c$ and decreases to V_G (the general rate of attack) as J decreases to J_c.

Benton and Henke[11] have made measurements of ℓ_{max} for low-energy cosmic rays in cellulose nitrate stacks, but the second factor just mentioned made it difficult for them to achieve resolution of individual elements.

ETCHING RATE AS A MEASURE OF Z AND A

The most promising method of charge resolution, originally developed for cosmic ray identification in plastic detectors,[12, 13] exploits the fact that V_T is a function of $J-J_c$. For Lexan® this function is accurately exponential,[13] which makes V_T extremely sensitive to small variations in ionization rate for highly ionizing particles. For some other plastics V_T is a weaker function.[12] Instead of trying to etch out the entire recordable range (i.e., where $J > J_c$), one etches under carefully controlled conditions for a fixed time and measures the cone lengths at each exposed surface crossed by the particle, as indicated in Fig. 3. the analogy to electronic particle identifiers is obvious: The cone lengths $L = \int V_T \, dt$ provide a set of measurements of radiation damage density at measurable distances from the end of the range of the particle, just as voltage pulses in thin silicon crystals provide measurements of increments of energy deposited within known thicknesses before the particle loses all its energy in a thick crystal. Figure 4 shows some of the original measurements made in cellulose nitrate with this controlled etch rate method.[15]

Fig. 2 Short tracks intersecting rare, very long tracks in a crystal with a high track density from the Patwar meteorite.[9] The crystal was etched for a very long time and then ground so that the badly attacked top few microns of surface were removed. The occasional very long tracks of cosmic rays with $Z \gtrsim 30$ intersected many short tracks of cosmic rays with $Z \sim 24$ to 30. The long tracks provided access for the etchant to reveal the total recordable range of the slowing cosmic rays with $Z \approx 24$ to 30.

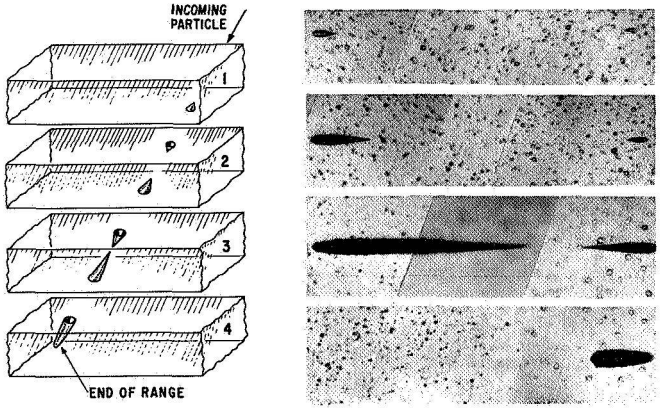


Fig. 3 Etched portions of the track of a Ca cosmic ray slowing down in a stack of Lexan sheets. The rate of change of etched cone length with distance is a unique function of atomic number and mass below threshold at the top of the sheet, but increases above threshold within the sheet so that a cone of short length is etched at the bottom of the sheet. In sheets 2 and 3 the lengths of the etched cones increase as the velocity of the particle decreases until finally it stops in sheet 4.

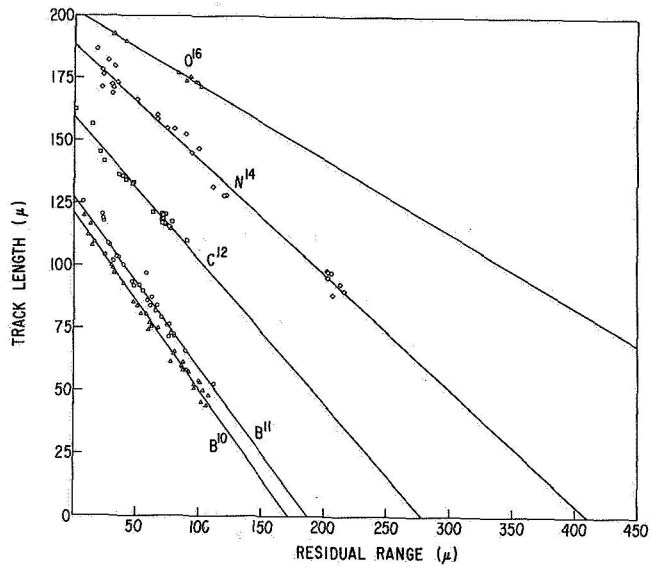


Fig. 4 Measurements of cone length as a function of residual range measured from the low-energy end of the cone for ions in two-layer stacks of 250μ thick cellulose nitrate. The ions were produced in the Yale accelerator and degraded by various amounts from 10 MeV/nucleon.[15]

In general, the longer the range of the particle, the more measurements of its local radiation damage rate can be made and the more accurately can its identity be established. By using Lexan detectors, Price et al.[13] were able to resolve atomic numbers of cosmic rays from Z≈12 to 28 and to provide the first data on their relative abundances at energies below a few hundred MeV/nucleon, some of which are shown in Fig. 5. Additional work on this subject is reported in this session by Enge and Bartholoma, by O'Ceallaigh et al., and by Blanford et al. Also in this session, Magdalena and Monnin report studies of tracks of heavy ions from an accelerator in cellulose nitrate and PVC. Figure 6 shows some measurements made in our laboratory on Ne, O, and F ions using cellulose triacetate detectors.

One of the most exciting applications of this high resolution technique is to the identification of super heavy cosmic rays, which may extend well beyond uranium in mass. Figure 7 shows the present status of comparative measurements on the same relativistic, superheavy cosmic rays that passed through a stack of interleaved plastics and emulsions.[14] The emulsion measurements were made by P. H. Fowler and co-workers. To assign atomic numbers to the events in the plastics, a relation between V_T and Z was

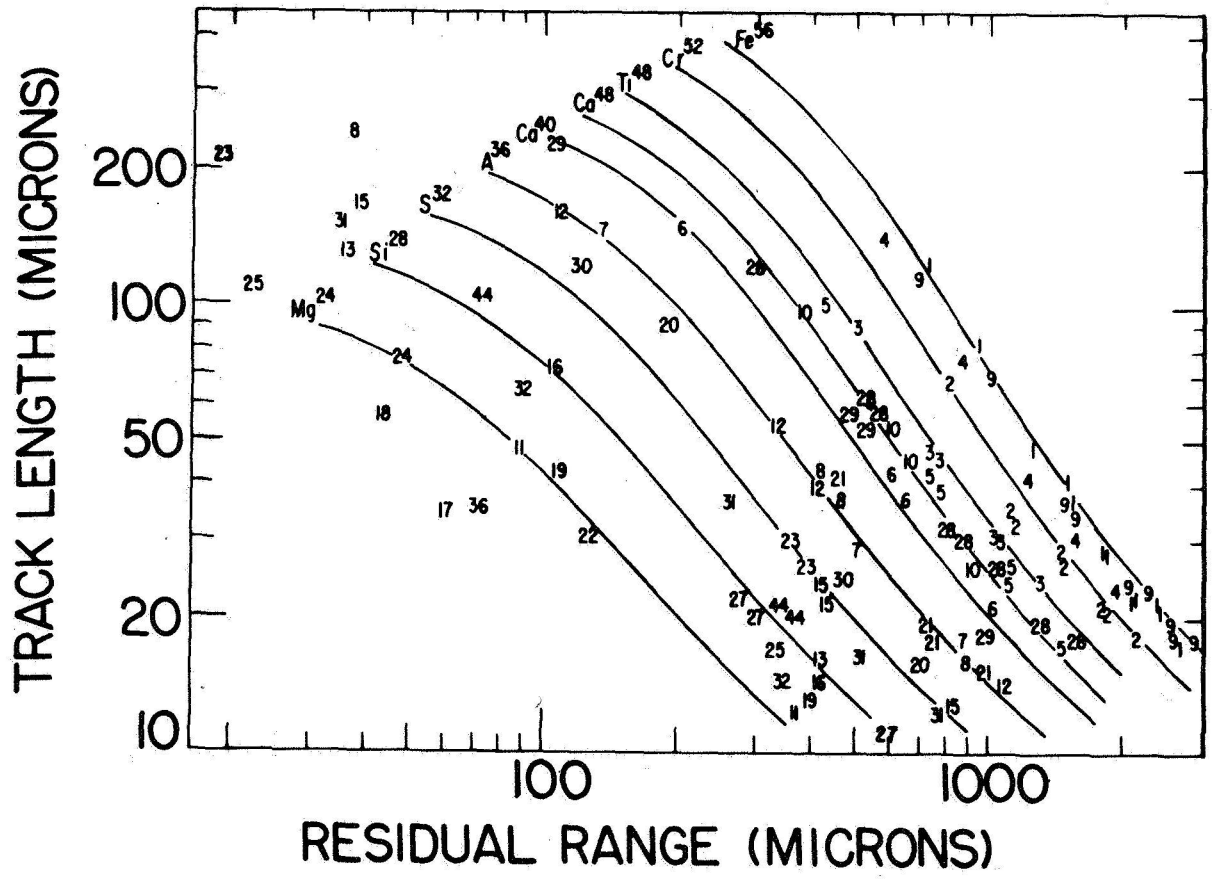


Fig. 5 Cone length vs residual range for a representative sample of cosmic rays that came to rest in a Lexan stack during a balloon flight over Ft. Churchill in 1967.[13] Numerals are used both as data points and to identify events. The curves were calculated from an exponential response.

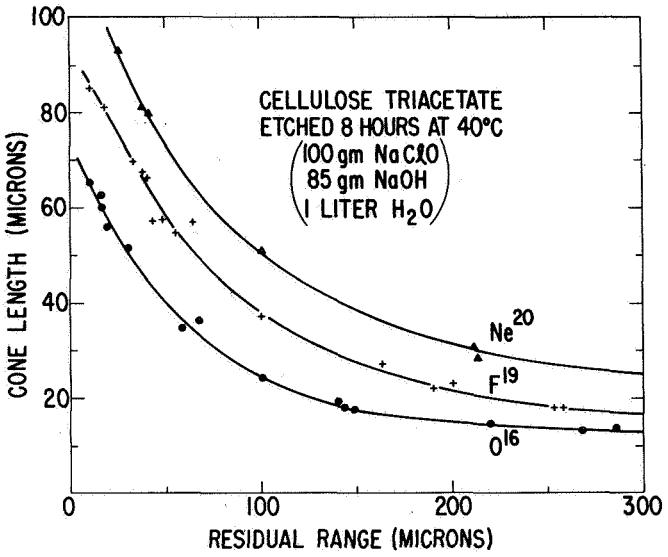


Fig. 6 Unpublished data by the authors on cone length vs residual range for Yale Hilac ions in Eastman Kodacel cellulose triacetate (without plasticizer).

established by making measurements of cone length vs residual range in similar sheets of the same plastic etched in an identical way after having been exposed to ions in the low-energy cosmic radiation and to known ions in accelerators. Equation (1) was used and the velocity of the relativistic superheavies was taken to be 0.95c. Stringent lower limits on this velocity were set by requiring that there be no observable increase in V_T from top to bottom of a 1 g/cm^2 stack. Probably improvements in charge assignment by both the emulsion technique and the plastic technique will bring the data into better agreement.

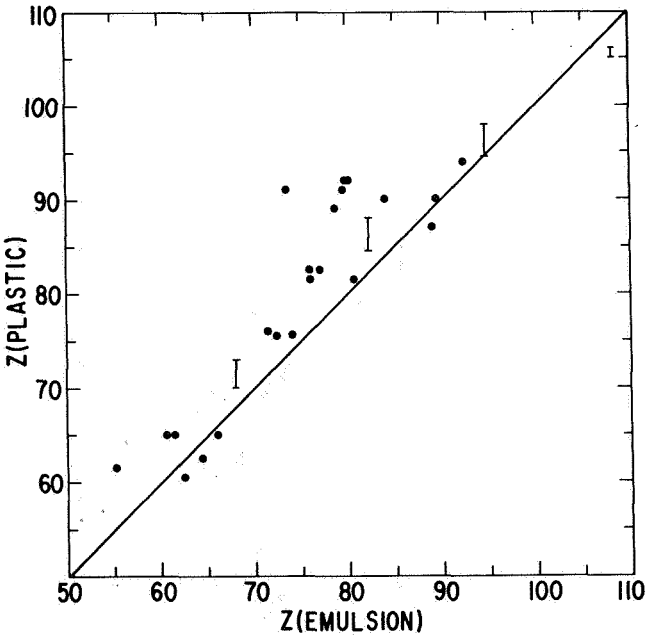


Fig. 7 Preliminary results on charge assignment to relativistic, superheavy cosmic rays by measurements made on the same events in interleaved stacks of plastics and nuclear emulsions. The stacks, with a total area of 32m^2, were exposed for 40 hours over Palestine, Texas, in September 1968.

IDENTIFICATION OF SHORT-RANGE PARTICLES

Lightly ionizing, short-range particles, which do not traverse a single sheet of detector, can be identified in plastics by an etch + re-etch scheme, illustrated in Fig. 8. With this method, G.E. Nichols, R.L. Fleischer, and I have identified He, Li, and Be fragments emitted during the ternary fission of Cf^{252} including observations of the breakup of Be^8 into two alpha particles following the decay of Li^8 fragments (Fig. 9). At Brookhaven, S. Katcoff and L. Hussain are calibrating Lexan detectors for the identification of light particles emitted in high-energy reactions, using the etch + re-etch scheme.

Heavily ionizing short-range particles, such as fission fragments, can in principle be identified by measuring etch rates in glass or a crystalline solid. Fleischer, Woods, and I have recently found[16] that V_T is an increasing function of J in various kinds of glasses, as shown in Fig. 10. As a consequence,

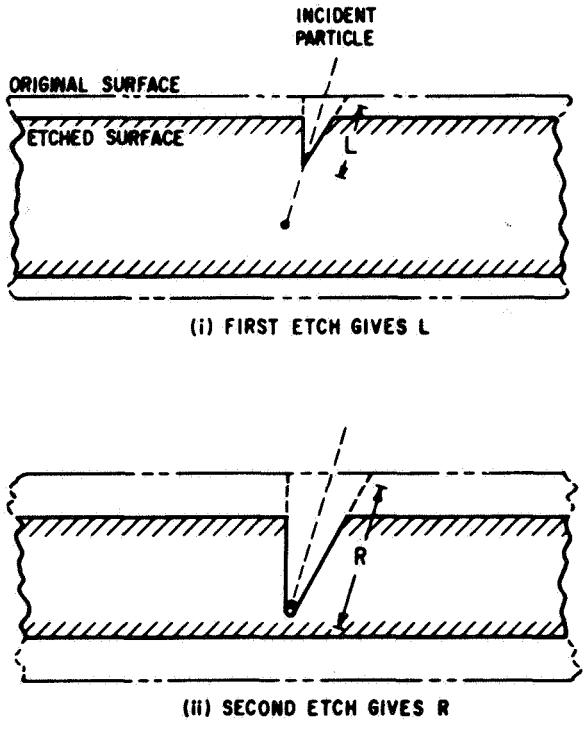
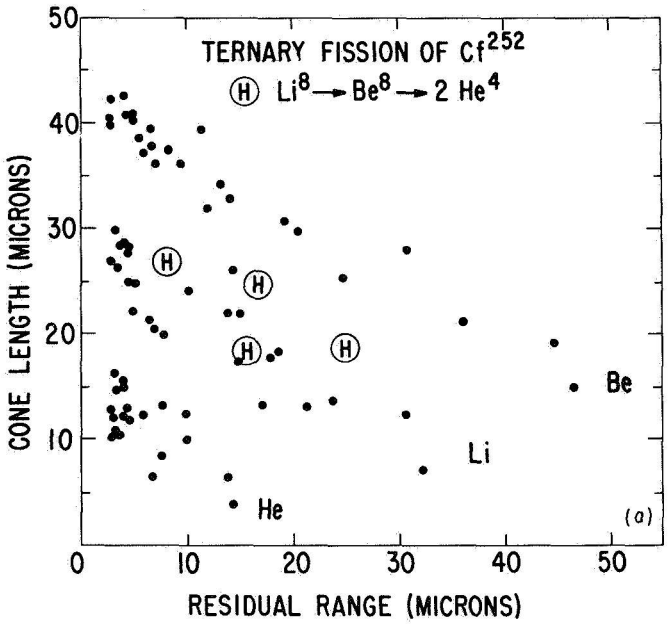


Fig. 8 Method of identifying particles that do not penetrate a sheet. Cone length, L, is measured after a first etch. After a long second etch the cone has extended to the end of the range of the particle, which allows the residual range from the end of the original cone to be deduced by subtracting the old from the new length.



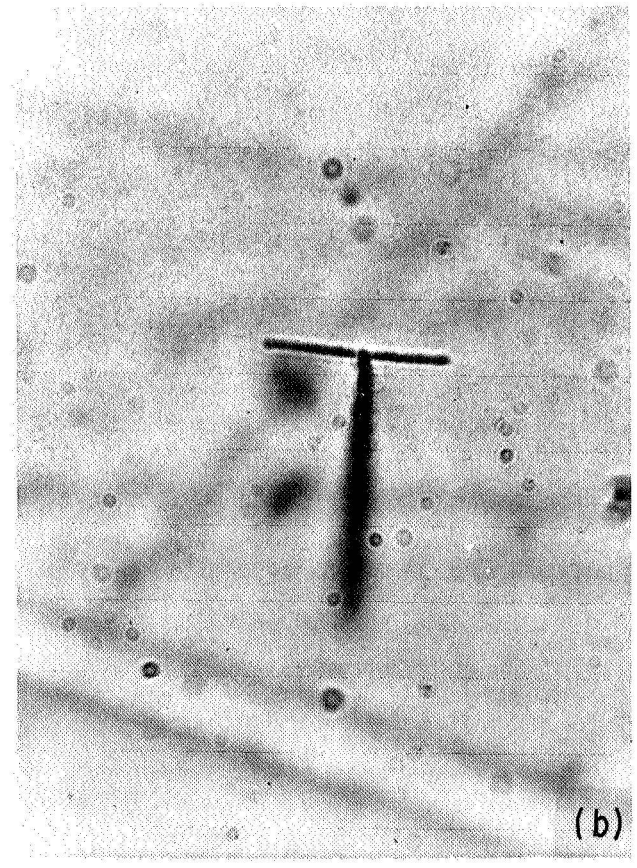


Fig. 9 Distribution of cone lengths vs residual range for light fragments emitted from Cf^{252}, after penetrating a 50μ absorbing foil (used to stop all normal binary fission fragments and 6.1 MeV alphas). The photomicrograph shows a track of a Li^8 fragment which has decayed into Be^8, which then breaks up into two alpha particles.

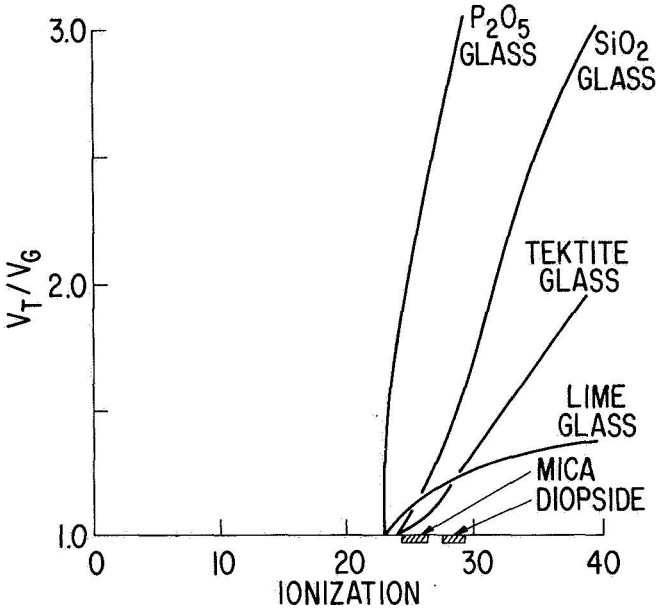


Fig. 10 Etching rate as a function of ionization rate for a series of inorganic glasses. V_T/V_G is computed from the measured values of $\theta_c \equiv [\arcsin (V_G/V_T)]$, the critical angle for revealing tracks by etching. Ionization values are from the relation inferred by Price, Fleischer, and Moak.[22] Cf^{252} fission fragments of various energies were obtained by using a series of stopping foils of known thickness. Thresholds for mica and diopside are shown for comparison.

after a long etching time the diameter of the pit surrounding a fission fragment track is an increasing function of the initial energy of the fission fragment. This has been beautifully demonstrated by Fiedler and Hoppner in a paper in this session. Of course, if one is trying to distingusish fission fragments from heavy ions or recoiling compound nuclei, more than one measurement along the same event is required to gain separate information about identity and range. Further work aimed at identification of very heavy, short-range particles is in progress.

WHAT TO EXPECT IN RESOLUTION IN THE FUTURE

We can already identify atomic numbers of ions up to at least Z=30 that come to rest in a stack of plastics. I believe that with further work we will be able to resolve isotopes of particles which have a range of at least a few hundred microns and which differ in mass by at least 4% or 5%; that we will be able to resolve charges of all relativistic nuclei with Z>30; and that we will be able to measure atomic numbers of fission fragments to within one or two units. Miniaturized, multilayered plastic detectors with automatic readout for fission fragment identification are already in the planning stage.

To achieve the highest resolution the detectors must be quite uniform in composition and they must

be irradiated and later processed under very carefully controlled conditions. We have found that Lexan polycarbonate sheets manufactured by General Electric are of adequate uniformity and have an unlimited shelf life. We have found that Eastman Kodacel (cellulose triacetate) made on special order without any plasticizer is of adequate uniformity and has not deteriorated in one year. No commercially available cellulose nitrates are sufficiently uniform in composition or of adequate long-time stability. Benton[17] has made films of cellulose nitrate which are superior to most commercial films, but by its very nature cellulose nitrate deteriorates slowly in air at room temperature and for this reason may have limited use.

The chemical reactivity of the damaged region along the particle trajectory should depend only on radiation damage rate and not on external factors. Some precautions that should be taken in achieving this goal are summarized below. They are discussed in detail in Peterson's PhD thesis.[18]

1. Starting material. To avoid variations in composition, it is best to use sheets manufactured in the same batch.

2. Irradiation environment. The final reactivity along tracks in some plastics depends on the ambient temperature and composition of the atmosphere around the plastic during irradiation. Calibrations in accelerators should be done in an environment as similar as possible to that existing during exposure of the detector to the particles to be studied.

3. Steady-state reactivity. After irradiation the reactivity increases at first rapidly and then at a diminishing rate, reaching a steady state after a time that depends on the plastic and on ambient temperature. Figure 11 illustrates this effect in Lexan exposed to Ne ions. We allow Lexan detectors to stand in the dark for about one month at room temperature before processing them. After one month the rate of change of reactivity is negligibly small. The detectors can then be kept indefinitely in a freezer at -30°C.

4. Annealing of damage. This effect has been thoroughly studied by Peterson,[18] because it can seriously limit resolution. Probably few workers realize that if they etch at temperatures as low as 50°C they are likely to lose resolution! The problem is that the fractional decrease in reactivity caused by elevated temperatures depends on radiation damage rate. Reactivity along tracks of particles close to threshold decreases more rapidly with annealing than does reactivity along tracks of heavily ionizing particles. For example, tracks of Ne ions in Lexan are significantly affected by a 24-hour anneal at 50°C, whereas tracks of cosmic rays with $Z \gtrsim 16$ are not. Some annealing data for cosmic ray tracks in Lexan are shown in Fig. 12. Since the time at which our original paper was written on cosmic ray identification with Lexan detectors, we have chosen to etch at temperatures below 50°C. Even though the etching time required to produced a given size track is much longer

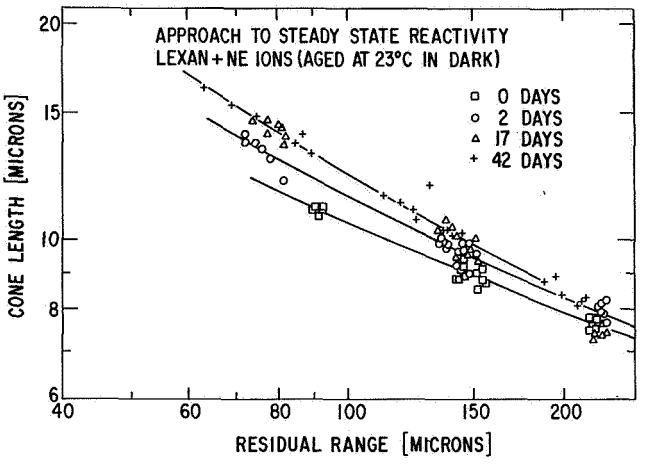


Fig. 11 Change of reactivity along Ne ion tracks in Lexan with time of aging at 23°C in air in the dark (Ref. 18).

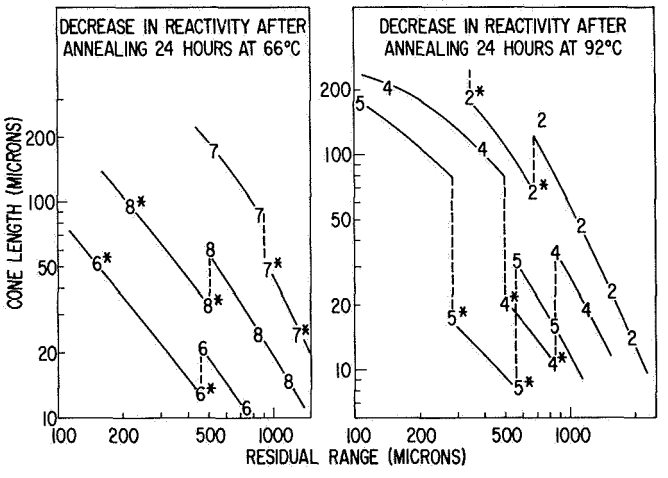


Fig. 12 Effects of annealing at various temperatures on reactivity along tracks of heavy cosmic rays in Lexan.[18] Each number designates a particular cosmic ray event. An asterisk means that the sheet was annealed before etching. All sheets were etched for 168 hours at 30°C.

at 50°C than at 70°C, the extent of track fading is diminished because the activation energy for annealing is higher than that for etching.

5. Ultraviolet exposure. In 1968, Crawford et al. (Ref. 19) made the very exciting discovery that the reactivity along tracks can be drastically increased by exposing the plastic to ultraviolet radiation in the presence of air. An example of the magnitude of the effect is given in Fig. 13. Other forms of ionizing radiation, such as x-ray and energetic electrons, also increase the reactivity along the tracks but at a lower rate. To avoid changing their reactivity it is essential to exclude ultraviolet light from plastic detectors after they have been exposed to charged

particles. However, provided the exposure could be made uniform throughout the detector thickness, a controlled dose of ultraviolet radiation could, of course, be used to increase the detector signal by a desired amount as Fig. 13 shows.

6. Etching conditions. The etching rates V_T and V_G increase exponentially with etch temperature and somewhat faster than linearly with the concentration of the solution. Traces of certain additives such as Benax surfactant, which is very useful in improving the etching characteristics, may significantly change V_G and V_T. Rigid control of temperature, concentration, and purity of solutions is essential.

7. Response function. For the same plastic, the relationship between V_T and J, as well as the value of J_c, depend very much on composition of the etchant. For Lexan an aqueous hydroxide solution gives an exponential response; if the solution is diluted with equal parts of ethanol the response becomes nearly linear and the threshold J_c is considerably lowered. Somogyi et al.[21] have found that J_c for cellulose acetate is lower when $KMnO_4$ is added to hydroxide solution than when hydroxide solution alone is used.

8. Anisotropy of response. Under certain conditions V_T may not be independent of dip angle. In this session, O'Ceallaigh et al. report that in Lexan detectors etched at 23°C in a solution of 25 grams NaOH +100 ml water + 100 ml ethanol the dependence of V_T on dip angle is quite noticeable. To get good resolution they have separately analyzed cosmic rays within small intervals of dip angle. In aqueous solutions no dip angle dependence is observed.

CONCLUSIONS

What is the ideal detector? There is no one ideal detector that suits all needs. Cellulose nitrate still appears to have the highest sensitivity. It can detect low-energy, singly charged particles.[20, 21] With it we have resolved isotopes of B in accelerator bombardments[12] and with effort we might be able to resolve isotopes of Li and Be at reasonable flux levels in a small area. Unfortunately it is not uniform enough for high resolution of isotopes of cosmic rays at very low flux levels, where a very large area must be used. Cellulose triacetate without plasticizer shows promise that it may be adequate for resolving isotopes of some elements in the cosmic radiation. We are now using this plastic to study relativistic cosmic rays with $Z \approx 40$ to 70.

With its exponential response (when etched in aqueous hydroxide solution) Lexan now has the highest resolution of any plastic yet studied, for highly ionizing particles. We are making extensive use of large areas of Lexan in current studies of relativistic cosmic rays with $Z \gtrsim 60$ and of medium energy cosmic rays with $Z \gtrsim 10$.

Presumably another conference on dielectric track detectors will be held about two years from now.

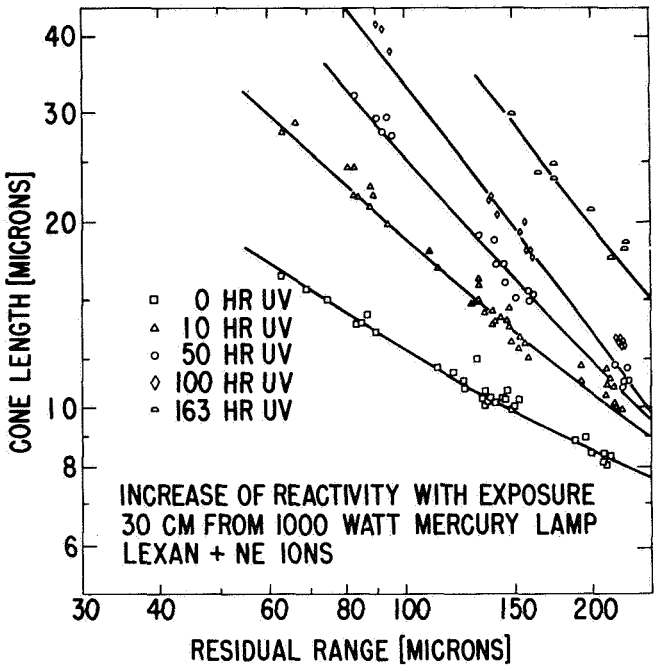


Fig. 13 Increase of reactivity along Ne tracks in Lexan resulting from exposure to a 1000-watt mercury vapor lamp after bombardment and before etching.[18] The theoretical curves for an exponential response are discussed by Peterson.[18]

I predict that during the intervening period the techniques of particle identification just discussed will have been applied with great success in each of the four ways outlined at the beginning of this report.

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III. COMPOSITION OF COSMIC RAYS OF ATOMIC NUMBER 12 TO 30

III. COMPOSITION OF COSMIC RAYS OF ATOMIC NUMBER 12 TO 30+

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Summary

Improvements in the chemical processing of Lexan polycarbonate track detectors have made it worthwhile to collect new data on the elemental abundances of cosmic rays with $Z \geq 12$ and with energies below 250 MeV/nucleon. Lexan stacks were exposed in July, 1968 at a residual pressure of 1.8 mbar over Ft. Churchill. The new abundance distribution is consistent with the one reported in Phys. Rev. Letters 21, 630 (1968), but the resolution is higher ($\Delta Z \approx .3$ charge unit, $\Delta A \approx 2$ amu at the 1σ level) and the even-Z to odd-Z ratio is larger (~4.5). The high abundance of Mn at low energy is confirmed (Mn/Fe $\approx .5$). These measurements in the region $Z \leq 30$ and $0.1 < \beta < 0.45$ are used to predict curves of response of Lexan to cosmic rays with $Z > 30$ and β up to 0.95.

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*Speaker

+Supported in part by NASA under contract NAS 9-8868.

1. Introduction

In a series of papers[1-3] we have shown that heavily ionizing particles can be identified by measuring the conical pits that are etched along their trajectories in certain plastics that have been treated in a caustic solution of controlled composition and temperature. The rate of growth of the etch pits is an increasing function of ionization rate. Following a fixed etching time, the lengths of the etch pits are measured at various residual ranges along the trajectory of a particle that has passed through one or more plastic sheets in a stack. The variation of pit length with residual range is a unique function of atomic number, Z, and mass A.

Using Lexan polycarbonate sheets flown from Ft. Churchill at a residual pressure of 2.5 mbar, we were able last year to obtain the first data[3] on the charge composition of stopping cosmic rays with $Z \gtrsim 14$. We reported a high Mn abundance, comparable to that of Cr. Since then several methods have been used to study the same charge region, with conflicting results.[4-7] Because the cross sections for production of Cr and Mn from Fe are strongly energy-dependent, their abundances relative to Fe should be sensitive to their propagation history. We felt it was important to make further abundance measurements with Lexan, using improved etching procedures.

In addition to their intrinsic value, these measurements are an essential step in establishing the response of Lexan to cosmic rays with both high Z and high β.

2. Experimental Details

In another paper[8] we have discussed factors that may impair the resolution of plastic detectors. Most serious of these is the thermal fading of tracks during etching in a hot solution. In the present work we have tested two new etching techniques for Lexan that avoid track fading: (1) an "alcoholic etch" consisting of 25 gm NaOH + 100 ml water + 100 ml ethanol for 30 hours at 23°C, and (2) an "aqueous etch" consisting of 6.25 N NaOH solution + 0.05% Benax surfactant for 160 hours at 40°C, the Lexan having been preheated in air 2 hours at 50°C to drive off dissolved gases.

In each solution we processed Lexan stacks exposed in July, 1968, at 1.8 mbar over Ft. Churchill, as well as a stack bombarded with O^{16}, F^{19} and Ne^{20} ions at the Yale heavy ion accelerator. Unfortunately we found that with the alcoholic etch V_T, the rate of increase of etch pit length, depends not only on J, the ionization rate, but also on the dip angle of the track. By analyzing separately data for tracks at 10° intervals of dip angle, we deduced the response V_T versus J shown in Fig. 1. The data on extremely heavy cosmic rays in Lexan detectors

reported elsewhere in these proceedings[9] were obtained with the alcoholic etch, using the appropriate response curves in Fig. 1.

Because there turns out to be no dependence of response on dip angle for samples treated with the aqueous etch, we favor it over the alcoholic etch. The remainder of this paper concerns results obtained with it. Fig. 1 gives the response V_T versus J inferred from the raw data displayed in Fig. 2. Although for clarity we have left the identifying numbers off the points in Fig. 2, we emphasize that each event comprises several points (up to 20 for the heavy nuclei) that fall along the curves. As an example, note that one nucleus with $Z = 30$ was detected; the six measurements of etch pit length at various residual ranges for that nucleus fall beautifully along the calculated curve for Zn^{70}.

Curves of L versus R for all stable isotopes of the elements $8 < Z < 32$ have been calculated as described previously[3] and have been used to identify the cosmic rays. With the new aqueous etch V_T is a complicated function of J. It was convenient to use the polynomial function $V_T^{-1} = \sum_0^9 a_i J^i$ with coefficients chosen to make the curve marked Fe^{56} pass through the densest population of points in Fig. 2. The spacing between the curves is weakly dependent on the form of the ionization equation. The equation of Katz and Kobetich[10] based on energy deposited by delta rays was tried and rejected because the curves were too close together. The best

spacing of the curves was achieved with the empirical ionization equation

$$J = (Z_{eff}^2/\beta^2)[\ln(\beta^2\gamma^2) + 20 - \beta^2] \quad (1)$$

Within the region $8 \leq Z \leq 30$ and $0.1 < \beta < 0.45$, the calculated curves fit the data for known ions of O^{16}, F^{19} and Ne^{20} and give strong peaks at the abundant elements Mg, Si, Ca and Fe. For $\beta < 0.1$ the curves are unreliable. For $\beta > 0.45$ the etching rate along Fe tracks is too small to be measured. Soon to be flown at high latitude is a large interleaved stack of Lexan, steel absorbers and emulsions, designed to bring to rest a few rare, extremely heavy nuclei ($Z > 35$) with $0.45 < \beta < 0.85$. Until results are available we are using eq. 1 and the polynomial in J to predict values of V_T for $\beta > 0.45$ and $Z > 30$. Fig. 3 shows the predicted response for a few extremely heavy nuclei using the aqueous etch. In Fig. 1 the abscissa is given both in units of J and in units of Z at $\beta = 0.95$, corresponding to an average cosmic ray velocity at the latitude of Palestine, Texas.

3. Results

At the present time 104 events with $Z \geq 12$ have been analyzed. Fig. 4 gives the charge histogram for these events and the elemental abundances after correcting for recording efficiency and energy interval studied. We have cross-hatched those events (mainly light nuclei) for which only one etched cone could be measured. The other events are more reliable because from 2 to 20 etched cones were measured for each of them.

4. Discussion

We believe it should be evident from a comparison of Fig. 4 with the histogram in our previous paper[3] that we have achieved an improvement in resolution, even though the total number of events is small. Expressed in terms of one standard deviation of the individual points about the most probable curve for an event, about half the events above Ca have $\Delta A \approx \pm 2$ amu, one-fourth have $\Delta A \approx \pm 1.7$ amu and one-fourth have $\Delta A \approx \pm 2.3$ amu. In terms of atomic number, the typical standard deviation is $\Delta Z \approx \pm .3$ charge units. Below Ca the resolution tends not to be quite as good, mainly because not as many cones are etched. In contrast to the previous work, we are able now to see Mg quite clearly.

Between Mg and Zn our measured even-Z to odd-Z ratio is ~4.5 compared with 2.2 in our previous work. The peak at Mn (Z=25) is still present and appears rather convincing, although final judgment will be reserved until far more events are measured. One interpretation is that both Mn^{53} and Mn^{55} are produced with high cross sections in spallation of Fe at low energies, whereas the yield of lighter elements is much smaller.

Except for the high Mn abundance, our distribution at ~150 to 250 MeV/nucleon agrees quite well both with the low energy abundance distribution of Munoz and Simpson[7] to be reported at this conference and with the abundance distribution recently obtained by Webber (private communication) at energies above 1 GeV/nucleon and also to be reported at this conference. The agreement with Waddington and Freier[4] is less good, although the trends are similar.

After all the events in our flight (>1000) have been analyzed, we can reject the single-cone events and critically examine the data to see if isotope peaks stand out. It seems not at all out of the realm of possibility. Further improvement in resolution beyond the present stage may require improving the plastic detectors, which are flown as made commercially.

Finally, note that the rate of increase of V_T with J (Fig. 1) is more rapid for the aqueous than the alcoholic NaOH etch, even though its threshold ionization rate is somewhat lower. For this reason, and because of its absence of a dip angle effect, we expect to process all future Lexan stacks with the aqueous etch, including those exposed for long times to super-heavy cosmic rays.

5. Acknowledgments

We are grateful to G. E. Nichols and Mrs. H. Couch for much assistance, to Dr. K. Fukui for the balloon exposure, and to Dr. H. Bakhru for the heavy ion bombardments.

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Figure Captions

1. Response of Lexan detectors as a function of ionization rate and also of atomic number at $\beta = 0.95$. V_T is the rate of lengthening of the conical etch pits extending from the surfaces inward along the particle trajectory; V_G is the bulk etching rate. The broken curves give the response at various dip angles for an alcoholic NaOH etch (see text); the solid curve gives the response, independent of dip angle, for an aqueous NaOH etch.

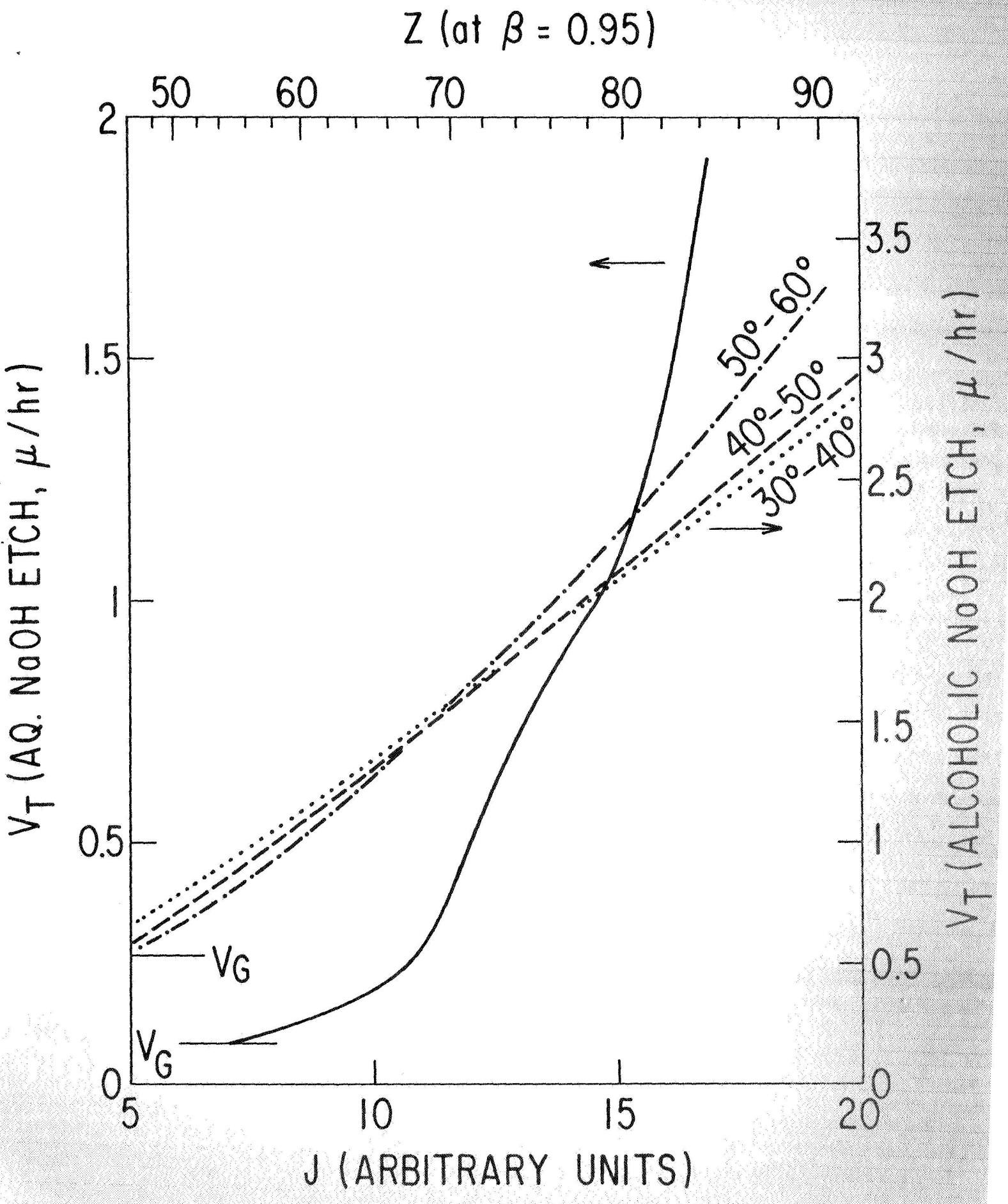
2. Etch pit lengths measured at various residual ranges in Lexan for about 140 cosmic rays with $Z > 10$ (circles) and for a group of ions of O^{16} (triangles), F^{19} (squares) and Ne^{20} (diamonds), produced in the Yale accelerator. The stacks of Lexan were etched in aqueous NaOH solution (see text). Calculated curves for a few nuclides are indicated.

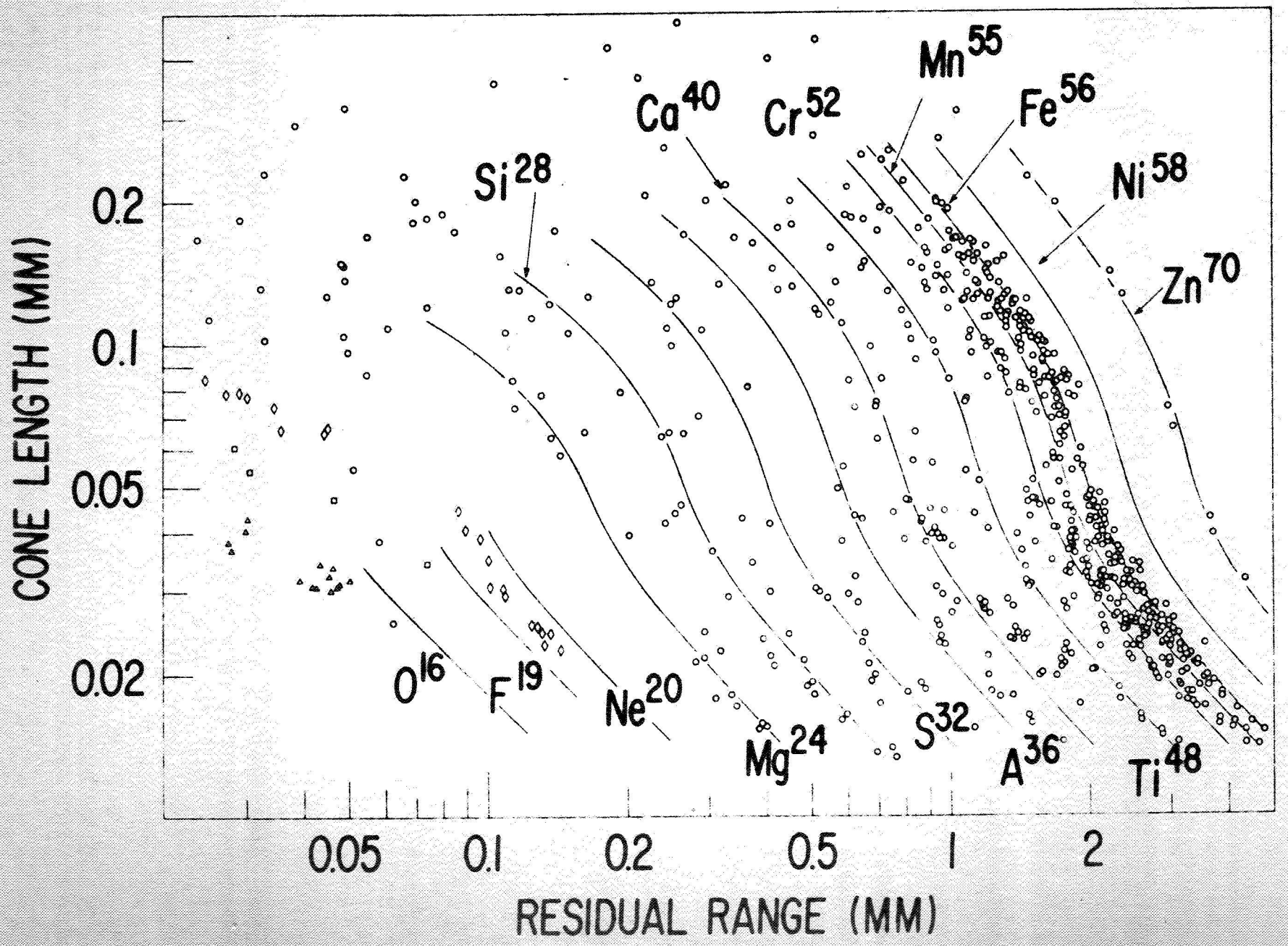
3. Etch pit length as a function of residual range of a few extremely heavy nuclei, calculated from the response curve of Fig. 1 for the aqueous etch, assuming the β-dependence given in equation 1. The points indicate kinetic energies of 300 MeV/nucleon (circles), 500 MeV/nucleon (squares), 800 MeV/nucleon (+) and 2000 MeV/nucleon (X). At the latitude of Palestine, Texas, only events to the right of the dashed curve are allowed.

4. Abundance distribution of our first 104 cosmic rays: (a) Each event was plotted as a rectangle of constant area centered on the most probable isotope and extending one

Figure Captions (Cont'd.)

mean deviation on each side. (b) Relative fluxes were determined by assigning each event to the nearest stable isotope and correcting for recording efficiency and for the same energy interval.


Z (at β = 0.95)
50
60
70
80
90
2
1.5
1
0.5
0
3.5
3
2.5
2
1.5
1
0.5
0
50°-60°
40°-50°
30°-40°
V_G
V_G
5
10
15
20
J (ARBITRARY UNITS)
V_T (AQ. NaOH ETCH, μ/hr)
V_T (ALCOHOLIC NaOH ETCH, μ/hr)


CONE LENGTH (MM)
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0.1
0.05
0.02
0.05
0.1
0.2
0.5
1
2
RESIDUAL RANGE (MM)
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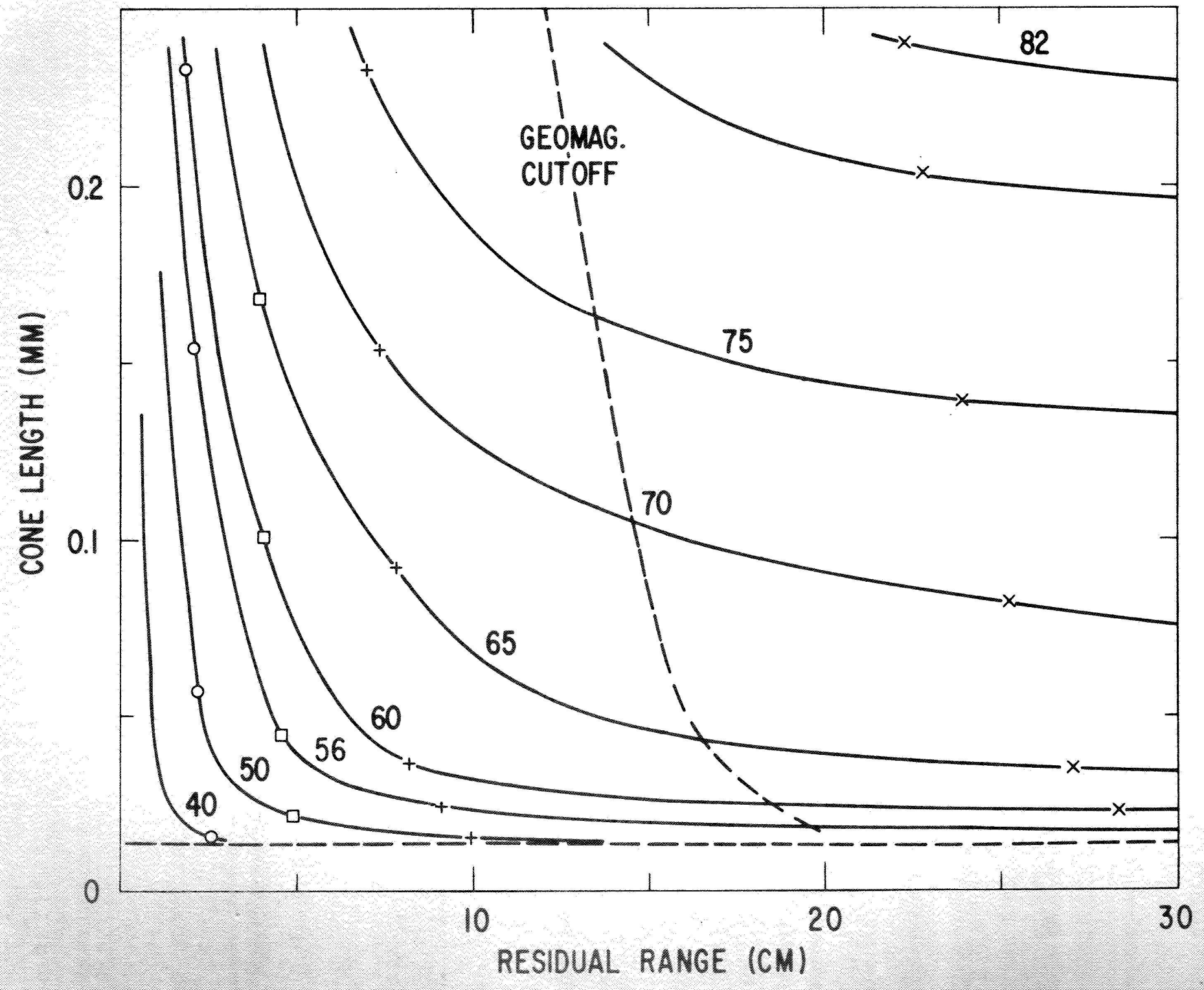
F19
Ne20
Si28
Mg24
Ca40
S32
Cr52
A36
Mn55
Ti48
Fe56
Ni58
Zn70


82
GEOMAG.
CUTOFF
75
70
65
60
56
50
40
0.2
0.1
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CONE LENGTH (MM)
10
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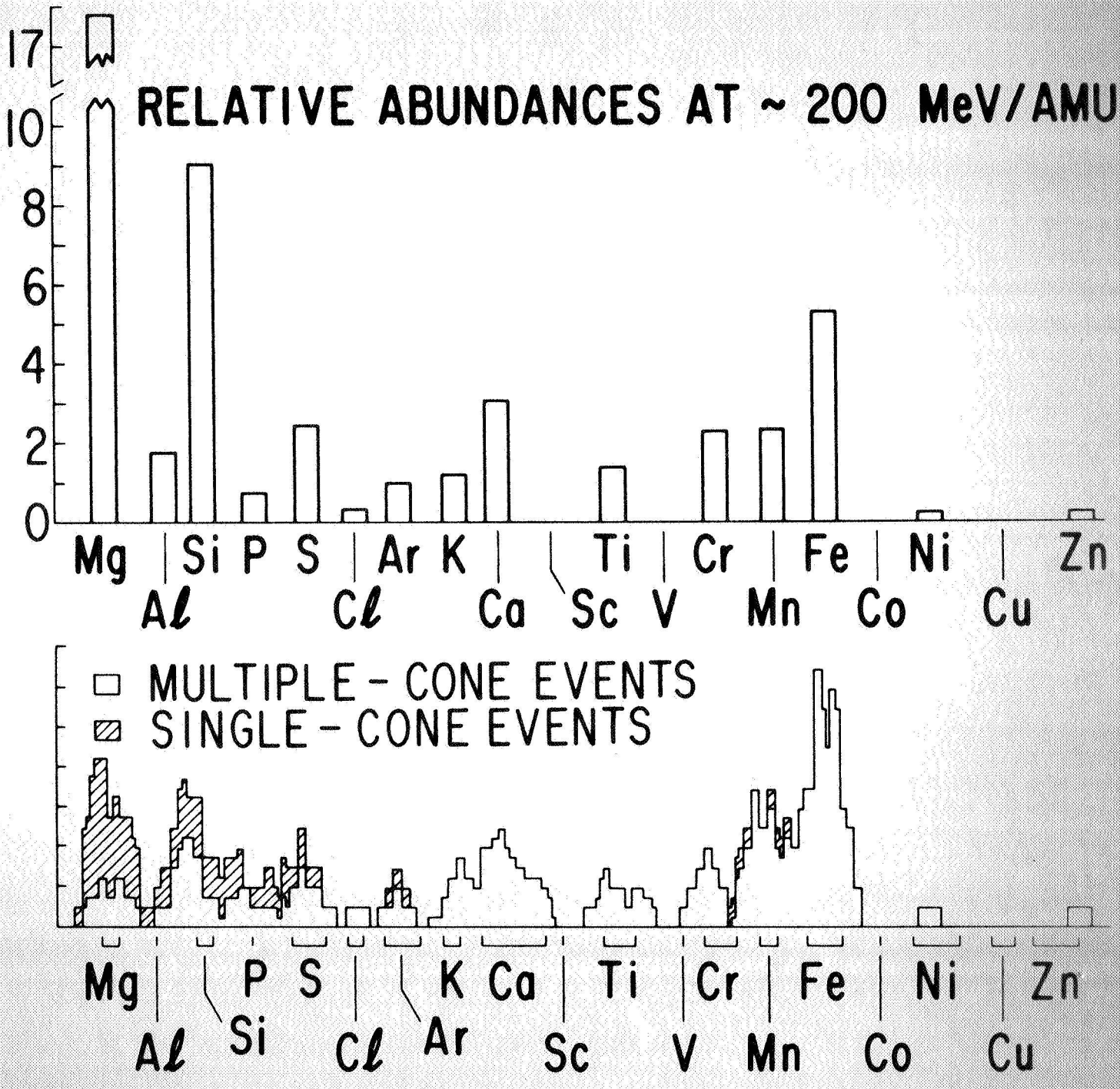
30
RESIDUAL RANGE (CM)


RELATIVE ABUNDANCES AT ~ 200 MeV/AMU
17
10
8
6
4
2
0
Mg
Si
P
S
Ar
K
Ti
Cr
Fe
Ni
Zn
Al
Cl
Ca
Sc
V
Mn
Co
Cu
MULTIPLE - CONE EVENTS
SINGLE - CONE EVENTS
Mg
P
S
K
Ca
Ti
Cr
Fe
Ni
Zn
Al
Si
Cl
Ar
Sc
V
Mn
Co
Cu

IV. IDENTIFICATION OF TRACKS OF SUPER HEAVY COSMIC RAYS IN PLASTICS

IV. Identification of Tracks of Super Heavy Cosmic Rays in Plastics[+]

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Summary

Tracks of 70 cosmic rays with $Z \gtrsim 40$ in stacks of plastics + nuclear emulsions flown over Palestine, Texas, have been identified in three ways: by the rate of lengthening of conical etch pits along tracks in calibrated Lexan polycarbonate and in Eastman cellulose triacetate (CTA) and by the optical density of the same tracks in emulsion. CTA responds to relativistic nuclei with $Z \gtrsim 38$ and Lexan to relativistic nuclei with $Z \gtrsim 54$. The responses of Lexan and CTA to nuclei with $Z > 54$ correlate very well and provide evidence for a Pb peak and for one event with $Z \approx 90$. Charge assignments in the plastics agree better with

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charge assignments based on the central density of the emulsion tracks than with those based on the density at a lateral distance of 10 microns. Up to $Z \approx 60$ the agreement is usually within ~5%, but there is a systematic trend with increasing Z for the charge assignment in emulsion to be higher than the charge assignment in plastic.

1. Introduction

A new world of extremely heavy, extremely rare cosmic rays has been revealed through the use of detectors with very large area-time factors - very old meteorite crystals[1], nuclear emulsions many m^2 in area[2], and huge plastic stacks[3]. It is important to ascertain the detailed charge distribution of these particles with $Z > 32$ and to establish whether the spectrum extends up into the trans-uranium region[4]. Charge identification depends on large extrapolations beyond previously studied regimes. With nuclear emulsions one compares the large signal from an extremely heavy cosmic ray with the small signal from an Fe nucleus at the same velocity and assumes a Z^2 response law[2]. With plastics one matches the response of the unknown nucleus of large β and the response of an Fe nucleus of small β and employs the appropriate (roughly β^{-2}) β response law[5,6]. Obviously it makes sense to use several independent methods to identify the same events. In this

paper we report simultaneous measurements of charges of 70 nuclei with $Z > 40$ using three detectors - Lexan polycarbonate, Eastman unplasticized cellulose triacetate, and Ilford G-5 nuclear emulsion.

2. Experimental Details

In a joint program at Bristol University, at General Electric, and at Washington University, three stacks consisting of plastics + emulsion + iron absorbers were exposed in September, 1968, on three flights at ~3g/cm^2 altitude over Palestine, Texas. All of Sky Monster I (area-time factor 600 m^2 hours) and half of Sky Monster II (area-time factor 300 m^2 hours) were surveyed in the present work. Other papers in these proceedings deal with the analysis of the emulsions in these stacks[7] and with measurements of fluxes of nuclei with $Z > 40$ in the third stack, Barndoor III[8].

Each stack contained a motor-driven, shiftable, upper layer of G-2 emulsion + Lexan + Daicell cellulose nitrate so that events occurring at ceiling altitude could be singled out. The rest of the stack, from top to bottom, consisted of G-5 emulsion + Lexan + 2 Bayer triacetates + 2 Eastman triacetates + .5 g/cm^2 of iron absorber + G-2 emulsion + Lexan + Daicell. The total stack thickness was 1 gm/cm^2.

About 2×10^9 cosmic rays, including over a million Fe-peak nuclei, passed through the portions of the stacks analyzed here. Our plastics have thresholds for track-formation such that slow Fe

nuclei are recorded but minimum-ionizing Fe nuclei are not. The present work concerns relativistic nuclei with $Z \gtrsim 40$, which could rapidly be located in the following way:

(1) One layer of Eastman triacetate (CTA) was etched 50 hours at 40°C in 2 parts 15% NaClO + 1 part 6.25N NaOH. All etched tracks that completely penetrated the sheet thickness (path length $> \csc\delta \cdot 125\mu$, where δ is dip angle) were found by using the ammonia vapor in an ozalid copy machine to make prints of their positions on chemically treated paper[9]. The scanning rate is faster than 1 m^2/hour.

(2) The top and bottom Daicell sheets were etched 2 days at 23°C in 6.25N NaOH. Only tracks that could be traced from the CTA into both Daicell sheets and that lined up with the top Diacell in its ceiling position were accepted. Being appreciably more sensitive than CTA, the Daicell gave easily visible tracks of all nuclei studied in the CTA.

In addition, the G-5 emulsion was independently scanned in a low-power stereomicroscope for tracks appreciably heavier than Fe so that scanning efficiencies in plastic and emulsion could be compared. The G-2 emulsions were used to check whether the tracks occurred at altitude and to set lower limits on velocity. Charges were determined in the G-5 emulsion as described elsewhere[7].

The charge assignments in emulsion were used to select

suitable etching times for small areas cut from the second Eastman CTA layer so that the two conical etch pits at each track would have an easily measurable length but would not become connected. From optical microscope measurements of these etch pit lengths, track-etching rates were computed and are entered in Figs. 1 and 2. (Charge calibration of CTA by use of low β, low Z cosmic rays is underway but is not completed.) About 40 events in CTA remain to be measured, predominantly those with $Z < 50$.

All three Lexan layers were etched 48 hours at 23°C in the alcoholic NaOH solution described in an accompanying paper[6]. Track-etching rates were measured in these layers and charge values were assigned on the basis of the calibration graph in Fig. 1 of that same paper[6], assuming $\beta=.95$ and taking dip angle into account. This calibration was based on an analysis of cosmic rays with $12 \leq Z \leq 30$ and $0.1<\beta<.45$, using an empirical ionization equation to extrapolate in Z and β. Any events for which the etching rate increased significantly from the top to the bottom Lexan sheet were rejected as not being minimum-ionizing. Charge assignments in Lexan are entered in Fig. 1. The dotted extension of the parabolic correlation curve in Fig. 1 was used to assign rough values of charge to those events that could be measured in CTA but not in Lexan. Final charge assignment in plastics - entered in Fig. 3 - was made by taking a weighted average of

the charge deduced in one sheet of CTA (from the correlation curve in Fig. 1) and in the three sheets of Lexan.

Two events from a previous flight - Barndoor II - had been measured in Lexan and in emulsion and are entered in Fig. 3 for purposes of comparison[3].

3. Results and Discussion

Table I summarizes the present capabilities of three plastics for rapid location of high-Z cosmic rays (which requires connected etched cones) and for charge identification (which requires cones of measurable length). The numbers quoted depend somewhat on sheet thickness and etching conditions. "Threshold Z" is that value of Z at $\beta=0.95$ for which the etching rate along a track is just equal to the bulk etch rate, so that even at a 90^{o} dip angle no cone is etched.

The correlation between measurements in CTA and in Lexan (Fig. 1) is good. Out of 25 events that could be measured in both plastics, 23 agree to within 2 charge units.

The correlation between measurements in CTA and in emulsion (Fig. 2) is not as good at high Z ($\gtrsim$70). Since it is worse for non-altitude events (not included in the figures) with a large spread of velocities, we believe that some of the spread in Fig. 2 could be due to velocity spread of the altitude events.

Among the other possibilities being tested in current flights is a time-dependence of the emulsion response due to its outgassing during the flight.

In Fig. 3 we see that at high Z the estimated charge in emulsion tends to be considerably higher than the estimated charge in either of the plastics. The disagreement is most severe for the most interesting event of all - one that appears to have $Z \approx 108$ in emulsion but only $Z \approx 90$ to 92 in plastics. This event showed no perceptible change in ionization through a path length of ~ 1 gm/cm^2 between the top and bottom Lexan sheets, from which we calculate that $\beta \gtrsim .92$ and that its charge in plastics could have been overestimated by no more than 1 or 2 charge units.

The difference in charge assignments by the two methods tends to disappear for $Z \lesssim 60$, which leads us to believe that the β-dependence assumed in calculating the response in plastics[5,6] is not seriously in error.

We now consider the Z-dependence. Within the stack the mean free path for loss of the K-electrons by even the heaviest relativistic nuclei is sufficiently small that their charges should equal their atomic numbers. In plastics the response at $\beta = .95$ was calculated from the response to Fe nuclei at β between 0.2 and 0.45, where we took the charge to be $Z^* = Z[1 - \exp(-125\beta/Z^{2/3})]$. The track-etching rate for the heaviest nucleus observed was the same as that of an Fe nucleus at $\beta \approx 0.2$,

for which the above relation gives $Z^* \approx 24.5$. Even if this value were in error and the true effective charge were as high as 26, our calculated charge for the heavy nucleus would be raised from 90 only to 95.5, which does not remove the discrepancy. A similar revision of the effective charge of Fe at an ionization rate equal to that of a relativistic nucleus with Z calculated to be 80 would increase the value only to 81.5.

When Mott scattering instead of the usual Rutherford scattering approximation is properly taken into account by including high orders of the Born approximation[10], we deduce that the production of delta rays of energies pertinent for normal track formation in both emulsions and plastics goes as Z^2 to a close approximation. The contrary conclusions reached recently by Semikoz[11] are invalid since he only considered the first and second Born approximations. We are then left with a disagreement in charge assignments at high Z that is not yet understood.

Referring to the charge histograms in Fig. 4, we consider that the peak in the distribution in plastics at a value near Pb ($Z = 82$) and the absence of the short-lived elements $84 \leq Z \leq 89$ are suggestive that the resolution of these detectors may be quite high and that the charge scale for plastics may not be too far wrong. Since a number of events with $Z < 50$ have not yet been measured in the CTA, the histograms should not

be construed as abundance distributions. Further, roughly half of the plastics on Monster II and all of the plastics on Barndoor III remain to be subjected to controlled etching, as well as those on our current flights at Palestine and Minneapolis. It seems premature until their analysis is completed to speculate further on the significance of the abundance distribution or on the possibility of trans-uranium cosmic rays.

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7. P.H. Fowler et al, these proceedings.

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TABLE I. Charge Determination Data in Plastics

	Threshold Z ($\delta=90^{o}$)	Practical minimum Z ($\delta=45^{o}$)	Minimum Z for rapid scan ($\delta\gtrsim30^{o}$)
Daicell	~26	28-29	30-32
Eastman CTA	not established	~38	52
Lexan			
a) Alcoholic etch	~45	~54	~72
b) Aqueous etch	~54	~59	~70

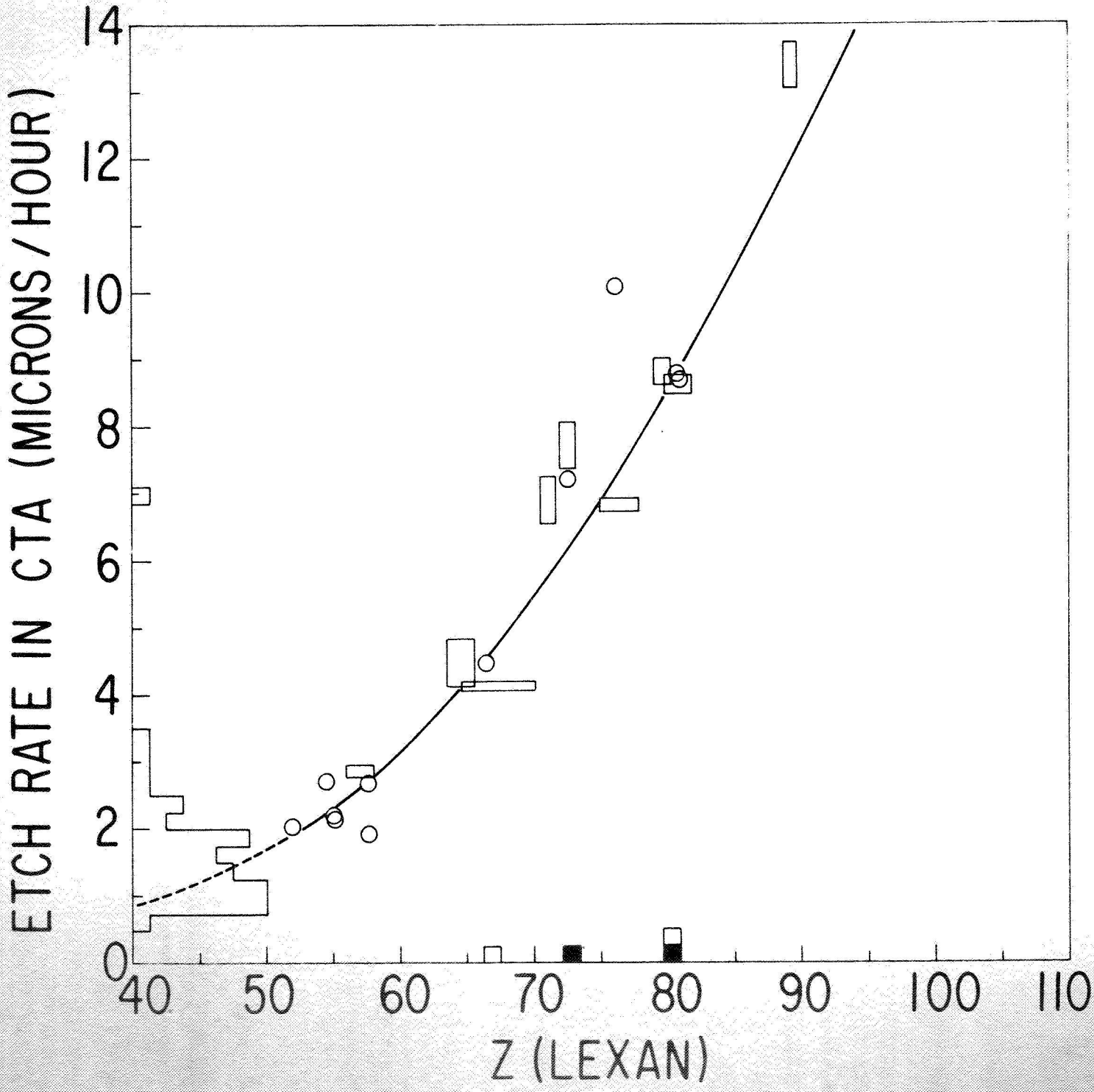
Figure Captions

1. Rate of lengthening of etch pits along tracks in Eastman cellulose triacetate (CTA) as a function of atomic number inferred from etch pit measurements on the same events in Lexan as described in the text. The histograms along the ordinate and abscissa pertain to events that were measured in only one of the two plastics. The two shaded events were measured in Barndoor II, which contained no CTA.

2. Rate of lengthening of etch pits along tracks in CTA as a function of atomic number inferred from measurements on the same events in emulsion.

3. Atomic number (weighted average) inferred from etch pit measurements in CTA and Lexan as a function of atomic number inferred from measurements on the same events in emulsion.

4. Charge assignments for those events that have been measured in <u>both</u> (a) plastics and (b) emulsion. The data were taken from Fig. 3. The distribution is biased toward high Z because some events with $Z < 60$ have not been measured yet.


ETCH RATE IN CTA (MICRONS / HOUR)
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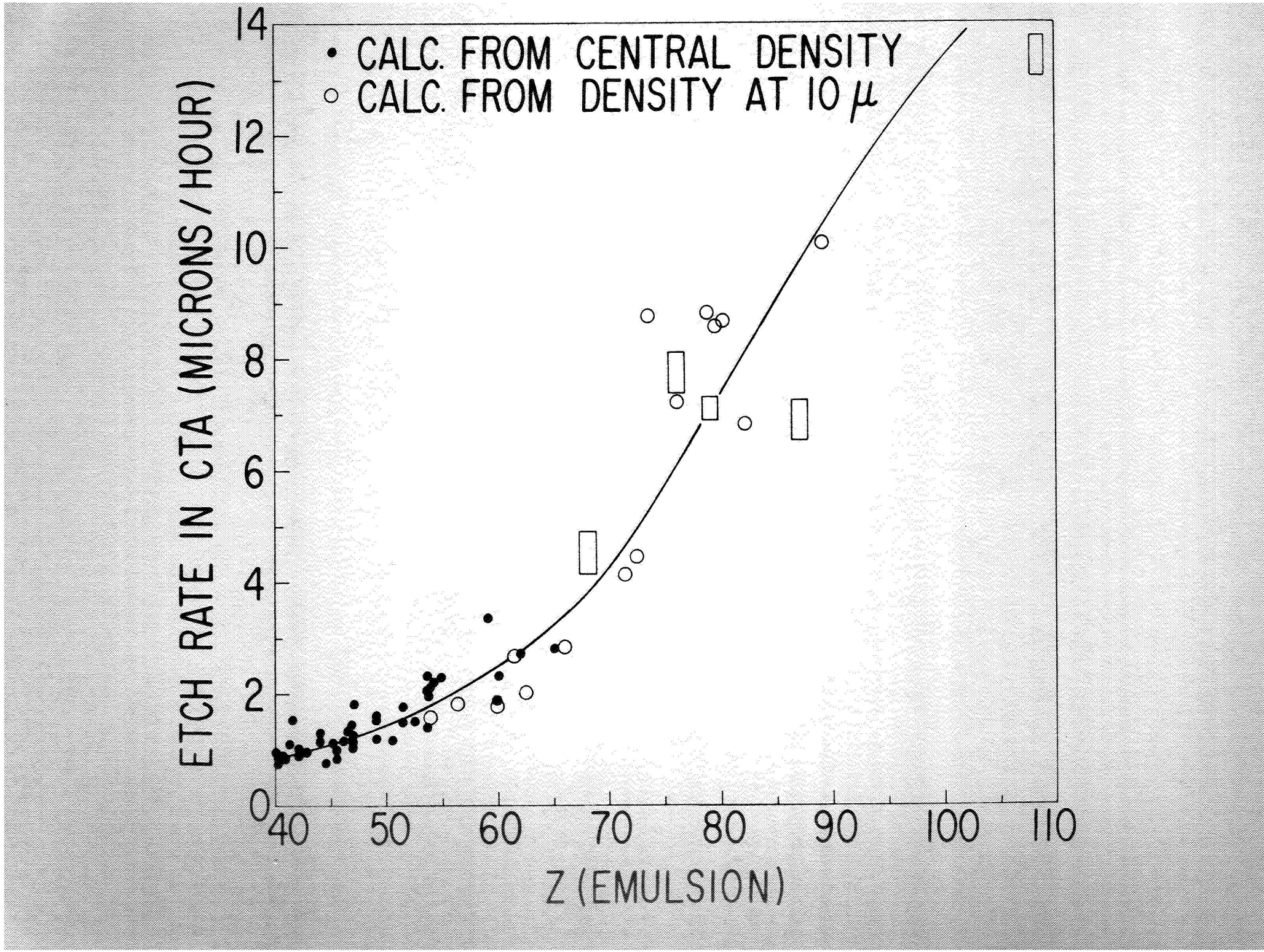
2
4
6
8
10
12
14
40
50
60
70
80
90
100
110
Z (LEXAN)


• CALC. FROM CENTRAL DENSITY
○ CALC. FROM DENSITY AT 10 μ
ETCH RATE IN CTA (MICRONS / HOUR)
0
2
4
6
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10
12
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50
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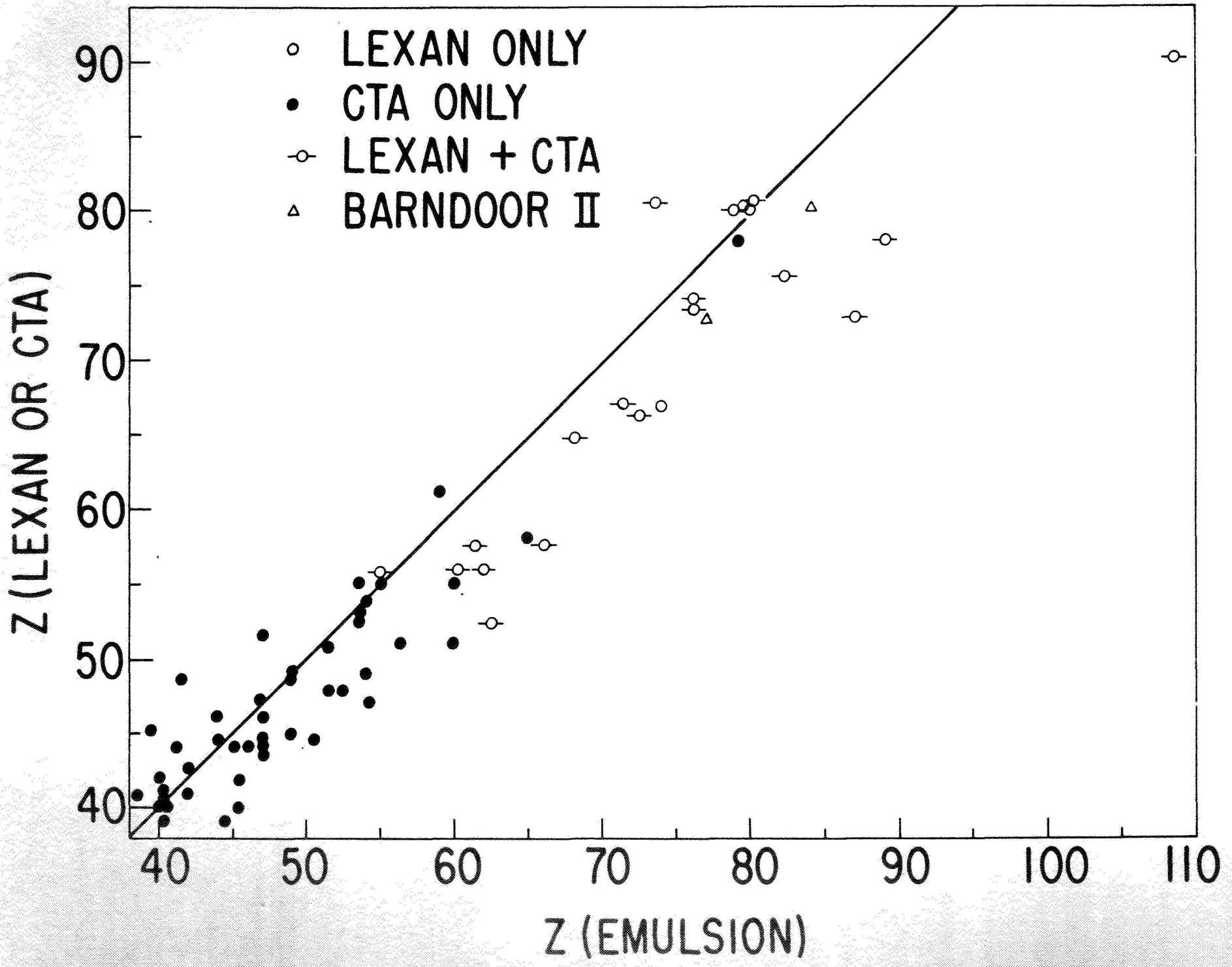
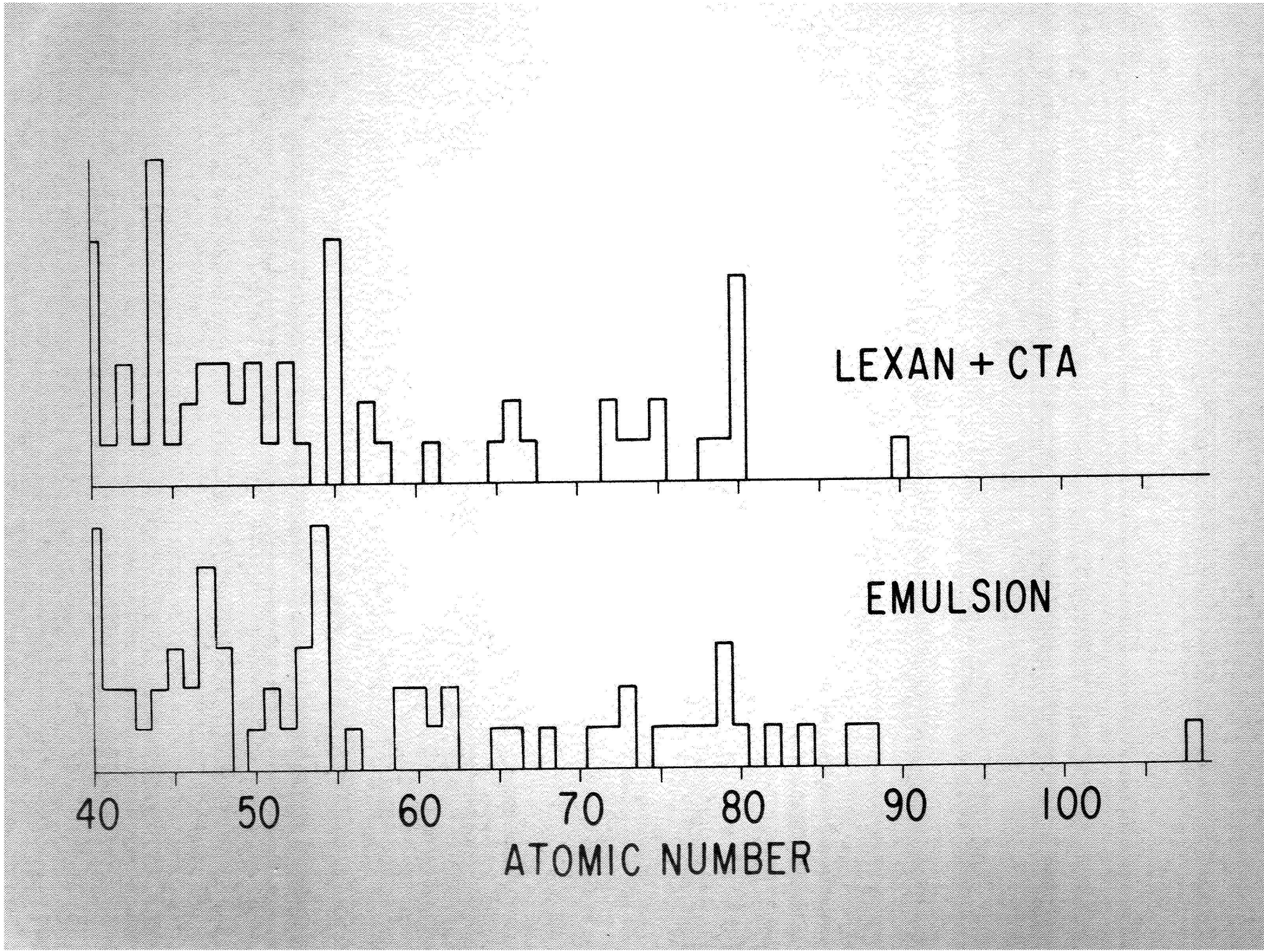
70
80
90
100
110
Z (EMULSION)


LEXAN ONLY
CTA ONLY
LEXAN + CTA
BARNDOOR II
Z (LEXAN OR CTA)
Z (EMULSION)
40
50
60
70
80
90
100
110


LEXAN + CTA
EMULSION
40
50
60
70
80
90
100
ATOMIC NUMBER

V. SUMMARY OF RESULTS AND RECOMMENDATIONS

V. Summary of Results and Recommendations

1. Acceptable Detectors for Cosmic Ray Identification

Of the numerous plastics studied, two with outstanding qualities were found acceptable: Lexan polycarbonate in 250μ thick sheets (type 8070-112, General Electric Chemical Materials Department, Mt. Vernon, Indiana, at $1.72 per pound) and Kodacel unplasticized cellulose triacetate in 125μ thick sheets (available on special order at $25 per pound from Eastman Chemical Products, 480 Cochitiate Road, Framingham, Mass.).

2. Qualities of the Plastics Chosen

a. Uniformity of response throughout a roll and through the sheet thickness.
b. Absence of flaws that etch preferentially.
c. Retention of latent (undeveloped) tracks for up to one year at room temperature in the dark.
d. Outstanding optical quality of etched tracks.
e. Etch rate that continues to increase at the highest ionization rates studied, with no sign of saturation.
f. Unlimited shelf life.
g. Sensitivity and resolving power for relativistic particles from $Z \approx 30$ up to at least 100.

None of the cellulose nitrates or polyesters met many of these conditions, nor did Bayer's cellulose triacetate. The Eastman triacetate (CTA) is sensitive to $Z \gtrsim 38$; the Lexan is sensitive to $Z \gtrsim 54$. Still to be found is a plastic that has good resolution in the region below $Z \sim 38$ and also meets all the other requirements. Commercially available cellulose nitrates that record particles with Z down to ~ 29 have many objectionable features that make them un-

acceptable. Further studies like those in section I may lead to new, more sensitive detectors and should be continued.

3. Calibration of Response of Lexan and CTA

Outstanding work by D. D. Peterson, under the direction of the principal investigator, has led to an understanding of the response of Lexan over the charge interval $12 \leq Z \leq 30$ and within the velocity interval $0.1 < \beta < 0.45$, A full exposition of this work will shortly be available as his Ph.D. thesis at Rensselaer Polytechnic Institute. At the ionization minimum ($\beta \gtrsim 0.92$) the calculated response of Lexan appears to be correct at least for $Z \sim 54$ to ~ 65, based on studies of the same events both in emulsions and in Lexan. At higher Z, there is a trend for the estimated charge in emulsion to be higher than the estimated charge in Lexan. We cannot yet say which method is in error. The response of Lexan to low energy O^{16}, F^{19}, and Ne^{20} ions (<10 MeV/nucleon) is lower than predicted, by an amount that increases with decreasing energy. It is not yet possible to say whether the effective charge or the β-dependence has been represented incorrectly. Further work at $\beta < 0.1$ needs to be done.

CTA has not yet been calibrated with stopping cosmic rays. It has, however, been calibrated rather well at $\beta > .92$ by intercomparing signals in CTA, Lexan, and emulsion from the same relativistic nucleus. At all Z the signals in CTA and Lexan correlate extremely well. At $Z \lesssim 60$ the charges deduced in emulsion and CTA tend to agree. At higher Z the trend is for the emulsion charge to be higher than the CTA charge. Current experiments with huge stacks of plastics and emulsions should clear up this discrepancy. Further support for the completion of these experiments would be desirable.

4. Environmental Effects

The standard procedure following an exposure is to age the plastics at room temperature in the dark for one month, then to process them as quickly as possible, keeping unprocessed sheets in a freezer at -30°C until ready for etching. New etching procedures at 40°C have eliminated any problem of track fading during processing.

A question remaining to be answered is the long-term constancy of response as a function of temperature and pressure during irradiation.

5. Assessment of Lexan and CTA

This work has confirmed the absolutely remarkable capability of Lexan for reliable, high resolution particle identification. Using the new etch developed during the period of the contract, we believe we are near to resolving certain isotopes of medium-mass stopping cosmic rays, and our charge resolution is extremely good. At very high Z we simply have no standard with which to compare, since the response of emulsion is not yet understood at high Z. We believe that it will turn out that the resolution of Lexan is better than 1 charge unit for even the heaviest nuclei.

CTA has the feature of being able to resolve lighter elements than can Lexan. Its resolution appears quite good. We recommend that effort be made to develop a more selective etchant such that the ratio of track-etching rate to bulk-etching rate is increased. Then the span of relativistic particles accessible for study might be extended down to Z as low as ~30.