Rev. Mexicana Astron. Astrof., **14**, 776-782 (1987)

RESOLUTION AND SENSIBILITY INCREASE IN CR-39 PLASTIC COSMIC RAY DETECTOR

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RESUMEN. Los rayos cosmicos son una parte más de la información que el Universo nos envía. De hecho es una valiosa información que no debemos desperdiciar. Una forma permanente de regristrar esta información, son los SSNTD (detectores de estado sólido por trazas nucleares), en donde existe una búsqueda permanente para incrementar la resolución y sensibilidad de dichos detectores con el fin de perder la menor información posible. El detector plástico que proporciona actualmente la mejor resolución y sensibilidad es el polímero CR-39 (polímero del dietilenglicol bis alil carbonato). Un material similar (POLINAOE) ha sido desarrollado en el INAOE para el estudio de partículas solares y radiación cósmica galáctica. Se ha establecido de manera completamente empírica que la resolución y sensibilidad de estos materiales plásticos se modifica cuando el material es impurificado con ciertos aditivos, tales como el dioctil ftalato. En este trabajo se reportan resultados experimentales y su correspondiente interpretación en un intento de profundizar en el entendimiento de estos efectos, dentro del marco de incrementar la resolución y sensibilidad del material detector.

ABSTRACT. Cosmic rays are part of the information that the Universe send to us. As a matter of fact, it is a valuable information that we should not loose. A permanent way of registering this information is by using SSNTD (solid state nuclear track detectors) where a continuous search has been done for incrementing the resolution and sensibility of such detectors with the purpose of loosing the least possible information. At present the best resolution and sensibility has been obtained with a plastic detector known as CR-39 (polimer of dietylen glycol bis allyl carbonate). A similar detector material (POLINAOE) has been developed at the INAOE for studies of solar particles and galactic cosmis rays. It has been stablished in a completely empirical way that the resolution and sensibility of these detectors are modified when the material is doped with and additive such as dioctyl phtalate. Here we report experimental results and our interpretation, in an attempt to go deeper in the understanding of these effects, within the frame of our intentions to increase the resolution and sensibility of the detector material.

Key words: INSTRUMENTS — DETECTORS

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I. INTRODUCTION

In the world of astronomy a substantial quantity of information comes from cosmic rays. To detect cosmic rays, we a need different kind of detectors that those used for light. A suitable kind of these detectors are the SSNTD, materials in which particles cause a damage while passing through them, damage that can be developed by etching technics, to see under the microscope the amplified particle track.

In nuclear track studies there is a constant search for improving the resolution and sensibility of the detector materials. For resolution we mean here the ability to distinguish between tracks of a given kind of particles of different but close energies, or particles of same energy but different mass,atomic number or effective charge. For sensibility we mean the capacity of the material to detect particles of enough low energy, that is that even particles of low energy produce a damage on the material because the ratio of etching track velocity to etching general or bulk velocity is high enough ($V_t/V_g \gtrsim 1$). Since the particle energy loss (dE/dx) depends on parameters such as the mass A, the atomic number Z and the effective charge q*, we can also refer resolution and sensibility for those other parameters.

It has been known for some time that as far as plastic detectors are concerned, when they are doped with some additives, the resolution and sensibility may change with respect to the plain (non doped) plastics. These are empiricalresults and the mechanism for explaining this change in resolution and sensibility has not yet been found. In the particular case of CR-39 it has been shown (Tarlé 1981) that when this is doped with dioctyl phtalate the resolution of the material increases with respect to that of CR-39 plain, though the sensibility sligthly decreases; it has also been shown that the effects of phtalates and terephtalates are practically the same. In this work it is attempted to go deep in the understanding of the processes involved in the alterations of resolution and sensibility for nuclear track detection when a plastic is doped. At last instance it is searched how to increase both resolution and sensibility, to set limits for that improvement without changing drastically the the physico-chemical properties of the material.

The present work is in some extent related with a previous work (Pérez-Peraza et al. 1983,1984) where the main goal was to study the effect of finite temperature of the detector material on the nuclear track during particle energy deposition. Among the several conclusions of that work is that the control of both resolution and sensibility depend on both, the macroscopic and microscopic elasticity properties of the material. The former are represented through Young's modulus (Y) and the latter by the interatomic elastic constant (k_s), which in turn is directly proportional to the dissociation energy and inversely proportional to the square of the interatomic distance in the equilibrium position. Therefore, the natural suite is to search for experimental techniques allowing to control those macroscopic and microscopic parameters, and in turn to get control on resolution and sensibility. We know from several works that doppants are able to modify both resolution and sensibility so, in some way they must act on the macroscopic and microscopic parameters, as we will hypothesize in the next section.

In the search for understanding the intercorrelations between the doping techniques and the macroscopic and microscopic parameters, we proposed in a preliminar work (Laville et al. 1985) that additives like phtalates and terephtalates act either as polimer chain openers or as blockers of cross-linking, whereas additives of the kind of benzoates may act only as blockers (fig. 1).

In that work, it was established that benzoate additives do not increase the resolution as high as phtalates or terephtalates, but it was better than the resolution without additives in CR-39 plain. Also it was found that the sensibility did not decrease with benzoate as much as doped with phtalates or terephtalates. We concluded that the predominant effect in increasing resolution was chain opening.

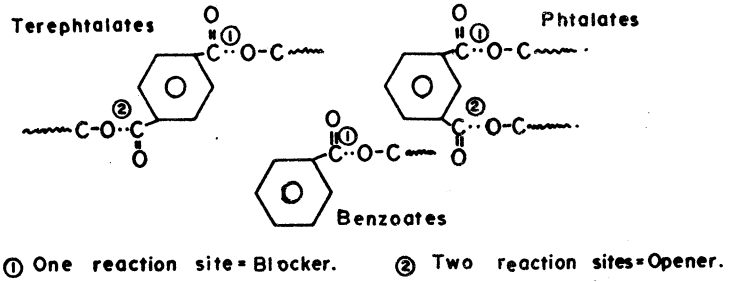

Fig. 1. Reaction sites in phtalates, terephtalates and benzoates.

II. THEORETICAL APPROACH

Our depart thesis in relation with the conclusions on the previous work, is that the Young modulus may be increased by increasing the separation between polymeric chains, that is, making softer the reticulation; or may be decreased by closing the separation between chains, that is, making the reticulation more compact. On the other hand, the interatomic spring constant may be increased or decreased by increasing or decreasing the cohesion energy respectively. The increase or decrease of the macroscopic elasticity does not necessarily entail the increase or decrease of the microscopic elasticity and cconversely. How do these processes are translated on resolution and sensibility of the material, to nuclear tracks of penetrating projectiles? If separation between chains is increased, the interaction cross section between projectile and target atoms diminishes, the energy loss of projectils is lowered and the particle range increases leaving a longer track. This entails an increase in resolution with respect to a more compact reticulation, because it is easier to discriminate between tracks of particles of neighbor energies as the reticulation becomes softer and bond density decreases: let us consider two particles of energies Ei, Ej above the threshold energy, such that $Ei-Ej=\Delta E$. Once particle of energy Ej has been stopped, it is easier for particle of energy Ei to cause still further damage on the plastic before stopping, on a softer net, whereas in a more compact reticulation the bond density is higher and so, for the same energy difference ΔE ,particle energy Ei is stopped very close to that of energy Ej, causing similar damage. In other terms with the separation of chains, damage along the track increases and this causes that tracks become more distinguishable among them. Under this point of view, resolution decreases by compacting the reticulation (fig. 2).

Now concerning sensibility, if separation between polymeric chains is increased, there is a compromise between, for one side, the increase in sensibility because of the decrease of dissociation energy of atomic bonds due to the enlargement of the reticulatioin cells with the subsequent decrease of cohesion (the chain net becomes softer, such that particles of lower energy are above the threshold energy) and from the other side, the decrease in sensibility by the fact that the cross section decreases with

cell enlargement. However this compromise is solved in the case of additives that contain a benzoic ring (as it is the case of the previously mentioned) because the benzoic ring chemical structure has a high stability that creates a bigger energy of cohesion, that increases substantially the dissociation energy in the sites of reaction between additives and monomeric molecules. The result is a decrease in sensibility as established by Tarlé (1981). Therefore projectils of energy lower than a certain threshold value are not able anymore to cause a damage in the doped plastic as they do in the plain one. However once they have the threshold energy they are better resolved in a doped plastic.

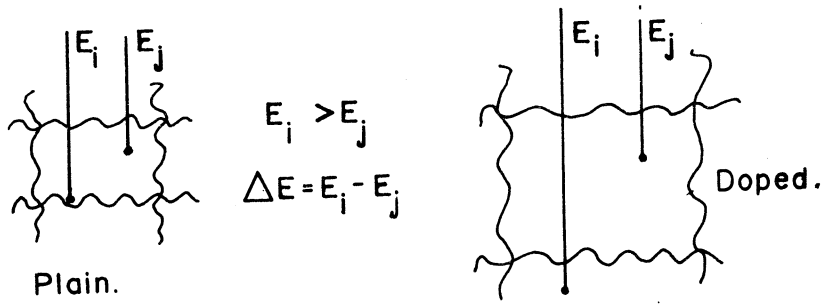

Fig 2.- Effect of the softening in the reticulation.

Now according to our thesis, four different things can happen during polimerization when we add the additives. Acordingly they are:

1.- OPENING: When molecules of the additives have two reaction sites as it can be the case of phtalates and terephtalates (see fig. 1) they interpose between the monomer molecules of two different chains, reacting with them on both sites during crosslinking of the monomer molecules thus having as a result the opening between chains and the softening of the cells, otherwise would have become more compact by the single effect of crosslinking without the additive (fig. 3).

2.- BLOCKING: Blocking of the chains is expected when the additive molecule have only one reaction site (see fig. 1), for instance benzoates, in which case the molecule reacting with one free radical opf a monomer chain will avoid that this particular chain will undergo crosslinking with another chain in that particular reaction site. The result is that the cohesion of the net becomes softer in those blocked sites. This is translated in a density decrease of the reticulation in relation with the non doped plastic, this is, though the cells become larger they are not so open or soft as in the case of the two reaction sites additives (fig. 4). We have to remember that phtalates and terephtalates can also work as blockers, when they present only one reaction site.

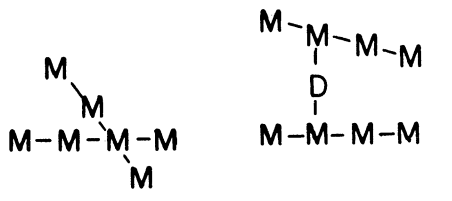

Fig 3.- Opening hypothesis.

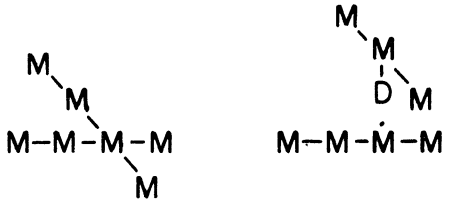

Fig 4.- Blocking hypothesis.

3.-RETICULATION: Suppose now that during polimerization instead of a dopant molecule, an individual monomeric molecule react with two monomeric chains causing crosslinking between them. The result is that the reticulation cells in those particular crosslinking sites becomes larger even when crosslinking occurs by means of a dopant molecule, because of the monomer molecules being physically bigger than the dopant molecules (fig. 5). Remember that this can happen even if no dopant is added.

4.- FILLING: Our last hypothesis supposes that now the dopant molecules place themselves between monomeric chains but without reacting with them. Since in this case the dopant molecule is not broken, the dopant molecule is larger than the monomeric molecules Therefore during conformation of the reticulation, the cells in those places becomes larger (fig. 6). We have to notice that benzoates can be smaller than the monomeric molecules as it is our particular case with benzyl benzoate, unexplaining how by this hypothesis, resolution can be increased with this particular benzoate.

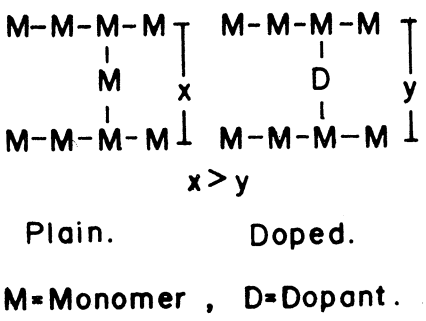

Fig 5.- Reticulation theory.

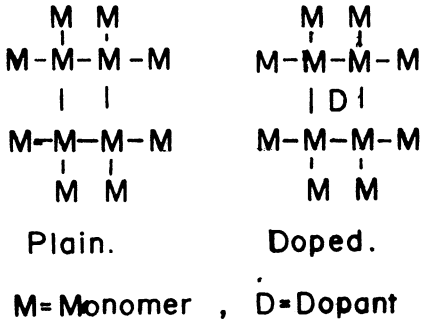

Fig 6.- Filling theory.

III. EXPERIMENTAL PROCEDURE

1.-ADDITIVES AND POLYMERS: Three different polymer sheets 5x5 cm^2 and 380 μm. thick were made. One plain and two doped. The two dopants used were dioctyl phtalate (DOP) and benzyl benzoate (BB). All three sheets were polymerized using benzoyl peroxide (BP) as initiator in an oven at a constant temperature of 70 ° C and a curing period of 72 hours (Table I.)

TABLE I.- Polymer sheets 5x5 cm^2. 380 μm. thick.

Polymer name	Initiator	Initiator conc.	Dopant	Dopant conc.
POLINAOE	BP	3%	-	-
POLINAOE(DOP)	BP	3%	DOP	3%
POLINAOE(BB)	BP	3%	BB	3%

NOTE: Initiator and dopant concentration is percentage by weight.

2.-IRRADIATION: Of the 5 cm^2. sheets, seven squares of 1 cm^2. of each plastic were cutted and irradiated with particles from a ^{252}Cf and ^{241}Am radioactive sources. The samples were placed in an irradiation fan. The fan had several slots perpendicular to the irradiation channels so that absorbers may be interposed in the particles trajectories before they could hit the plastics, and so different energies could be obtained. The absorbers where previously calibrated to give us the energy of the particle hitting the detector. The 21 samples were identified according to their additive and the irradiation they were going to receive. Once the samples and the absorbers were placed in the irradiation fan it was assembled in the versatile irradiation chamber (Balcazar-García and Pineda 1983) where they were irradiated with α particles.

3.-ETCHING AND READING: After the samples were irradiated these were etched in a NaOH solution 6.25 N at 70° C until the tracks diameters were measurable. This happened at an etching time of 3.5 hours. Fifty diameters were measured for each sample and the stastistical media was calculated. The same procedure was repeated for 5.5, 7.5, 9.5 and 11.5 hours, etching times. In addition one of the seven samples of each kind of plastic was irradiated with fission fragment from the Cf source. One fission fragment track was selected in each plastic and identified perfectly by a nearby marking. In this way we can track the evolution of the fission track diameter in time for the several etching times. It is well known that the slope of a graph of track diameter of the fission fragment against etching time is the bulk etching velocity (V_g).

IV. DATA ANALYSIS

Track diameters as a function of energy behave exponentially (Balcazar-García and Cuauhtecatl 1983):

$$\phi = k_1 \exp(-k_2 E)$$

k_1 and k_2 are characteristic of each etching time and for each type of plastic. To evaluate them is necessary to graph the data for just one etching time, in a semilogarithmic scale. Here we evaluate the constants for etching times 7.5 and 9.5 hours for the whole set of samples. Now, knowing the empirical expression between track diameter ϕ and energy E, we want to relate with V_t. Using $V=V_t/V_g$ and the well known expression:

$$\phi = 2 v_g t \sqrt{(v+1)/(v-1)}\,.$$

we obtain:

$$V = k_1^2 \exp(-2k_2 E) + 2Vt \Big/ 2Vt - k_1^2 \exp(-2k_2 E)\,.$$

where t is the correspondant etching time for which k_1 and k_2 were calculated.

form the latter expresion we can plot V against E and from the fact that $V=V_t/V_g$ we can plot V_t against E for the 7.5 and 9.5 etchinmg times. From figures 7,8,9,10, we clearly see the difference in slopes for the three plastics. We have to note here that as steeper is the slope of these graphs, the better the resolution of the plastic in question i.e. for small changes in energy bigger changes in V or V_t for steeper slopes. Then it is seen that the one with more resolution is the doped with DOP, then the doped with BB and at last the plain one.

V. DISCUSSION AND CONCLUSIONS

As we mentioned earlier, the increment in resolution may happen by one of the four hypothesis or by a combination of them. From figures 7,8,9,10, we discard the reticulation hypothesis because if true, we would expected that the plain plastic would have more resolution than the other ones, as this kind of reticulation happens in the absence of additives.Now, if we notice that plastic doped with DOP increases the resolution better than the one doped with BB and the latter one increases has a better resolution that the plain plastic, then if the filling theory were true, the BB additive should not have effect at all because of its molecules being smaller than the monomer molecules, filling nothing at all. The preceeding reasoning discards the filling theory. The difference in resolution between DOP and BB, confirms our earlier result that DOP works mainly as an opener but that it can work in a lesser extent,also as a blocker, this is shown by the improved resolution of the plastic doped with BB compared to the plain one

and considering that this additive can only work as a blocker after discarding the reticulation and filling theories.

From the careful analysis of the four hypothesis and the results shown, we can conclude that the most effective dopants are the ones that act mainly as openers as DOP does. We suggest the search for dopants that can open the chains more to corroborate completely this theory and at the same time, this new dopants should be absent of the benzoic ring, so that sensibility is not decreased.

As a last word, reticulation and filling theories are highly improbable especially the reticulation theory as it opposes completely to the other three hypothesis and does not check at all with the results presented here and on our earlier work.

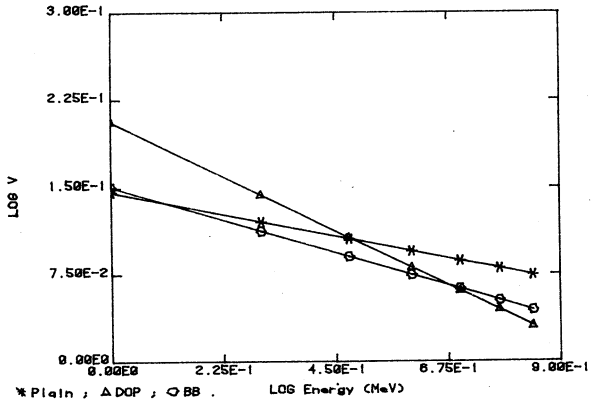

Fig 7.- V against E (7.5 hours).

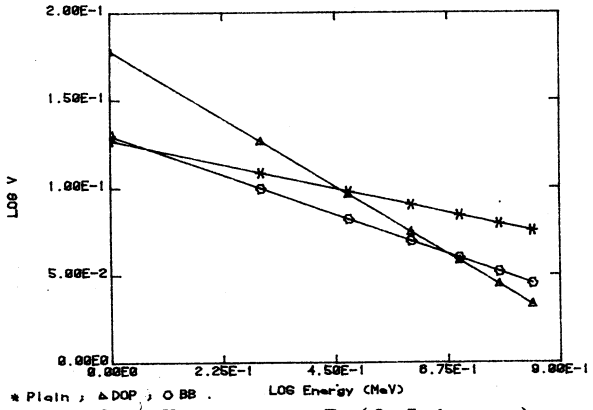

Fig 8.- V against E (9.5 hours).

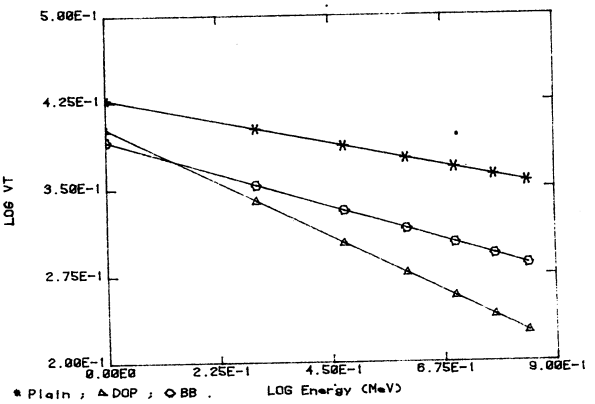

Fig 9.- V against E (7.5 hours).

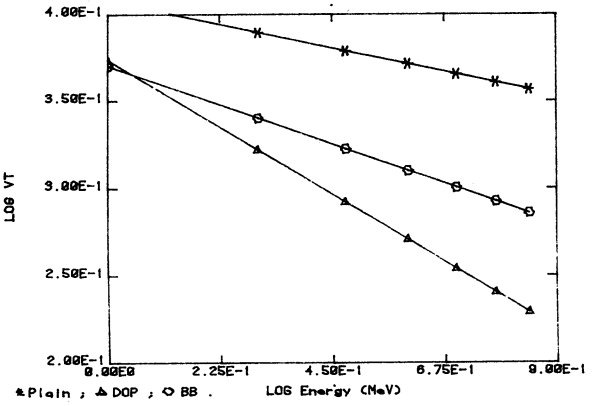

Fig 10.- V against E (9.5 hours).

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