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Mini Review

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Cosmic neutrons radiation damage in carbon nanotubes for space applications

Abstract

The main phenomenological aspects of the nuclear reactions induced by high-energy cosmic radiations in Carbon compounds are given. Results of measurements made with polycarbonate detectors at the International Space Station are shown and the main radiation damage mechanisms due to fast neutrons are discussed. Results of 0.02 DPA calculated value, indicate that carbon based matter start losing its intrinsic properties. Deterioration rate is further accentuated, when nuclear reactions are considered. Results evidence carbon nanometers structure radiation damage resistance of and show its suitability in space technology.

Keywords: space technology, nanotubes, linear energy transfer, elastic dispersion, nuclear track methodology, astronautic dosimetry

Volume 2 Issue 2 - 2018

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Received: January 24, 2018 | **Published:** March 20, 2018

Introduction

Radiation of solar, galactic and extra-galactic origin[1] that interact with the materials used in spacecraft and satellites in geostationary orbits or in interplanetary missions, experiment radiation absorption. High energy neutrons and energetic particles (HE) of high atomic number (Z) indicated as HZE,[2] induce nuclear reactions. The spatial vectors used in cosmic exploration subjected to radiation during their stay in extraterrestrial or interplanetary space, continuously receives a high LET (Linear Energy Transfer) radiation; consequently, in matter, elemental and molecular modifications are expected by elastic dispersion given in displacement per atom (DPA) and depending on the nature (energy and type) of the impinging particle, nuclear reactions. In this instance, the study is restricted to these phenomena taking place in the carbon nanotubes. Nanomaterials have a tensile force 100 times greater than steel and one sixth of their weight, they are excellent conductors of electricity and heat, employed often for their strongly bonded atomic structure.[3] Due to these intrinsic properties, they are employed, for example in the mechanical structure at the International Space Station (ISS); it is expected, in the future, to be used, more frequently in the construction of transport vectors and equipment required during the colonization of the planet Mars. In this study, nuclear track methodology (NTM) is applied to determine radiation damage to carbon compound, the passive charged particle detecting device, exposed to cosmic radiation[4] at the ISS International Space Station. The "Zvezda" service module of the Russian Federal Space Agency was employed to transport mentioned samples.

Materials and methods

The NTM is related to Poly Allyl Diglicol Carbonate or PADC) and applied for charged particle detection in the linear energy transfer (LET) that range above the threshold value of ~10 keV μm-1. Application in cosmic ray primary and secondary particles, including recoil nuclei, projectile and target fragments and secondary neutrons have shown excellent response[5,6] including in studies of ternary fission of light elements as the reaction n (^{12}C, 3 α) n`) in which three alpha particles are produced.[7] Depending on the energy of the cosmic neutrons, other nuclear reactions are also likely to occur such as: ^{12}C(n, T) ^{10}B; ^{12}C(n, D) ^{11}B; ^{12}C (n, p) ^{12}B. It is important to mention that the exposure to reaction induced by fast neutrons, damage on the long term, organs and tissues of the astronaut.[8] The most important reaction in astronautic dosimetry is ^{16}O(n, α) ^{13}C with a threshold value at 10 MeV and the effective cross section of the order of 1000 mb. Energetic neutrons >15 MeV after interacting with nuclei, either ^{16}O or ^{12}C, induce the formation of charged particles (such as p, D, T, α) that leave a low volume damage and high ion density in the absorbing matter (for example in carbon nanotubes). A massive displacement of the atoms is a result of the Coulomb explosion (high intensity electrostatic field). A similar phenomenon occurs with the nuclear reaction fragments for having a high LET value or stopping power in keV / μm). They induce atomic displacements in the molecular structure, with a not negligible intensity; often the consequence is the formation of a damage 1000 nm long with a diameter of 50–100 nm, in which the atoms depart from their original position modifying the mechanical properties of the materials (in the passive detector forms the so-called latent nuclear tracks). The selection of polyallyl diglycol-based carbonate detectors (PADC frequently reported in the literature as Columbia Resin number 39 or simply CR-39™) is particularly suitable to radiation damage studies. The passive matter chemical composition $(C_{12}H_{18}O_7)_n$, contain 6.6 % hydrogen in weight, C (52.6%) and O (40.8%). Through chemically treated latent nuclear tracks, the following are determined: i.- the rate of displaced atoms (DPA), ii.- the cosmic ray mass and energy trough damage left by nuclear reactions charged particle products or residual nuclei following transformation (n, x); (n, fission).[4] As mentioned the latent tracks require for visualization an enlargement of a factor of 100 or more; that must be carried out by means of a suitable chemical treatment. This is achieved, for example, by an etching solution of NaOH, 6N at 70°C; the chemical action is applied for 6 or more hours, depending on the radiation nature under study. This method provides enlarged tracks to determine by analyzing geometric shape, size and frequency, the identification of the impinging particle and therefore the damage cause. In the case of a carbon structure the displaced atom alters the microscopic properties of matter and in the case of carbon structure, is referred to as the Wigner's Disease (WD); the phenomenon described by Eugen Wigner in relation to nuclear reactors moderated by graphite. The importance of the WD effect in nanotube is determined by means of DPA, (which is interpreted as a dose i.e. energy per unit mass); usually in the mathematical formalism, the approximate Kinchin and Pease model is applied;[9] to determine DPA rate or R_d, the following equation applies:[10]

$$R_d = N\int_l^{\infty} \varphi\left(E_n\right) dE_n \int_{E}^{\Lambda E_n} \sigma_d\left(E_n, E\right) \nu\left(E\right) dE \qquad (1)$$

In which the product of the number of atoms per cm³ is N, the scattering cross section σ_d and the particle flow involved, $\phi\left(E_n\right)$, these two depend on the incident neutron energy (E_n). The value of the parameter DPA is determined from the previous equation in dependence of time *t*:

$$\therefore dpa = \frac{R_d t}{N} = t\int_{E}^{\infty} \Lambda \sigma_d\left(E_n\right) \varnothing\left(E_n\right) dE_n \qquad (2)$$

Using the equation (1) and

$$R_d = \frac{N\Lambda\sigma^{el} E_n}{4E_d} \int_0^{\infty} \varphi\left(E_n\right).dE_n \qquad (3)$$

Equation (2) assumes the following expression:

$$\therefore DPA = \frac{R_d t}{N} = t\frac{\Lambda\sigma^{el} E_n}{4E_d} \int_0^{\infty} \varphi\left(E_n\right) dE_n \qquad (4)$$

In which E_n, E_d are the average neutron and atom displacement energy respectively having considered the reduced mass given by:

$$\therefore \Lambda = \frac{4A}{\left(1+A\right)^2} \qquad (5)$$

The final expression assumes the following compact form:

$$dpa = t\frac{\Lambda\sigma^{el} E_n}{4E_d} \qquad (6)$$

In the case of carbon nanomatter, the following values apply: L=0.284; En= 5MeV; Ed =24 eV; elastic cross section 3[b], neutron flux of 107 n. cm^{-2}. s^{-1}.

The predicted values based on the above method can be then compared with those observed from PADC exposed to cosmic radiation at the ISS. Digitalized images such as given in Figure 1 are analyzed. Individual tracks corresponding to C and O ion recoils, are larger than most of those corresponding to lower mass charged particle. Then selecting the larger tracks, value for specific etched track density, (r_{spec}) is provided.[5] From earlier studies for C and O recoils or (r/F) the following calibration factor 2.4×10^{-6} applies. Calibration factor for the combined ^{12}C(n, n'x) and ^{16}O(n, n' x) reactions (En>5 MeV) is: 4.8×10^{-7} track per unit neutron fluence.

Results

Typical etched nuclear track induced by energetic neutrons interacting with carbon and oxygen nuclei (nuclear splitting in three energetic alpha particles, are given in Figure 1. From the analysis of digitalized images as those given in Figure 1, the reaction type and frequency was determined. Results are reported in Table 1. The values of the DPA were calculated employing equation (6).

Target elements given in (Table 1)(Table 2) are of natural isotopic composition. On Table 2, the most relevant reaction is indicated with their reaction rate for solar high neutron energy. The neutron energy threshold to induce three fragment fission on carbon nuclei is 15 MeV, for oxygen target is much lower, 10 MeV.

Some other information related to results reported are given as indicated in the next section below.

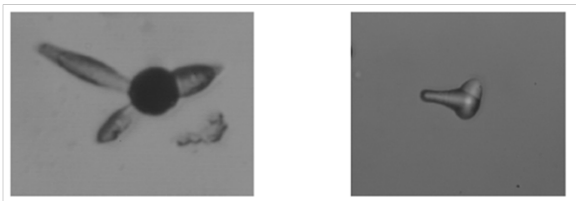

Figure 1 Etched nuclear tracks in PADC exposed to cosmic rays. The damaged region left by three alpha particles are evidenced.[7] The track pit size on the picture is 10 micro m.

Table 1 Theoretically predicted DPA.s-1 and charged particle nuclear reaction rate taking place in solid matter due to cosmic energetic neutrons, are reported

$E_n > 5$ **MeV** **Element**	**DPA.s^{-1} x 10^{-15}**	**E_d threshold (eV)**	**Reaction rate x 10^{-2} events.cm^{-2}.s^{-1}**	**Note**
C_{nat}	1.4	23.6	0.02	Nanotube
O_{nat}	0.069	32.2	20	Human environment
Al_{nat}	6.0	16.0	20	Metallic structure
Fe_{nat}	10	24.0	0.25	Metallic structure

Table 2 Predicted nuclear reaction rate taking place in carbon nanotube and oxygen isotope for comparison. Neutron and gamma ray induced reaction, of importance in extraterrestrial environment. Reaction reported in first column are those that were detected on a space mission to the ISS employing PADC6

$E_n > 5$ **MeV** **Target isotope**	**Reaction rate x 10^{-2} events.cm^{-2}.s^{-1}**	**Note: production of**
n (^{12}C, 3α) n'	10	He gas
^{12}C (n,t) ^{10}B	3	Tritium radioactive gas
^{12}C (n,d) ^{11}B	0.7	Deuterium gas
^{12}C (n,p) ^{12}B	0.2	H_2 gas is produced
^{16}C(n, α) ^{13}C	10	He gas
^{16}O (γ,n) ^{15}C	0.1	Photo-neutrons
^{12}C (n, n' x)	2.4 x 10^{-4}	^{12}C recoil
^{16}O (n, n' x)	2.4 x 10^{-4}	^{16}O recoil

Discussion

The cosmic neutron has been recognized as the main source of atomic displacement and reaction rate related to radiation damage during long term missions to the Red Planet. Regarding the DPA by neutrons and the induced nuclear reactions including cosmic rays we have values reported in Table 1, that provide information on the matter and human exposure to HEZ radiation. In the determination of the mentioned results as first approximation, three groups of neutron energies $E_{n',i}$ (i = 0.025 eV, 2 MeV and 200 MeV) were considered specifically in the calculations of the DPA and nuclear transformations rates with neutrons including gamma ray with $E_{□□}$ > 15 MeV in that the effect on braking by channeling or ion channeling was not considered. We have observed that the atomic dislocation impact on carbon nanotubes, is one thousandth in comparison with e.g. The Al. However, if we include the carbon material mass factors (tons) and the exposure time (years), we observe that using 1 m³ of material in a mission to the planet Mars and assuming two spatial neutron

Citation: Bohus LS, Martínez OSA, Palacios D,et al. Cosmic neutrons radiation damage in carbon nanotubes for space applications. *Phys Astron Int J.* 2018;2(2):94–96. DOI: 10.15406/paij.2018.02.00067

groups of 1 and 100 MeV, the number of dislocated atoms is comparable with the Avogadro's number. By a thermodynamic effect (annealing) the atoms dislocated by neutrons, statistically occupy positions that they have occupied previously i.e. low WD-value. The neutrons of the spectrum with energy of the mentioned groups induce ternary fission in the graphite in addition to the reactions with the generation of hydrogen and its isotopes in addition to helium. These gases occupy interstitial spaces thus inducing an additional factor that worsen material properties. The most frequent and important damage, occurs with the reaction n(^{12}C, 3α)n' and it is of such magnitude that decrease the material mechanical properties, in particular the tensile strength worsens by 5% per year of permanence in the interplanetary space. From data comparison given in Table we may observe the the difference is significant however considering several approximation affect the predicted values. Nevertheless we may observe that values indicate the c and O recoil process is more frequent compared to charged particle production.

Table 3 Experimental values obtained analizing digitalized etched nuclear track detectors. PADC exposed on board of the ISS. Experimental and calculated values are given in column 2 and 3 respectively

Nuclear track related to reactions	Measured value by etched track	Predicted DPA per unit fluence	Note
^{12}C (n,x) x charged particles: t, d, p, α	0.48 × 10^{-6}	1.16 × 10^{-6}	Including carbon 12 splitting in 3 alphas
^{16}O (γ ,n) ^{15}C	Not considered	Not considered	Photo-neutrons not included
^{12}C (n, n' x) +^{16}O (n, n' x)	2.4 × 10^{-6}	22.5 × 10^{-6}	^{12}C +^{16}O recoil tracks

Conclusion

Cosmic radiation damages induced in carbon nanostructure have been studied by passive devices. Results show that the phenomenon is relatively of low frequency for the assumed fluencies i.e. space mission of around 250 days, indicating that it is possible to use nanocarbon tubes or in general carbon based bulk matter as structural material in the space-ship technology for longer period of time than the mentioned space missions. Predicted values determined in this study differ from published data however it is demonstrated that carbon nanotubes maintain their properties for long exposure time in a cosmic environment. Space vectors are subject to radiation damage that is a limiting factor since resistance to traction may be reduce with exposure time; in fact is reduced by 40% when exposed to cosmic neutron spectrum; also the number of displaced atoms is 17% of the total while for exceptionally large flow (of 10^{15} n/s) the DPA is 0.02. This value corresponds to the case of exposure during a three years space mission to the planet Mars assuming typical solar events. From radiation damage (DPA), it can be observed that carbon nanotubes for space applications, do not offer major technical advantages compared to other materials for instance Al or Fe. However, considering nuclear reactions induced radiation damage, the outcome is diametrically opposite, in fact carbon nanotubes offer several advantages when employed as space-ships matter. For a mission to the planet Mars with a load of only 10% (in weight) elemental C-protuberant matter, the expected displacement number is 17%. The number is not negligible; however, due to the presence of the relaxation effect, the material suffers lower deterioration to radiation. Results put in evidence that for larger flows (case of solar explosions or the black spots observed by Galileo in 1640) a DPA-value can be high as 0.02, indicating that the material looses steady fast its intrinsic properties. The case is further accentuated, when nuclear reactions are considered. It is worth mentioning that based on the technology and transport vectors capacity currently available, the colonization (limited to six years) of the Red Planet with crew of six astronauts require more than a thousand tons of various materials (fuel, supplies, radiological shielding, construction material, support machinery, among others) that implies one hundred launches. We understand that for a comprehensive phenomenology further study should be convenient, nevertheless results given here indicate that radiation damage in carbon nanostructure has an important aspect in the continuously evolving and growing space technology.

Acknowledgements

Authors are grateful to: Dr Palfalvi who provided the PADC exposed detectors, Russian and Hungarian Space Agency for the detectors transport to the ISS and to Tasltrak of Bristol UK for providing the PADC high quality detectors.

Conflicts of inertest

Authors declare there is no conflict of interest.

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