Journal of Physics: Conference Series

Electron beam induced microstructural changes and electrical conductivity in Bakelite polymer RPC detector material: A positron lifetime study

To cite this article: K V Aneesh Kumar *et al* 2015 *J. Phys.: Conf. Ser.* **618** 012032

Electron beam induced microstructural changes and electrical conductivity in Bakelite polymer RPC detector material: A positron lifetime study

K V Aneesh Kumar[1], S Ningaraju[1], L M Munirathnamma[1], H B Ravikumar[1] and C Ranganathaiah[1,2]
[1] Department of Studies in Physics, University of Mysore, Mysore-570006, India.
[2] Govt. Research Centre, Sahyadri College of Engineering & Management, Adyar, Mangalore – 575 007, India

E-mail: cr@physics.uni-mysore.ac.in:crang1@rediffmail.com

Abstract. In order to explore the structural modification induced electrical conductivity, samples of Bakelite RPC polymer detector materials were exposed to 8 MeV of electron beam with the irradiation dose from 20 kGy to100 kGy in steps of 20 kGy. The microstructural changes upon electron beam irradiation have been studied using Positron Annihilation Lifetime Spectroscopy (PALS) and Fourier Transform Infrared (FTIR) Spectroscopy. Positron lifetime parameters viz., o-Ps lifetime and its intensity show chain scission at lower doses (20 kGy, 40 kGy) followed by cross-linking beyond 40 kGy due to the radical reactions. The reduction in electrical conductivity of Bakelite material beyond 60 kGy is correlated to the conducting pathways and cross-links in the polymer matrix. The appropriate doses of electron beam irradiation of Bakelite material may reduce the leakage current and hence improves the performance of the detector.

1. Introduction

Resistive plate chambers (RPCs) [1] have found extensive applications in high energy and astrophysics experiments as the active detectors for muon detection. The RPCs made up of high resistive materials like Bakelite and glass are being used as the detectors for iron calorimeter (ICAL) to study atmospheric neutrinos in the proposed India based neutrino observatory (INO) project in India [2]. As the RPC detectors are continuously exposed to charged particles like cosmic ray muons, there will be some modification in the microstructure of RPC material in the long run. The vital problem faced in such experiments is that Bakelite RPCs exhibit undesirable high leakage current compared to the glass RPCs [3]. However, Bakelite has several advantages over glass detector material like low cost, easy to handle in the fabrication of RPCs and as such it is very important to understand the origin of this leakage current. This problem has not been rectified in the past and it is interesting to find whether the high leakage current owes its origin to the microstructural changes of the Bakelite material upon exposure to charged radiation. In recent years, irradiation is treated as an effective tool for structural modification of polymers and plays a significant role in the material modifications. In the present study, authors carried out experimental investigations on the effects of electron beam irradiation on the microstructure of the polymer based Bakelite RPC detector material by one of the well established techniques viz., Positron Annihilation Lifetime Spectroscopy (PALS) [4] and an attempt has been made to correlate the free volume change due to the electron beam irradiation on the electrical conductivity.

2. Experimental

2.1. Sample Preparation and Electron Beam Irradiation

Bakelite samples (P-120, Matt finished NEMA LI-1989, Grade XXX) of density 1.22 g/cm^3, manufactured by Bakelite Hylam, India were procured from VECC Kolkata (INO-Lab), India. The chemical structure of Bakelite is shown in figure 1. The pairs of rectangular samples having dimensions 1 cm x 1 cm x 0.175 cm were exposed to the electron beam of energy 8 MeV in air at Microtron Centre, Mangalore University, India, for different doses up to 100 kGy in the interval of 20 kGy. These samples were used for PALS, electrical conductivity and FTIR measurements.

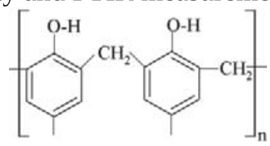

Figure 1. Chemical structure of Bakelite.

2.2. Positron Annihilation Lifetime Measurements

Positron annihilation lifetime spectra were recorded for the as received and electron beam irradiated Bakelite samples using positron lifetime spectrometer. The detailed description of PALS technique was given in our earlier paper [5]. The o-Ps lifetime (τ_3) is related to the free volume hole size by a simple relation given by Nakanishi *et al*. [6], which was developed on the basis of theoretical models originally proposed by Tao [7] for molecular liquids and later by Eldrup *et al*. [8].

2.3. FTIR Characterization

The Fourier Transform Infrared (FTIR) Spectra for the as received and the electron beam irradiated Bakelite samples at different doses were recorded using KBr pellet method in the range of 4000-400 cm^{-1} with JASCO-460 Plus, Japan FTIR Spectrometer with a resolution of 4 cm^{-1}.

2.4. Electrical Conductivity Measurements

The electrical conductivity of the as received and electron beam irradiated Bakelite samples at different doses were measured using a standard set up with Keithley 2636A model with Source meter and computer program Lab Tracer 2.0.

3. Results and Discussions

3.1. Positron Annihilation Lifetime Spectroscopy

PALS results reveal that the free volume size increases from 64.90 Å^3 to 69.46 Å^3 on electron beam irradiation up to 40 kGy and then decreases continuously to 58.93 Å^3 at 100 kGy (figure 2). This can be explained as follows; Positronium (Ps) formation takes place preferentially at the free volume cavities, which are the regions of low electron density exist mainly in the amorphous domains of Polymers. Upon irradiation, the scission of polymer chains will increase the size of the free volume cavities and hence the o-Ps lifetime (τ_3) [7]. Above 40 kGy, there is a remarkable decrease of o-Ps lifetime (τ_3) about 71 ps and free volume size (V_f) by about 6 Å^3 starting from 40 to 100 kGy irradiation dose in comparison to as received sample. This is attributed to the cross-linking of Bakelite polymer chains due to the radicals formed by the scission of polymeric chains in the initial stages of irradiation; this will hinder the polymer chain mobility [9]. On the other hand, the o-Ps intensity (I_3) decreases up to 60 kGy and a small increase at 80 kGy (only about 0.5%) followed by a decline up to 100 kGy dose as seen from figure 3.The decrease in I_3 may be due to inhibition of positronium (Ps) formation. The cleavage of bonds in Bakelite yield phenyl radicals, these radicals undergo reactions with the electrons of the spur created during slowing down of positrons, thereby inhibits the formation of positronium (Ps) [10]. Also, there is a possibility of the interaction of free radicals with o-Ps, this may cause ionization or oxidation. All such interactions may contribute to the inhibition of o-Ps formation. The variations of o-Ps intensity from 60 to 100 kGy is due to the structural modification of Bakelite polymer induced by the chain scission followed by the cross linking of the polymer chains. These arguments are adequately supported by FTIR results.

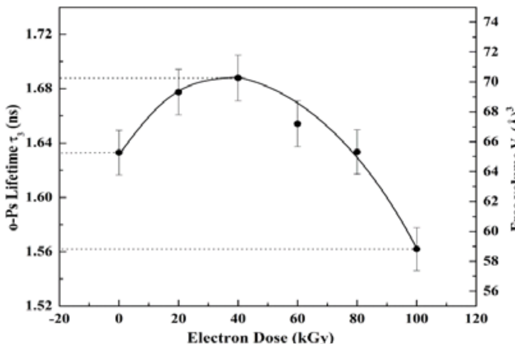

Figure 2. Variation of o-Ps lifetime (τ_3) and free volume size (V_f) as a function of electron dose.

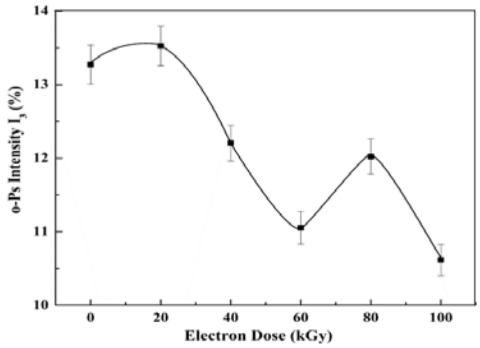

Figure 3. Variation of o-Ps intensity (I_3) as a function of electron dose.

3.2. FTIR Studies

The chemical modifications caused by electron beam irradiation on Bakelite are understood using FTIR scans shown in figure 4. The absorbance bands at wave numbers 1457, 1647, and 2923 cm^{-1} correspond to the functional groups viz., C-H aliphatic bridge, C=C aromatic ring and stretching vibrations of CH_2 alkane respectively [5, 11]. Since no changes in these wave numbers observed upon electron beam irradiation suggest that they do not influence structural modification of Bakelite. On the other hand, the absorption band at 3394 cm^{-1} corresponds to Phenol/OH in the as received sample gets shifted to 3420 cm^{-1} at 20 kGy and remains at this value for higher electron doses. As the radiation dose increases, the % of transmittance also increases which can be attributed to the chain scission resulting to the formation of more number of phenolic radicals. This may further leads to the formation of OH^- and H^+ ions [12, 13]. The decrease in % of transmittance of O-H group at higher electron doses suggests the cross linking of polymeric chains of Bakelite. This is possibly due to the formation of hydrogen bonds by the radicals released during the scission of phenolic groups [11, 14]. The absorbance band at 1053 cm^{-1} corresponding to single bond C-O stretching vibrations of -CH_2OH- group shifted to 1036 cm^{-1} at 20 kGy and then increases to 1055 cm^{-1} and remains at 1055 cm^{-1} for higher dosages. These results indicate that the chemical changes due to the rearrangement of free radicals after the chain cleavage seems to contribute to the microstructural modifications in the Bakelite sample. These results agree well with the PALS results.

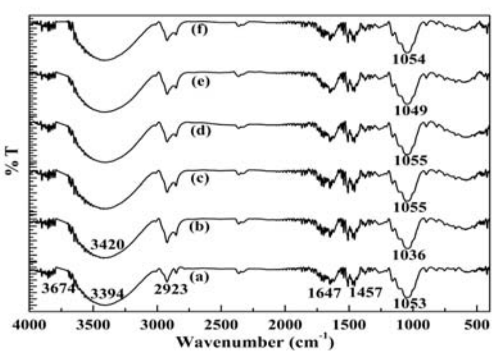

Figure 4. FTIR Spectrum. [(a) to (f) represents FTIR spectra of as received to 100 kGy electron beam irradiated Bakelite samples in the interval of 20 kGy.]

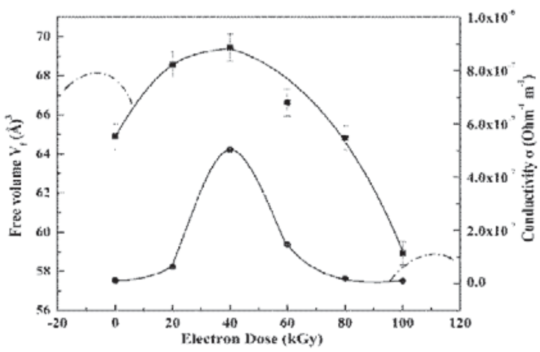

Figure 5. Variation of free volume size (V_f) and electrical conductivity (σ) as a function of electron dose.

3.3. Electrical Conductivity Results

Figure 5 shows the variation of free volume size and electrical conductivity as a function of electron dose and they exhibit behaviour upon electron beam irradiation that as the free volume increases, conductivity increases and vice-versa. The electrical conductivity (σ) of the as received sample was 1.23×10^{-8} $(\Omega m)^{-1}$. It was observed that the electrical conductivity increases on irradiation of electron beam dosage up to 40 kGy and then decreases to 1.08×10^{-8} $(\Omega m)^{-1}$ at 100 kGy. The increase in conductivity for the low electron doses may be due to the scission of hydrogen bonded phenolic groups lead to the formation of OH^- and H^+ radicals [5, 15]. At higher doses (above 60 kGy) cross linking is the predominant process and the formation of cross-links of the network hinders the movement of chains inside the polymer. This indicate the formation of cross-links of Bakelite polymeric chains and reduces the formation of free radicals and reduce their mobility and hence reduction in electrical conductivity [16, 17]. This is also evident from the reduced free volume size (V_f) obtained from the PALS study.

4. Conclusions

PALS results revealed that chain scission and cross linking are the predominant processes under electron beam irradiation on polymer based Bakelite RPC detector material. FTIR spectroscopy indicates the scission of hydrogen bonded phenolic groups leading to the formation of OH^- and H^+ radicals in the low dosage region. The electrical conductivity for 40 kGy electron irradiated sample is more compared to the as received sample indicative of increased mobility of a large number of OH^- and H^+ ions produced from the cleavage of hydrogen bonded phenolic groups. The increased cross-links at higher electron dose may possibly reduce the leakage current although it depends on the surface properties of the materials as well. We propose that improvement in the performance of Bakelite RPC detector is possible with electron beam irradiation resulting in reduced leakage current.

Acknowledgement

The authors are grateful to DST, Govt of India for sanctioning the INO-DST Project to UOM, Mysore.

References

[1] Santonico R and Cardarelli R 1981 *Nucl. Instr. Meth.* **187** 377-380
[2] Biswas S, Bhattacharya S , Bose S, Chattopadhyay S, Saha S, Sharan M K and Viyogi Y P 2009 *Nucl. Instr. Meth. Phys. Res. Sect. A* **602** 749-753
[3] Carlson P, Crotty *et al*. 2001 Study and optimization of RPCs for high rate applications *IEEE Nuclear Sci. Symp. Medical imaging Conf.* (USA, 4-10 November 2001) pp 1-17
[4] Shaojin J, Zhicheng Z, Yangmeib F, Huimingb W, Xianfengb Z, Rongdiana H 2002 *Eur. Polym. J.* **38** 2433
[5] Aneeshkumar K V, Ravikumar H B and Ranganathaiah C 2013 *J. Appl. Polym. Sci*.**130** 793-800
[6] Nakanishi H, Wang S J and Jean Y C 1988 Microscopic surface tension studied by positron annihilation, in: Sharma S C (Ed.) Positron annihilation in fluids Singapore, *World Scientific* pp 292- 93
[7] Tao S J 1972 *J.Chem. Phys.* **56** 5499-5510
[8] Eldrup M, Lightbody D and Sherwood 1981 *Chem. Phys.* **63** 51-58
[9] Ismayil, Ravindrachary V, Bhajantri R F, Praveena S D, Poojary B, Dutta D and Pujari P K 2010 *Polym. Degrad.Stab.* **95** 1083-91
[10] Shariff G, Sathyanarayana P M, Thimmegowda M C and Ranganathaiah C 2002 *Polymer*.**43** 2819
[11] Lee Y, Kim D, Kim H J, Hwang T S and Rafailovich M 2003 *J. Appl. Polym. Sci*. **89** 2589-96
[12] Loo J S C, Ooi C P and Boey F Y 2005 *Biomaterials*.**26** 1359-67
[13] Chen Y, Chen Z, Xiao S and Liu H 2008 *Thermochim. Acta*. **476** 39-43
[14] Roczniak K, Biernacka T and Skarzynski M 1983 *J. Appl. Polym. Sci.* **28** 531-542
[15] Va'vra J 2003 *Nuclear Symp. Conf. Record 2003 IEEE*. **5** 3704-08
[16] Siti A, Mohd N, Azizan A, Mohd Y A R and Ibrahim A T T 2010 J. *Nat. Sci*. **2** 190
[17] Liu H, Hu X B, Wang J Y and Boughton R I 2002 *Macromolecules*.**35** 9414-19