



The Effect of Co-60 Gamma Irradiation on Chemical, AC and DC Electrical Properties of Ammonium Dihydrogen Orthophosphate Non-linear Optical Crystal

B. C. Hemaraju, A. P. Gnana Prakash*

Department of Studies in Physics, University of Mysore, Mysore - 570 006, Karnataka, India.

Received 15th April 2016; Revised 03rd May 2016; Accepted 05th May 2016

ABSTRACT

Single crystals of ammonium dihydrogen orthophosphate crystals were grown by slow evaporation technique. The grown crystals were irradiated by Co-60 gamma radiation with cumulative doses of 1, 5, 10, 15, and 20 Mrad. The functional groups of unirradiated and irradiated crystals have been identified and confirmed by Fourier transform infrared studies. The AC and DC conducting properties were studied before and after irradiation. The chemical, AC and DC conductivity was changed significantly after gamma irradiation.

Key words: Non-linear optical material, Co-60 gamma irradiation, Fourier transform infrared, AC and DC conductivity.

1. INTRODUCTION

Non-linear optical (NLO) crystals play a very important role in solid state lasers, which are sometimes used in instruments of orbital space missions, space-based light detection, and ranging systems [1,2]. In the field of optics and photonics NLO crystals contributing number of laser applications. The development of solid state lasers for spaceborne applications needs to know the radiation tolerance of the crystals. Spaceborne systems will be required to withstand several years of exposure to high-energy cosmic rays, gamma rays, and charged particles such as electrons, protons, or heavier particles [3]. Exposure of NLO crystals for such radiation can cause the defects in the crystal system such as vacancies, trapping centers, and color centers. The study of radiation-induced defects and electronic interaction becomes essential in various spaceborne system design. Therefore understanding the effects of ionizing radiation on crystals are very much essential. Therefore, the effects of Co-60 gamma irradiation on ammonium dihydrogen orthophosphate (ADP) crystals were studied from 1 to 20 Mrad. The chemical properties of the grown crystal were studied by attenuated total reflectance (ATR) - Fourier transform infrared (FTIR) before and after irradiation. The AC and DC measurement were conducted before and after every cumulative dose.

2. EXPERIMENTAL

AR grade sample of ADP was dissolved in double distilled water and stirred well for about 3 h using

a magnetic stirrer to get saturated solution. The solution was then filtered using Whatman filter paper to remove the impurities and allowed to crystallize by slow evaporation of solvent at room temperature. Good transparent crystals of size $10 \times 10 \times 3 \text{ mm}^3$ were obtained in a period of about 4-week and are shown in Figure 1.

The grown crystals were irradiated in Co-60 Gamma Chamber-1200 with a dose rate of 9.187 kGy/h at Department of Studies in Physics, University of Mysore, India. The different gamma radiation dose given to NLO crystals were 1, 5, 10, 15 and 20 Mrad. The changes in functional groups of the grown crystals were recorded using JASCO FTIR-4100 with ATR attachment. The AC conductivity, dielectric constant and dielectric loss of unirradiated and irradiated crystals have been measured using HIOKI 3532-50 LCR Hitester in the frequency range from 100 Hz to 5 MHz. The DC conductivity was performed using Keithley dual channel source meter (model 2636A).

3. RESULTS AND DISCUSSION

3.1. FTIR Spectroscopy

The infrared spectral analysis provides useful information regarding the molecular structure and functional groups of the compound. The infrared spectrums of unirradiated and irradiated ADP crystals were recorded in the frequency range $500\text{-}4000 \text{ cm}^{-1}$ and are shown in Figure 2. The bands appearing

*Corresponding Author:

E-mail: gnanaprakash@physics.uni-mysore.ac.in

Phone: +91-9449223826

between 500 and 4000 cm^{-1} are due to internal vibration of the co-ordinated groups [4,5]. The band in the high-energy region between 3488 and 3186 cm^{-1} is due to the O–H vibrations of water, P–O–H group and N–H vibrations of ammonium. The peak at 1688 cm^{-1} is due to the bending vibration of water molecule. The peak at 1515 cm^{-1} is due to the bending vibration of ammonium [6]. Thus, as a result of irradiation, there is slight change in the chemical nature of the sample by the absorption of peaks due to the formation of free radicals and other absorption band appeared in the irradiated samples [7]. After 1 Mrad of total dose, the chemical nature of the ADP was unchanged up to a total dose of 20 Mrad.

3.2. AC Conductivity

The dielectric properties are related with the electric field distribution with in solid materials and it is one of the useful methods for characterization of

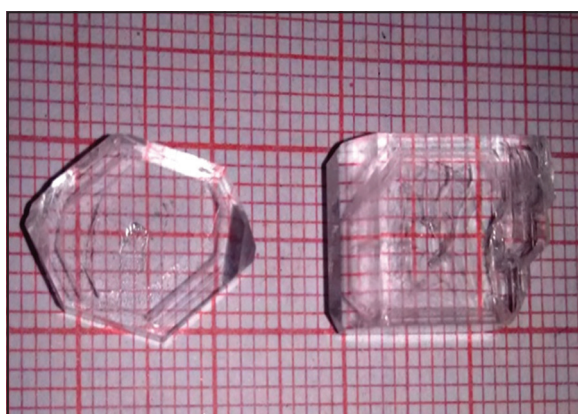


Figure 1: Photograph of as grown ammonium dihydrogen orthophosphate single crystals.

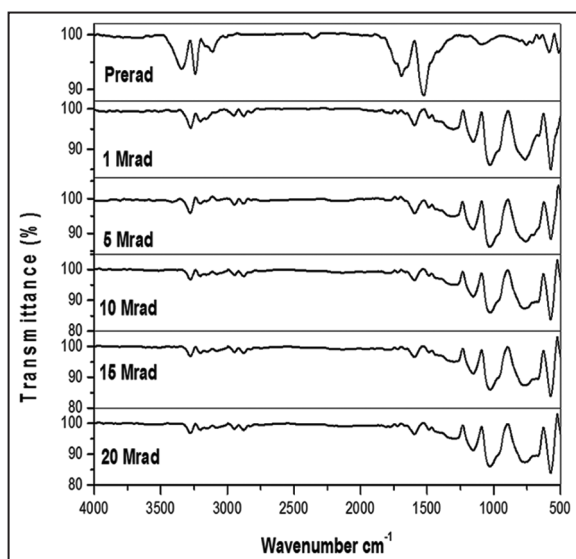


Figure 2: Fourier transform infrared spectra of unirradiated and irradiated ammonium dihydrogen orthophosphate crystal.

electrical response in crystalline material. The two opposite surfaces of the ADP crystal were polished and coated with air dry silver paste for the electrical measurements. The capacitance and dissipation factor of the unirradiated and irradiated single crystals were measured using HIOKI 3532-50 LCR Hitester in the frequency range of 100 Hz to 5 MHz. The dielectric constant ϵ_r has been calculated using the following equation as follows:

$$\epsilon_r = \frac{Ct}{A\epsilon_0} \tag{1}$$

Where, $\epsilon_0 = 8.854 \times 10^{-12}$ F/m is the permittivity of ϵ free space, t is the thickness of the crystal, C is the capacitance and A is the surface area of the crystal in contact with the electrode. Figure 3 shows the plot of dielectric constant against applied frequency for unirradiated and irradiated ADP crystal. It can be seen that the dielectric constant is found to decrease with increase in total dose up to 10 Mrad. After 10 Mrad of total dose, the dielectric constant is slightly decreased. The Co-60 gamma induced defects cause an increased space-charge contribution that increases the dielectric constant of the crystal.

The dielectric loss may be due to the perturbation of the phonon system by an electric field. The energy transferred to the phonons dissipates in the form of heat. From the Figure 4, it is observed that the dielectric loss is very high at low frequencies. Furthermore, the dielectric loss increases with increase of radiation dose up to 15 Mrad, and there is slightly decreased for a total dose of 20 Mrad. The response of AC conductivity with frequency ranging from 50 Hz to 5 MHz for unirradiated and Co-60 gamma irradiated ADP crystal is shown in Figure 5. It can be seen that the AC conductivity was found to increase with

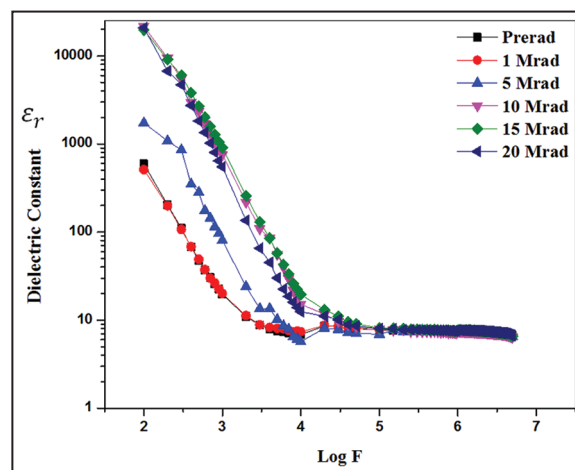


Figure 3: Log frequency versus dielectric constant for ammonium dihydrogen orthophosphate crystal.

increase in frequency and Co-60 gamma irradiation due to the defects created on irradiation [8].

3.3. DC Conductivity

The DC electrical conductivity study is essential to understand the behavior of charge carrier under DC field. The conductivity in ionic solid is mainly due to the presence of point defects in the lattice and due to different types of mobile charges as given in the relation;

$$\sigma = \sum_i m_i q_i e \mu_i \quad (2)$$

Where summation is taken over all the charged species i , m indicates the number of mobile charges of the type i having net charge $q_i e$ and μ_i represents the electrical mobility. The DC conductivity of unirradiated and irradiated crystal was calculated using the relation;

$$\sigma_{dc} = t/RA \quad (3)$$

Where, R is the measured resistance, A is the surface area of the crystal in contact with the electrode and t

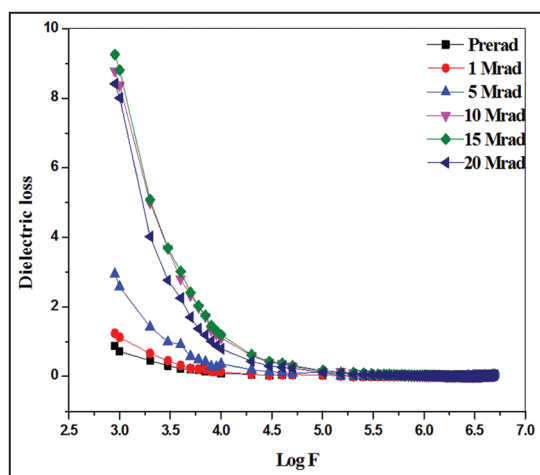


Figure 4: Log frequency versus dielectric loss for ammonium dihydrogen orthophosphate crystal.

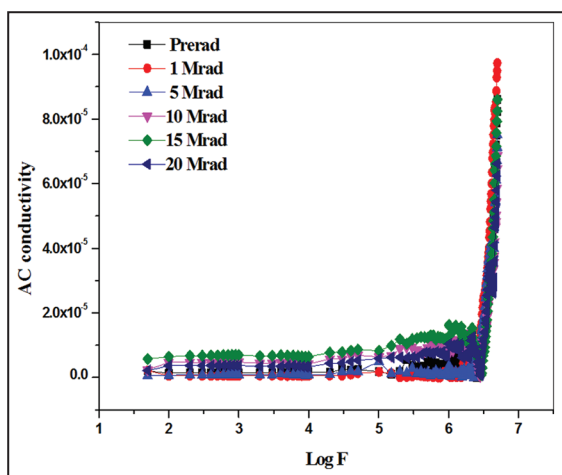


Figure 5: Log frequency versus AC conductivity.

is the thickness of the sample. The DC conductivity measurement was carried out using Keithley dual channel source meter model 2636A. I-V characteristic graph of unirradiated and irradiated ADP crystal is shown in Figure 6 and from which conductivity values are calculated. Figure 7 shows the variation of DC conductivity with radiation dose and it can be seen that the conductivity increases with increase in radiation dose up to 15 Mrad [9] due to gamma induced defects in the lattice. After 20 Mrad of total dose, conductivity is slightly decreased may be due to distortion in the hydrogen bond existing between the oxygen atoms of the adjacent PO_4 -group of ADP.

4. CONCLUSIONS

In this study, *in-situ* AC and DC measurements are carried out for Co-60 gamma irradiated ADP crystal up to a total dose of 20 Mrad. The dielectric constant, dielectric loss and AC conductivity were increased up to a total dose of 10 Mrad and there is slight decrease for 15 and 20 Mrad irradiated crystal. The

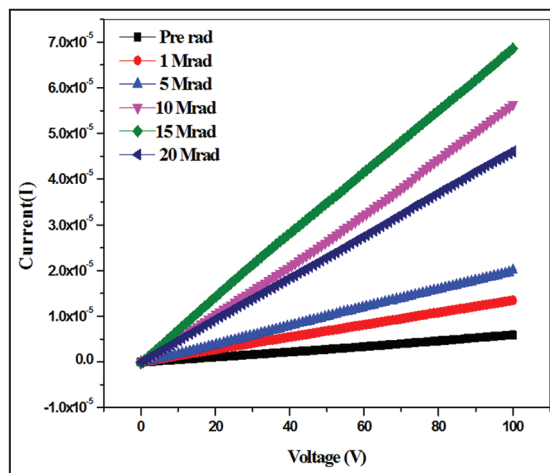


Figure 6: Plot of current versus voltage for ammonium dihydrogen orthophosphate crystal.

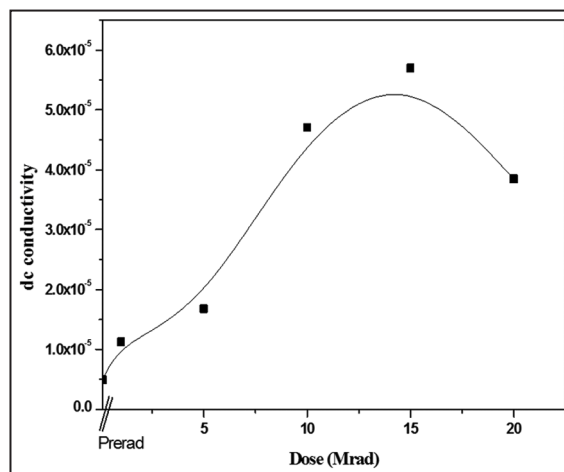


Figure 7: Variation of σ_{dc} of ammonium dihydrogen orthophosphate crystal with gamma radiation dose.

DC conductivity was found to increase with increase in total dose. FTIR spectral analysis shows there is a slight change in the absorption of bands up to 1 Mrad after that there is no change in the chemical properties up to 20 Mrad.

5. ACKNOWLEDGMENTS

This work is carried out under the research project sanctioned by UGC, Govt. of India [Project no. 41-911/2012 (SR)].

6. REFERENCES

1. A. Matkovski, A. Durygin, A. Suchocki, D. Sugak, G. Neuroth, F. Wallrafen, V. Grabovski, I. Solski, (1999) Photo and gamma induced color centers in the YAlO₃ and YAlO₃: Nd single crystals, *Optical Materials*, **12**: 75-81.
2. S. M. Kaczmarek, W. Żendzian, T. Łukasiewicz, K. Stępką, Z. Moroz, S. Warchoń, (1998) Effects of gamma irradiation and annealing treatments on the performance of Cr;Tm;Ho: YAG lasers, *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, **54**: 2109-2116.
3. U. Roth, M. Tröbs, T. Graf, J. E. Balmer, H. P. Weber, (2002) Proton and gamma radiation tests on nonlinear crystals, *Applied Optics*, **41**: 464-469.
4. N. Goel, N. Sinha, B. Kumar, (2013) Growth and properties of sodium tetraborate decahydrate single crystals, *Materials Research Bulletin*, **48**: 1632-1636.
5. M. Kalidasan, K. Asokan, K. Baskar, R. Dhanasekaran, (2015) Effect of gamma ray irradiation on sodium borate single crystals, *Radiation Physics and Chemistry*, **117**: 70-77.
6. P. Rajesh, P. Ramasamy, (2009) Growth of dl-malic acid-doped ammonium dihydrogen phosphate crystal and its characterization, *Journal Crystal Growth*, **311**: 3491-3497.
7. M. Ahlam, M. Ravishankar, N. Vijayan, G. Govindaraj, A. P. Prakash, (2012) Investigation of gamma radiation effect on chemical properties and surface morphology of some nonlinear optical (NLO) single crystals, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, **278**: 26-33.
8. T. Kanagasekaran, P. Mythili, P. Srinivasan, N. Vijayan, D. Kanjilal, R. Gopalakrishnan, P. Ramasamy, (2008) On the observation of physical, chemical, optical and thermal changes induced by 50 MeV silicon ion in benzimidazole single crystals, *Materials Research Bulletin*, **43**: 852-863.
9. S. C. Sabharwal, B. Ghosh, R. Y. Deshpande, (1976) Electrical conductivity of gamma irradiated ADP and KDP crystals, *Radiation Effects*, **30**: 123-124.

*Bibliographical Sketch



A.P. Gnana Prakash is an Associate Professor at Department of Studies in Physics, University of Mysore, Mysore, India. He completed MSc. and MPhil. in Solid State Physics from Gulbarga University, India and Ph.D. from Mangalore University, India. He worked as a Post-doctoral Fellow at Department of Physics, National Dong Hwa University, Taiwan and School of Electrical and Computer Engineering, Georgia Institute of Technology, USA. He has more than 13 years of research/teaching experience and published more than 70 research articles in reputed journals. His main research interests are growth and characterization of nonlinear optical crystals and radiation effects on semiconductor devices and circuits.