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Electronic Processes and Radioluminescence for Crystalline Systems with Different Structure Perfection

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The electronic processes causing the radioluminescence of organic molecular scintillators, i.e. single crystals, polycrystals, plastics, liquids, are discussed. Pure and binary systems were investigated. The analysis is based on the review of experimental results of radioluminescence intensity and pulse shape measurements, both well known and new, and on the modern theory of radioluminescence process in organic condensed media ^[1]. Specific quenching of the radioluminescence, which appears in the cases of high density of excitation, is analysed for organic solids with different structure perfection.

Keywords: radioluminescence; polaron states; molecular solids; liquids

INTRODUCTION

Scintillators are the objects in which the process of radioluminescence (RL) is very effective and take place during the time not longer then

microsecond. Organic scintillators possess their own place among the great amount of the objects, which are believed to organic molecular condensed media, i.e. organic crystals, polymers and liquid solutions of organic luminophors. Organic scintillators (like the effective inorganic ones) are dielectrics. Specificity of scintillation pulse formation is caused by the fact of involving in the process of very high energies exchange on it first step. Really, such energies are about $10^4 \dots 10^6$ eV, and the energies, which describe the process of excitation energy transfer, or even the ionisation potentials of molecules are the negligible values in such a scale. In other words, the features of an object structure means nothing on this first step of scintillation pulse formation, i.e. during the time of formation of regions of high activation density (RHD). During the times about 10^{-11} s these regions (spurs, tracks) are cooling down, and after that the physical processes of transport in them of charge states and excitation energy are defined by nature and features of a sample. Nevertheless, during the first step of energy exchange processes the proportions between the energies deposited inside and outside RHD are defined. It means that a specific quenching of RL, a proportion between it fast and slow component, and other main rules of RL pulse formation have just been defined. This paper is devoted to investigation of some aspects of relationship of influence of the first and last step of RL pulse formation on scintillation characteristics of a sample.

SAMPLES, EXPERIMENTAL DETAILS.

Single crystals of p-terphenyl with different structure perfection were obtained using the special technologies of their growth. Pure single crystals as well as doped with molecules of diphenylbutadiene (DBD) ones were studied. Polycrystals were obtained by pressure from grinding single crystals ^[2]. Results of measurements of RL pulse intensity and shape are used for analysis. Fig.1 shows the calculated

number N of molecules of DBD in the track volume for moment of excitation ($t=0$), and time limits of RL slow component analysis ($t=50$ ns and $T=2\mu$ s). The natural limits of technology give no possibility to verify the numbers of molecules of luminescent solvent as many as can be done by effect of track expanding.

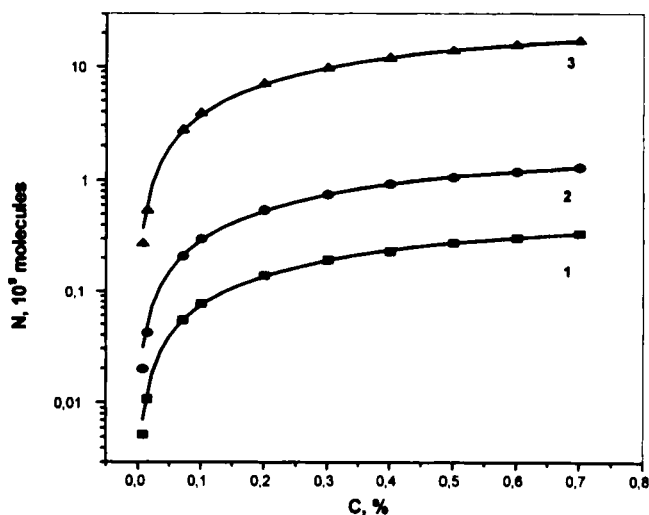


FIGURE 1 Number of DBD molecules N in track volume for different concentrations of DBD C in p-terphenyl polycrystal.

The values of

$$\xi_{ij} = [M/E]_i / [M/E]_j, \quad (1)$$

where E is the energy of ionising radiation and M is the number of scintillation photons in the RL pulse for the cases of excitation by radiation of i - and j - type, were calculated. The ξ - values are not

dependent on energy of ionising radiations, and therefore give a “pure” quieting effect for types of irradiation been studied. The $\xi_{\alpha\gamma}$ – value, i.e., gives the information about specific quieting for the case of α -excitation, because in the case of γ - excitation no track is formed. It was shown that for single crystals of p-terphenyl $\xi_{\alpha\gamma}$ is about 0.07 and for polycrystals is about 0.05...0.06, and slightly decrease with C increase. The light output for polycrystals was lower than for single crystals.

DISCUSSION

Two main points of this analysis have to be: the differences between pulse shapes and intensities of prompt radioluminescence for the objects with different structural perfection, and the same one differences for delayed radioluminescence as well as.

The prompt radioluminescence pulse is formed in the multistage process of ionising radiation energy transformation, i.e. hot secondary particles – charge carriers – molecular excitation – light photons emission, which takes place in the regions of low density of activation. For effective scintillators the stage of charge carrier generation, transport, and recombination is of a primary importance. As it was shown in previous work ^[3], the most probable explanation of fast component of scintillation pulse formation must based on the idea of molecular –lattice polaron theory ^[4]. The temperature dependence of mobility for molecular and lattice polaron are not only different but can “compensate” each other making the mobility of lattice – molecular polaron constant for wide temperature range ^[4]. Really, the well-known fact of negligible effect of temperature on radioluminescence light output for organic molecular solids in temperature range $-60 \dots +60^{\circ}\text{C}$ is the direct evidence of such an explanation of RL mechanism. This mechanism takes account of the generation of dynamic traps of polarization origin those can move through the organic system as molecular – lattice polaron states, and those recombination results in

excitation of the molecules in organic solids. According to such an explanation, when the tracks are overlap and mean distance decreases, the mean time needed for such a recombination becomes shorter. Delayed radioluminescence pulse is formed according to the same multistage process of the energy transformation of ionising radiation as in the case of prompt radioluminescence formation, but in RHD. In this case, the probability of exchange interactions between charge states is of primary importance. It results in generation ratio of singlet (S) and triplet (T) excitons 1:3 ^[1]. It causes the process of T-T-annihilation in such a region, the singlet channel of which results in generation of photons of the slow component of scintillation pulse. Fig.1 shows that the effect of tack expanding is more important for slow component formation than even such an important technical characteristic as the concentration of additional agents, which are the luminescent centres of crystal. Nevertheless, the results of ξ_{ay} - values measuring conform that a structure perfection of crystalline system can influence on it characteristics. As it shown in ^[5] the variation of the root-mean -square random orientation of mosaic structure of a single crystal from 26' to 160' results in decrease of slow component intensity more than by three times. These facts become obvious when we take into account that for the last stage of scintillation pulse formation the process of a T- exciton transport is of primary importance. Specific quenching of the RL for single crystals and polycrystals is different. The structure perfection of the object cannot effect on primary processes of excitation by ionising radiation, i.e. on the excitation with the energies of $10^5 \dots 10^6$ eV. Therefore, the difference in specific quenching of the radioluminescence for single crystals and polycrystals can only cause by the processes of charge carrier and molecular excitation trapping on deep traps of a structural origin.

CONCLUSIONS

In the process of RL pulse formation for crystalline organic molecular systems, the type of excitation plays the most important role, and defines the primary conditions of the process. On the second place is the influence of the object parameters, which define the conditions of exciton transport. Deviations from normal conditions of exciton transport, which are typical for this type of objects (e.g. variation in traps concentration), have a comparably small effect on RL pulse formation process.

Acknowledgments

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