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Nature of carrier traps in polyethylene and possible implication in aging process

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Résumé

Les charges d'espace sont impliquées dans les phénomènes de rupture et de vieillissement des isolants polyéthylènes dans lesquels la nature des sites de piégeage reste méconnue. Les techniques de luminescence peuvent contribuer à l'identification de ces sites ce qui permettrait le contrôle de la charge stockée. Le polymère considéré est un polyethylène basse densité sans additif ni anti-oxydant. La photoluminescence permet d'identifier les espèces émissives qui sont nécessairement des défauts ou impuretés chimiques. La luminescence induite par la recombinaison radiative de charges piégées conduit à l'identification des centres optiques électriquement actifs, donc des niveaux de piégeage. La nature chimique de ces centres permet d'envisager les mécanismes de vieillissement électrique sous un jour nouveau.

Abstract

Space charges are involved in electrical aging and breakdown of polyethylene-based materials, but the nature of the trapping sites is still not well known. Identification of trapping centers in these materials would allow to control the amount of trapped charges. Luminescence techniques provide a way to reach this objective. We worked on additive-free low density polyethylene without anti-oxidant. Photoluminescence experiments were carried out to identify the emitting species which are necessarily defects or chemical impurities. Charge recombination-induced luminescence allows to identify the electrically active luminescent centers which are therefore potential candidates as trapping centers. The chemical nature of these centers leads to envisage aging mechanisms under a new angle.

Introduction

Space charges in polyethylene-based materials is the subject of more and more concern. Their implication in electrical aging and breakdown has been recognized through numerous experiments [1]. They constitute a serious limitation in the use of synthetic insulation in high voltage direct current cables. One aspect of the problem is related with the bulk distribution of the charges which can change the field distribution in the insulation, leading to local stress enhancement. The net result is that the material can be submitted to local stress much higher than the nominal one. A better control of material charging is needed, and it could be achieved through material tailoring. However, this can only be done through the knowledge of the nature of the trapping centers which provide sites where electrical charges can be stabilized.

There is very scarce information available on the nature of the trapping centers in polyethylene and its derivatives (e.g. cross-linked polyethylene), mainly because the techniques that can be used to identify the nature of traps are not numerous. A widespread approach is to correlate electrical measurements (conduction current, thermally stimulated depolarisation current, space charge distribution) with a change in the chemistry (by introducing functional groups or additives) or the microstructure (by controlling molecular parameters). However, these techniques yield indirect information on the nature of traps and it is not straightforward to change the chemistry without modifying the structure of polymers. Luminescence techniques provide an alternative to electrical measurements for identifying the nature of traps through the analysis of the emission spectrum.

Polymer luminescence can be excited by various excitation sources, being either electrical or not in origin [2]. Among those, photon absorption, electric field, chemical reaction, mechanical stress, ionizing radiation, thermal energy, lead respectively to photo-, electro-, chemi-, tribo-, radio-, and thermo- luminescence. Note that this terminology is a generic one which makes reference to the experimental conditions under which luminescence is produced rather than to the emission process itself, e.g., electrical processes can be involved in triboluminescence experiments. Luminescence in the visible part of the optical spectrum corresponds to energy between 4.1 eV (3000 A) and 1.55 eV (8000 A), i.e. from half to a fraction of the forbidden gap (≈ 8 eV) of polyethylene materials. Any light emission in this range is therefore characteristic of electronic transitions taking place in the band gap, i.e., they are characteristic of localized levels, some of which being candidate as trapping levels.

In this paper, we discuss the nature of deep traps in additive-free low density polyethylene by using two luminescence techniques. In a first step, we apply photoluminescence to get the emission spectrum associated with electronic levels in the forbidden gap of the polymer. It provides the optical fingerprint of the material's chromophores. In a second step, we deposite