절연물 폴리머의 전하이동과 전계발광

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Charge transport and electroluminescence in insulating polymers

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Abstract : Polymers submitted to thermo/electrical stress suffer from ageing that can drastically affect their functional behaviour. Understanding the physico/chemical processes at play during ageing and defining transport regimes in which these mechanisms start to be critical is therefore a prime goal to prevent degradation and to develop new formulation or new materials with improved properties. It is thought that a way to define these critical regimes is to investigate under which conditions (in terms of stress parameters) light is generated in the material by electroluminescence (EL). This can happen through impact excitation/ionization involving hot carriers or upon bi-polar charge recombination (a definition that excludes light from partial discharges, which would sign an advanced stage in the degradation process). After a brief review of the EL phenomenology under DC, we introduce a numerical model of charge transport postulating a recombination controlled electroluminescence. The model output is critically evaluated with special emphasize on the comparison between simulated and experimental light emission. Finally, we comment some open questions and perspectives.

Key Words : Thermo/electrical stress, Physico/chemical processe, Degradation, Electroluminescence

1. Introduction

Basically, excess energy in insulating materials under thermo-electric stress can dissipate either thermally by emission of phonons and/or optically by emission of photons. Thermal dissipation due to ionic or electronic transport has a positive feedback on the current leading eventually to a thermal runaway when critical conditions with an unbalanced heat supply are reached. This has been discussed at length in textbooks (see for example [1]) and experimental evidence of thermal breakdown has been provided [2]. Thermal runaway however did not give a key to understand electrical ageing, especially in the case of low loss dielectrics like polyethylenebased material. For those, current flow is so low that the elevation of temperature due to conduction is usually negligible. Alternatively, excess energy can dissipate - eventually optically - in a range that corresponds to covalent bond energy that is a few eV. We will concentrate on these optical phenomena that are considered as an ageing indicator [3]. Although EL can be excited in insulating materials by any kind of voltage shape of sufficient magnitude, we will restrict our presentation and discussion to the DC case for the sake of simplicity, taken polyethylene as a case study.

The theory of electroluminescence in large band gap materials predicts different excitation mechanisms depending on the type and distribution of carriers inside the dielectric. Light can be emitted upon inelastic interactions between mobile carriers (so-called hot carriers with kinetic energy >> thermal energy) and molecules constituting the dielectric. Although structural and chemical changes induced by hot electrons have often been evoked in electrical ageing [4, 5], a clear evidence of their existence in polymeric insulation is still lacking. The other excitation mechanism of EL is encountered when bipolar charge domains spatially interact, leading eventually to radiative charge recombination. This can happen for example when charge promotion occurs via injection from the electrodes, followed by charge transport in the bulk.

2. Model hypothesis

The model features injection, transport, trapping and recombination of electrical charges. Each kind of carriers can be mobile or trapped. Mobile carriers have a constant effective mobility (describing conduction through shallow levels, so called physical traps, related to the disordered structure). Alternatively, a description using a hopping mobility can be adopted but it does not affect deeply the simulation results. Deep trapping is described by considering a single level of deep trap (so-called chemical traps) for each type of carrier from where charges can escape by thermal activation. Recombination processes between mobile and/or trapped carriers are taken into account. By integrating in space the contribution from all the transitions, one gets the recombination rate that can be directly compared to the experimental EL. The model is clearly not taking into account hot carrier effects as a possible excitation mechanism for the EL. Further details on the model can be found in [6].

3. Results and Discusion

Figure 3 shows the recombination rate of mobile electrons with trapped holes as a function of the spatial coordinate x, computed for a field of 60 kV/mm without initial charge density. For short stressing time (<130 s), the recombination rate is null, just because negative and positive carriers do not coexist. Beyond that time, the recombination rate passes through a maximum (t=150 s) and continuously decreases afterwards. The peak corresponding to the maximum recombination rate (for mobile electrons) progressively moves towards the anode for times in the range 130-200 s. A symmetrical situation holds for recombination of mobile holes with trapped electrons in the cathode vicinity. Taken all together, the model forecasts a change in the location of EL during stressing, from the middle of the sample to the regions near both electrodes. Of course, the EL measurement is a space-integrated quantity and the location of the emission in the depth cannot be checked experimentally.

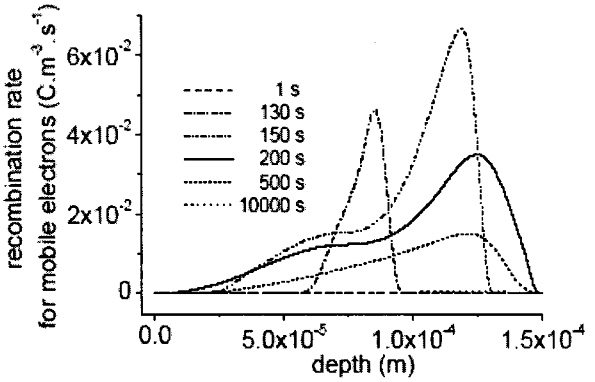

Fig. 1. Recombination rate profiles for mobile electrons/trapped holes as a function of time (cathode on the left, anode on the right, 60 kV/mm field, zero initial charge density). A symmetrical figure (recombination close to the cathode) holds for mobile holes/trapped electrons.

4. Conclusions

By considering experiments and modelling, we have shown that much of the EL features observed in PEbased materials can be interpreted on the basis of a recombination-controlled EL model. Involvement of hot carriers in the excitation mechanisms is believed to be important under time-varying voltage conditions where the model fails to reproduce the experimental behaviour.

Acknowledgement

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