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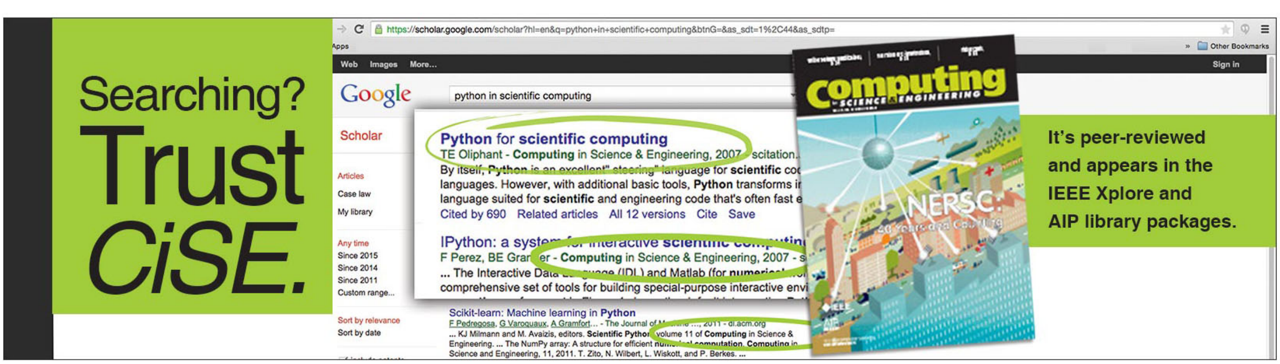

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Trap characterization in composite of solid-liquid using dual-level trap model and TSDC method

Yanhui Wei,[1,2] George Chen,[2] Guan-Jun Zhang,[1,a] Ning Liu,[2] and Guochang Li[1,2]
[1]*State Key Laboratory of Electrical Insulation and Power Equipment, Xi'an Jiaotong University, Xi'an, 710049, China*
[2]*School of Electronics and Computer Science, University of Southampton, Southampton, SO17 1BJ, United Kingdom*

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Charge trap is considered to be one of the effective characteristic parameters for qualitatively evaluating the aging status of insulating material. In this paper, the trap characteristics in oil-impregnated paper with different aging types (non-treatment, thermal treatment and electrical treatment) are investigated using a dual-level (shallow and deep energy) trap model based on space charge profiles and thermally stimulated depolarization current (TSDC) data. The simulated results based on the model are well consistent with the experimental results. On the other hand, the TSDC method can acquire much information related to the shallower traps, and the dual–level trap model can obtain much charge dynamics characteristics. It has been observed that thermally aging makes the shallow trap energy become deeper while electrically aging makes it shallower. Moreover, the trap density in oil-impregnated paper increases after aging regardless of thermal or electrical aging. [http://dx.doi.org/10.1063/1.4960214]

I. INTRODUCTION

Oil-impregnated paper has been widely used in high voltage transformers as it has shown excellent electrical and mechanical performance.[1] However, it gradually becomes aged when subjected to thermal, electrical or other stresses over its operation period. During the process of oil-impregnated paper deterioration, the formation of space charge may further promote its degradation and aging.[2–8]

Many researchers have put efforts on developing aging models of polymers based on trapped charge characteristics. For example, Dissado *et al*. established a model from the viewpoint of physics. It describes the relationship between insulation life and trapped charges, showing that trapped charge could accelerate the aging process and shorten the lifetime.[3,9] Cavallini *et al*. proposed an empirical model, which depicts the insulation life under the effect of trapped charges.[10] These models are all supportive to the statement that space charge and trap could be both effects and causes of aging. Moreover, the aging in physical and chemical property will correspond to the change of its microstructure, which is closely related to trap parameters.[6,11,12] Therefore, the trap parameters (e. g. trap energy and trap density) are considered to be employed to qualitatively assess aging state of the material. In recent years, trap parameters of polymers have been evaluated.[11,13,14] Specially, Chen proposed a dual-level trap model based on space charge characteristics in polymers.[11,12] It describes the aging characteristics in polymer by dual levels of traps, which are termed as shallow trap and deep trap. At present, most works about trap parameters focus on polymers, and rarely about composite of solid-liquid, especially for oil-impregnated paper.

[a]Electronic mail: gjzhang@xjtu.edu.cn.

In our previous research,[14] we have experimentally investigated the trap parameters of oil-impregnated paper via thermally stimulated depolarization current (TSDC) method. In this paper, a dual-level trap model is employed for oil-impregnated paper. Trap parameters of oil-impregnated papers with different aging sources, i.e. thermal and electrical, are calculated. In addition, the simulated results are compared with the experimental results obtained by TSDC method. Finally, trap characteristics in oil-impregnated paper with different aging types are investigated based on the model simulation and experimental results, and the aging types are qualitatively identified, such as electrical aging and thermal aging.

II. IMPROVED DUAL-LEVEL TRAP MODEL BASED ON SPACE CHARGE

According to our previous work,[6,12] in order to simplify the mathematic calculations, it was assumed that there are only dual-level traps within oil-impregnated paper and these two level traps are uniformly distributed across the sample. The improved dual-level traps model can be described as following based on space charge dynamics.

The charges are injected from the electrodes based on the Schottky law.[15] The injection current density can be expressed as

$$J = A_0 T^2 \exp(-\frac{qw}{kT}) \exp(\frac{q}{kT}\sqrt{\frac{qE}{4\pi\varepsilon_0\varepsilon_r}}) \tag{1}$$

Where w is the injection barrier, eV; A_0 is the Richardson constant; T is the absolute temperature, K; k is the Boltzmann constant; E is the applied electric field, V/m; ε_0 is the vacuum permittivity, F/m; ε_{r} is the relative permittivity of material; q is the elementary electron, C.

The dynamic processes of the injected charges and conduction charges in material can be described as

$$\frac{dn}{dt} = \frac{J}{qd} - Pn_m \tag{2}$$

Where n is the net charges density, $\mathrm{C/m^3}$, it refers to the sum of mobile charges n_m and trapped charges n_t. d is the deep of injected charges, μm; P is the charge conduction coefficient, s^{-1}; n_m is the mobile charges, $\mathrm{C/m^3}$.

In the model, the dynamic processes of trapping and de-trapping are considered. In order to simple the mathematics process, the charges movement process between the shallow traps and deep traps in the material is presumed to be negligible. The changing rate of trapped charge density can be expressed as

$$\frac{dn_t}{dt} = R_{cap} - R_{esc} - R_{rec} \tag{3}$$

It comprises of three terms, charge capturing rate by traps R_{cap}, charge escaping rate from traps R_{esp} and the recombination rate of trapped charges R_{rec}. Specially, these three terms can be expressed as following.

$$R_{cap} = n_m(N_t - n_{t1} - n_{t2})S \tag{4}$$

where N_t is the total traps density, $\mathrm{C/m^3}$; N_{t1} and N_{t2} are the total density of shallow and deep traps, $\mathrm{C/m^3}$; n_{t1} and n_{t2} are the occupied density of shallow and deep traps, $\mathrm{C/m^3}$, respectively; S is the trapping cross sectional area, $\mathrm{m^2}$. The physical process can be described as follows: when the mobile charges n_m move between the two trap sites with unit speed within unit time, these charges passing through unfilled traps of number density $(N_t - n_{t1} - n_{t2})$ with a capture cross section of S are captured and the capturing rate is given by equation (4).

$$R_{esc} = n_t v_0 \exp\left(-\frac{E_t{}'}{kT}\right) \tag{5}$$

In equation (5), n_t is the total charges captured by all traps, v_0 is the escape attempt frequency, s^{-1}. $E_t{}'$ is the trap depth affected by strong electric field based on Poole-Frenkel effect and it is shown by equation (6).

$$E_t' = E_t - \Delta V_{pf} \tag{6}$$

In equation (6), E_t is the initial trap depth and ΔV_{pf} is the lowered trap depth with Poole-Frenkel effect.

$$R_{rec} = Bn_t \tag{7}$$

where B is the recombination rate of charges with opposite polarity.

Substituting these three terms into equation (3), the changing rate of trapped charges can be expressed as

$$\frac{\mathrm{d}n_t}{dt} = -n_t \exp\left(-\frac{E_t'}{kT}\right) + n_m\left(N_t - n_{t1} - n_{t2}\right) S - Bn_t \tag{8}$$

The Schottky injection at the metal-insulator interface can be neglected after removing the applied voltage. According to equation (8), the changing rate of charges captured by shallow traps can be expressed as

$$\frac{\mathrm{d}n_{t_1}}{dt} = -n_{t_1} v_0 \exp\left(-\frac{k_1 E_{t_1} - \Delta V_{pf1}}{kT}\right) + n_m\left(N_{t_1} - n_{t_1}\right) S_1 - Bn_{t_1} \tag{9}$$

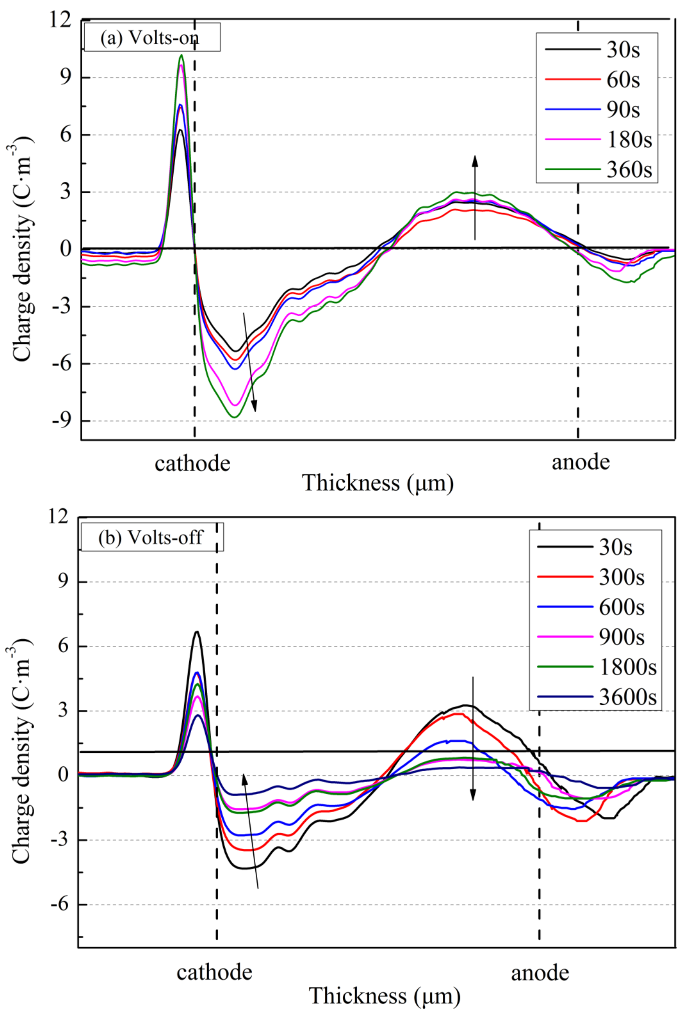

FIG. 1. Space charge behavior during volts-on within 360 s under 3.2×10^7 V/m and volts-off within 3600 s.

And the changing rate of charges captured by deep traps can be expressed as

$$\frac{\mathrm{d}n_{t_2}}{dt} = -n_{t_2}v_0 \exp\left(-\frac{k_2 E_{t_2} - \Delta V_{pf2}}{kT}\right) + n_m\left(N_{t_2} - n_{t_2}\right) S_2 - B n_{t_2} \tag{10}$$

It should be noteworthy that different materials have different trap distribution, so the modified parameters k_1 and k_2 are introduced, which represent the modified trap parameter, respectively. In this paper, the range of these parameters lies in between 1 and 3 based on space charge measurement within oil-impregnated paper.

In order to extract trap parameters based on the model, the space charge distributions during volts-on and volts-off are measured for the new sample, electrical aged sample and thermal aged sample, respectively. Here take the new sample for an example, Fig. 1 shows space charge profile of a new sample measured by the pulse electric acoustic (PEA) method. The thickness of sample is 130 μm, and the time of volts-on and volts-off are 360 s and 3600 s, respectively. In order to eliminate the effect of capacitive charges, the profile of space charge is processed with subtraction algorithm.[12]

According to the space charge distribution, the total quantity of charges in the sample can be calculated by equation (11).

$$Q_t = \int_0^d |\rho(x)|\, A\, \mathbf{d}\, x \tag{11}$$

where d is the thickness of the sample, A is the electrode area and $\rho(x)$ is the space charge density obtained by PEA method. Afterwards, charge density can be calculated according to the equation (12).

$$n = \frac{Q_t}{d \cdot A} \tag{12}$$

From Fig. 1, it can be observed that there are obvious bipolar charge injections at the two electrodes, and the electrons injected from the cathode is remarkably more than holes injected from the anode. After removing the applied voltage, the space charge decays sharply over the initial 300 s and then decays slowly, which indicates that there exist shallow and deep traps in the bulk of oil-impregnated paper.[7]

III. EXPERIMENTAL AND SIMULATION RESULTS

A. New oil-impregnated paper

In this paper, the charge conduction coefficient P, injection barrier w and trapping parameters have been set as unknown model parameters, which can be obtained by fitting the experimental data in Fig. 1 based on finite difference method (FDM), as shown in Table I. Fig. 2 depicts the charge density measured by the PEA and the simulated curves of mobile charges, trapped charges and net charges of new oil-impregnated paper based on dual-level trap model.

It can be observed that the trapped charges in shallow traps are much less than those in deep traps, and the mobile charges rapidly decrease once the removal of the applied voltage. Meanwhile, the shallowly trapped charges reduce to nearly zero with the decay time. However, the trapped charges in deep traps almost keep stable after about 1800 s, which illustrates that the charges

TABLE I. The key parameters and extracted trap parameters of new oil-impregnated paper based on the dual-level traps model.

Parameters	Value	Shallow trap	Value	Deep trap	Value
P (s^{-1})	0.007	k_1	2.7	k_2	1.2
w (eV)	1.123	E_{t1} (eV)	0.53	E_{t2} (eV)	0.87
		N_{t1} ($C \cdot m^{-3}$)	4.31×10^{15}	N_{t2} ($C \cdot m^{-3}$)	6.39×10^{23}

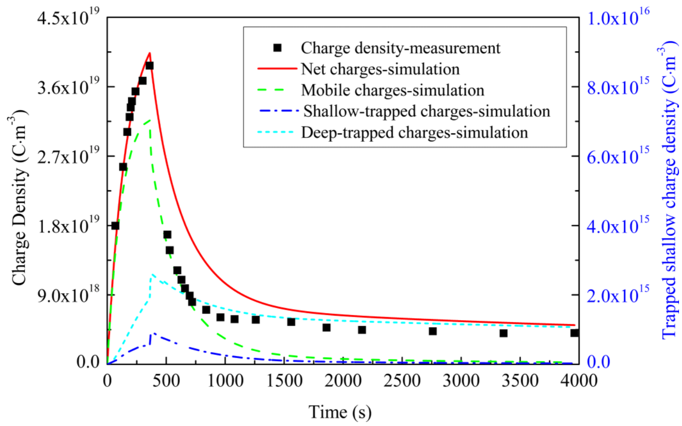

FIG. 2. Charge dynamics of new oil-impregnated paper.

captured by the deep traps need a considerably long time to dissipate. Hence, the ultimate net charges within oil-impregnated paper are mainly deeply trapped charges.

Trap parameters such as trap energy and trap density, may vary for the sample as the material experiences different aging sources such as thermal, electrical and the others. Any source of aging can cause the changes in the microstructure of a material, and trap characteristics are closely related to the microstructure of the material. In order to simulate the different aging sources, the electrical aging stress and thermal aging stress are implemented in oil-impregnated paper, respectively.

B. Electrically aged oil-impregnated paper

The samples treated by the electrical stress were obtained by cavity discharge with 2 times PDIV (Partial Discharge Inception Voltage) for 5 hours.[14] And then, the space charge distribution under two cases of volts-on and volts-off were measured, respectively. Afterwards, the simulated curves of mobile charges, trapped charges and net charges can be obtained based on the dual-level trap model using the same fitting algorithm, as shown in Fig. 3.

After electrical aging, both shallowly trapped charges and deeply trapped charges show an increasing trend comparing with the new sample. Within 1800s after removing voltage, the changes rate of new sample and electrical aged sample are about 2.43×10^{15} C/(m^3·s) and 3.47×10^{18} C/(m^3·s), respectively. It can be found that the decay rate of trapped charges become faster than that of the new sample. It illustrates much shallower traps may be generated. The main reason is that the surface of the sample is bombarded by charged particles during discharge and thus suffers mechanical damage.

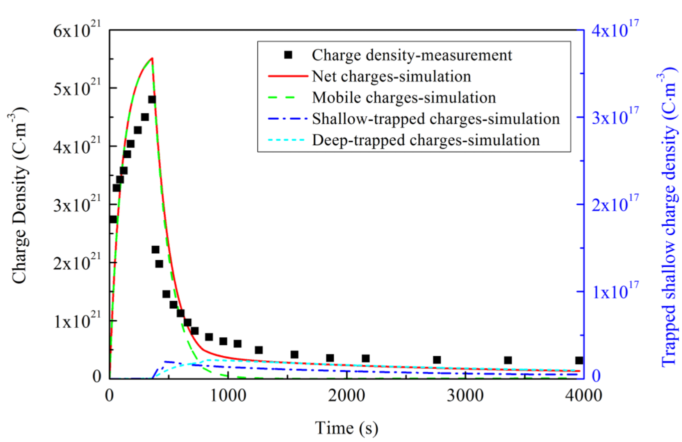

FIG. 3. Charge dynamics of electrical aged oil-impregnated paper.

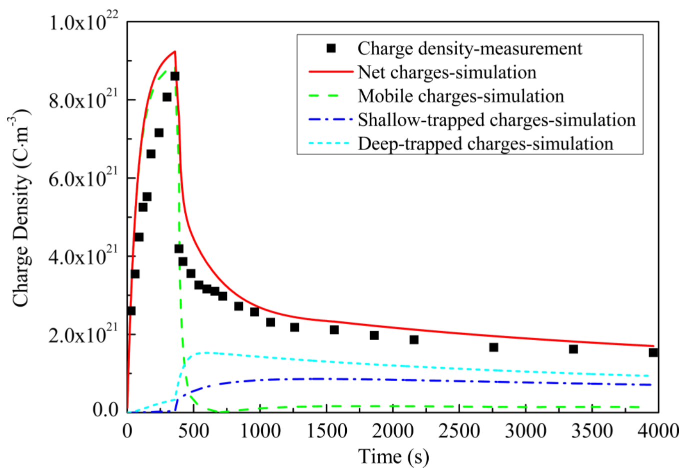

FIG. 4. Charge distribution of thermal aged oil-impregnated paper.

The defects and increased carbonization are generated, such as pits or pin-holes in the sample. These changes are mainly physical defects and result in much shallower traps.[16]

C. Thermally aged oil-impregnated paper

New oil-impregnated papers were thermally aged at 130°C for ~70h[17] and the degree of polymerization is 811. After that, the space charge profiles were measured during volts-on and volts-off period, respectively. Adopting the same calculation method, the distributions of mobile charges, trapped charges and net charges can be obtained, as shown in Fig. 4.

There has an interesting phenomenon that the shallow trapped charge density is no tend to zero over the time, as shown in Figs. 2 and 3. The net charge density is the sum of deep- and shallow-trapped charge density. Overall, the trapped charges are much more than that of the new sample and electrically aged sample. It implies that part of shallow traps get deeper after thermal aging. The most likely reason is that the fiber in oil-impregnated paper becomes loose after thermal aging and the inter-unit linkages in cellulosic chains are hence weakened. The changes are caused by chemical defects, which is also related to deep traps.[16]

In addition, Figs. 2, 3, and 4 illustrate that the net charges are almost equivalent to deeply trapped charges. It also indicates the deep traps dominate the aging process during long time period.

IV. COMPARISON AND DISCUSSION

In order to validate the applicability of the improved model in the oil-impregnated paper, trap parameters calculated based on the model are compared with experimental results measured by TSDC method. For three types of samples, i.e., new sample, electrically aged sample and thermally aged sample, the trap energy and trap density were obtained by TSDC method in our previous work.[14]

For the new sample, electrically aged sample and thermally aged sample, the comparison results are shown in Table II. Overall, the simulated results based on the model are well consistent with the experimental results, especially for deep traps. But for shallow traps, the trap energy obtained by the model is larger than that by TSDC method while the trap density is smaller. The charges captured by shallow traps will escape from trap sites within extremely short time, so that it is difficult to measure it by the PEA method.[18] Therefore, the trap energy simulated by the model based on space charge is larger than that of TSDC method. While the trap density is smaller than that of the measured results, due to part of shallow trapped charges have not been measured. For deeply trapped charges, the residence time is so long that almost all charges can be measured. The reason can be responsible for the well consistence of deep trap parameters between the two methods.

TABLE II. The comparison of model based simulation and TSDC method of oil-impregnated paper.

Aging types		Traps	Model results	TSDC results
New	Shallow	(eV)/ $(C \cdot m^{-3})$	0.53 4.3×10^{15}	0.34 1.1×10^{16}
	Deep	(eV)/ $(C \cdot m^{-3})$	0.87 6.3×10^{23}	0.82 5.3×10^{23}
Electrically aging	Shallow	(eV)/ $(C \cdot m^{-3})$	0.39 8.0×10^{17}	0.27 4.7×10^{18}
	Deep	(eV)/ $(C \cdot m^{-3})$	0.67 6.9×10^{23}	0.67 8.5×10^{23}
Thermally aging	Shallow	(eV)/ $(C \cdot m^{-3})$	0.60 1.3×10^{21}	0.51 7.1×10^{17}
	Deep	(eV)/ $(C \cdot m^{-3})$	0.80 8.9×10^{23}	0.76 8.6×10^{23}

The shallow trap of thermally aged sample is gradually transferred to deep trap. It presents a slight decay over the time and reaches a stable value, rather than tend to zero. It can be explained that thermal stress make the shallow trap energy deeper and less space charges escape from trap sites after removing applied voltage, result in some space charges captured by traps are still exist over the decay time.

The electrical and thermal aging can bring different physical and chemical defects to the oil-impregnated paper. The variation of physical and chemical defects will alter the ability of capturing charges of the material,[19] which can be described by the trap energy and trap density. Therefore, the parameters extracted from trap characterization are expected to assess aging types or aging status.

The comparison of two methods is shown as following: The TSDC method can measure charges escaped from trap sites at the moment of applied voltage removal. So it can acquire much information of shallower traps. By contrast, the dual-level trap model cannot obtain much shallower traps due to the measurement decay of space charge. However, the advantage of dual-level trap model is that the method can obtain much more charge dynamics information, i.e. the behaviors of mobile charges, trapped charges and net charges. Besides, it can directly reflect the effect of aging process on space charge dynamics, which is helpful to explore the aging mechanism.

V. CONCLUSION

An improved dual-level trap model based on space charge has been applied to the oil-impregnated paper. The simulated results based on the model are well consistent with TSDC measurement results.

(1) The comparison of the TSDC measurement and the dual-level traps model. The former can acquire much information related to the shallower traps. The latter can obtain much more charge dynamics, and it can directly reflect the effect of the influence of aging process on space charge dynamics, which is beneficial to better understand the aging mechanism.

(2) The comparison of aging types. The trap density and trap energy are closely related to the physical and chemical defects brought by different aging sources, which will alter the ability of capturing charges. Specially, the thermal aging makes the shallow trap energy become deeper while electrical aging makes it shallower. Trap density will increase after aging regardless of electrical or thermal aging.

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