SPACE CHARGE PHENOMENA INTERPRETATION TOWARDS ACCOUNTING FOR MATERIAL STRUCTURE OR QUALITY OF THE APPLIED TECHNOLOGY

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Abstract - *Focused on the interpretation of space charge evolution in paper materials, the article tries to identify some parameters derived from PEA measurements, able to be correlated with the chemical structure of these peculiar insulating materials (herein the mineral content), as a first step towards quality analysis of the manufacturing process.*

Keywords - Space charges, PEA method, Materials quality

1. INTRODUCTION

In recent years many studies have been performed regarding space charge phenomena inside or on the surface of certain materials, taking the aim of potential applications of some novel materials, especially in some fields not peculiarly / or not traditionally connected with the dielectrics area of interests, such as pharmaceutical, food industry or biophysical appliances, [1]. And this kind of studies seems to be further very actual and important for several reasons:
- the electric field can be applied towards simulating a response through which a better understanding of the material structure and behaviour can be gained (mainly regarding interfacial structure and technological electro-rheology);
- some systems are detrimentally affected by electric fields; such systems are usually sophisticated and require elaborate techniques to ensure or test their reliability over tens of years of exploitation under adverse conditions;
- many materials display useful responses to electrical fields, with peculiar reference on quality assessment; consequently, space charge measurement methods may work as an intelligent signal processing techniques, even for (bio-) electro-acoustic signals;
- current (and future) investigations in building up and movement of electrical charges in solid/soft condensed matter, followed by sophisticated 3-D computer modelling techniques, are a brief example in the field of developing novel electro-technologies, which include automatically the quality assessment procedures for the new obtained materials.

2. EXPERIMENTAL

The measurement principle is presented below, i.e. PEA - pulsed electroacoustic method, Fig.1-3, [1]. When a pulsed electric field is applied to a sample including trapped or induced charges, these charges generate acoustic waves, which propagate in the sample.

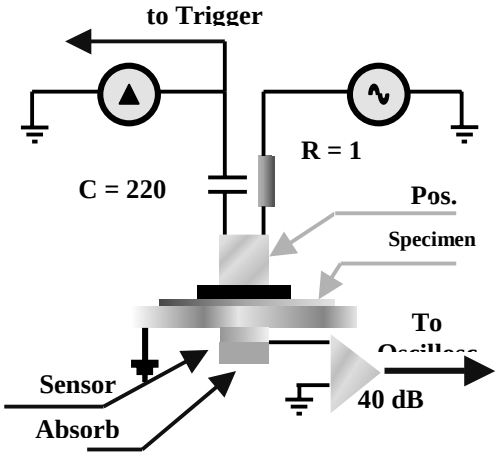

Fig.1 – Principle of PEA method and set-up

In fact, the dielectric specimen is submitted to a special cell – a peculiarly balanced acoustic system, composed by two electrodes, a conducting acoustic layer, a piezoelectric transducer and an acoustic absorber. A pulse source (pulse voltage up to 2kV and pulse width up to 200ns) and a HVDC source (up to 40kV) are coupled in parallel across the upper electrode, by a coupling capacitor, and, respectively, by a protecting resistor. The effect of the pulse is to generate pressure waves travelling through the sample in two directions. Reflections and transmissions occur normally at every (acoustic) interface. Hereafter, the acoustic wave is accurately detected by the transducer, as long as the acoustic absorber is able to delay and suppress the occurring reflections. The signal is further amplified by a broadband amplifier and displayed by a performance oscilloscope, Fig.1. Basically, the amplitude of the signal is related to charge quantity and the delay indicates the distance from the electrode, but a lot of other useful PEA derived parameters can be identified and investigated separately, [2,3]. In this way, the internal space charge distribution can be observed and processed by specialized software. Hence, the effect of applying the dc voltage lies in charge injection, accumulation

ISBN 972-98115-4-7 © 2001 IT

and/or drift within dielectric sample, and the charge dynamic can be accurately characterized by estimating the PEA quantities.

A drop of silicone oil is used to form a film between the specimen and the aluminium plate, in order to ensure a good acoustic coupling. The system resolution is better than 0.5 $C.m^{-3}$ in charge density and 4 μm or less spatially, depending on specimen structure and according to the experimental purpose. In order to observe the influence of the different electrode materials, stresses of equal and opposite polarities might be used in most cases. According to the scientific purpose and/or to the software features regarding response interpreting and calibration, this information can be specially outlined, or contrarily, suppressed.

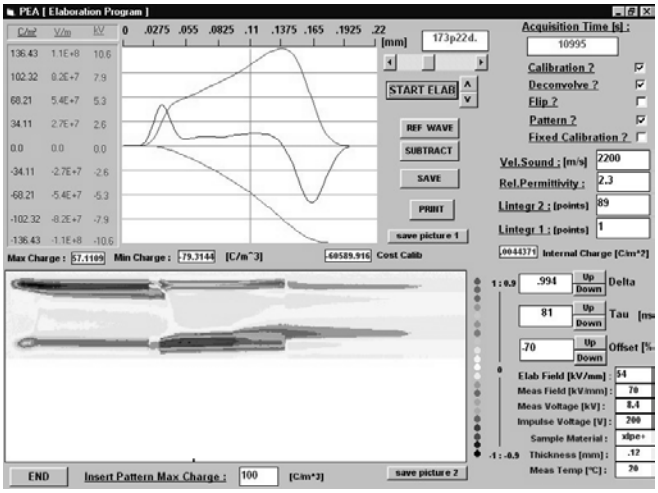

Fig.2 – Example of processing software for PEA-derived data.

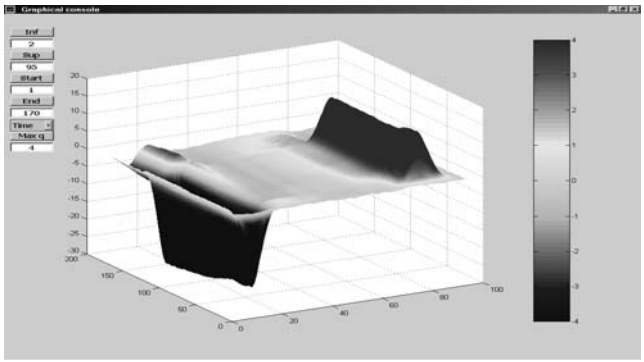

Fig.3 – Example of a space charge feature obtained by PEA method.

The software may be developed under Matlab, LabVIEW and/or C/C++ and must permit an extended signal interpreting via FFT and convolution techniques, being adaptable with the frequency response of any commercial transducer/amplifier combination. By means of the software, even the normal attenuation effect consequent to sample structure and/or technology should be compensate too, if necessary, [3].

Towards quality assessment, the continuous adaptation of the software becomes a must, i.e. by adding optional facilities involved by the study of materials with different (often unpredictable, or time variable) values of permitivity, conductivity and pulse speed. Some comparative measurements must be performed upon different liquid-solid mixtures, with the aim of identifying and measuring the peculiar values of PEA-derived parameters. This operation must presume the conceiving of totally different software for data acquisition, and a separate modulus for data processing. For on-line measurements, a new conception of PEA - virtual instrumentation (e.g. under LabVIEW) can be developed, by keeping only the main part of the calibration principle from the previous software. Such an example is presented in Fig.2, allowing the programming of facilities such as: trigger, input and reference parameters, characteristic data of material, temperature, time design of experiment, voltage design of experiment etc.

For laboratory purpose, the data processing should be used mainly off-line, to allow statistical interpretation, theoretical model testing, continuous re-calibration/deconvolving, and finally, the accurate identification of PEA-derived parameters. The analysis of charge evolution versus time, voltage or temperature is normally a must towards a complete estimation of the space charge feature, as presented in Fig.3.

As long as between the material's space charge behavior and the applied technology (described by the influence of some chemical/physical properties) was emphasized a clear correlation, we could finally account for the material structure, or for the quality of an applied technology, by use of well-defined values of some PEA derived parameters.

In order to create a dedicated PEA procedure for quality assessment, some extra technical circumstance should be met:

- to ensure that enough valuable and accurate information can be obtained during short time PEA measurements;
- to ensure a direct interpretation of the parameters values, preferably without further data processing or re-evaluation.

3. EXAMPLE AND DISCUSSION

Our application refers to papers for technical purposes, from sulphate hardwood – demineralized pulps. The mineral content in paper, a compulsory parameter for some dedicated appliances (e.g. cable insulation) can be controlled by use of a specific technological stage, named "demineralizing process". The demineralizing process parameters depend on cellulose base type, being in principal: the duration, and the equivalent of the reactive acid per 1 tone of absolutely dry cellulose. In our study, a constant equivalent of 60kg concentrated HCl per 1 tone of absolutely dry cellulose was used, for a variable treatment duration up to 70min, followed by a wash treatment with de-ionized water. The demineralizing process domain and mineral content limits are presented in Fig.4. Accordingly, 8 reference values of mineral content were appointed, in order to be correlated with the PEA derived parameters.

Normally, the polarization duration related to PEA method may presume even 10,000s, a very long time for quality assessment. But, as noted in [4] by analyzing the evolution of PEA derived parameters versus poling time, the internal charge reaches practically the regime (saturation) value after about 500s of the polarization process. More than this, the peculiar evolution of internal field suggests that the internal field is mostly established after 500s, even if the charge injection, extraction and transport processes may continue till the final equilibrium of charge is reached. On the other side, the charge characteristic obtained at 500s seems to reveal the highest magnitude of both positive and negative internal charges, representing i.e. the magnitude limit of the heterocharge process in insulating paper. Afterwards, the evolution towards regime conditions of charge is based mainly on charge extraction and recombination.

Hence, a reduced poling time t_P = 500s could be accepted for quick qualitative or comparative evaluations of different (paper) materials, especially when they are related technologically. This technical observation is in concordance with the imposed condition mentioned above for quality assessment, i.e. short time PEA measurements.

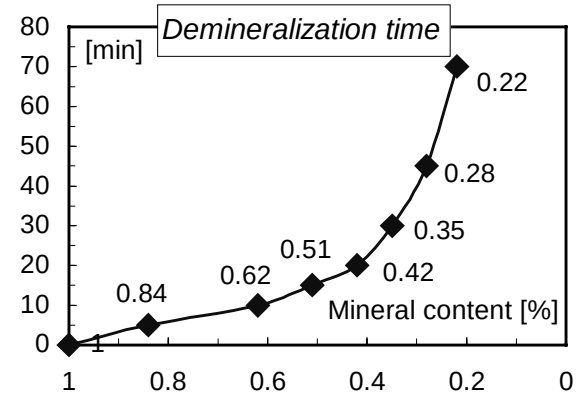

Fig.4 – Demineralizing process domain and parameters.

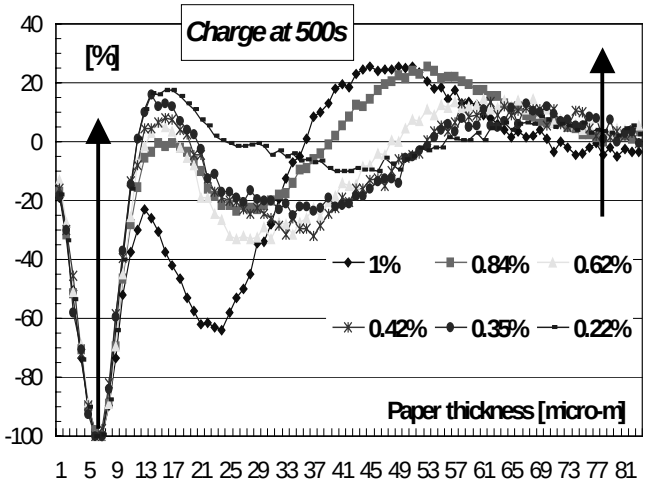

Fig.5 – Space charge profiles at different mineral content.

Consequently, the PEA measurements here presented were carried out upon oil-wetted and conditioned paper specimens, 70μm thick (having the mineral content related to the 8 reference values presented above), according to a polarization/depolarization program involving a poling field of about 80% of specimens breakdown strength, a poling time t_P = 500s and a depolarization time t_o = 100s.

The evolutions of space charge and internal field are presented in Figs. 5 and 6, as function of specimen thickness, at 8kV/mm poling field, for different mineral content (electrodes spatial positions are marked by arrows, and image charges within electrodes, of about 7 micro-m in exterior of specimen, are included). The characteristics are presented in [%], by reporting all instant values to the minimum value of the series, in order to facilitate the general interpretation of charge processes versus mineral content and to emphasize the relative regional growth of internal charge and field.

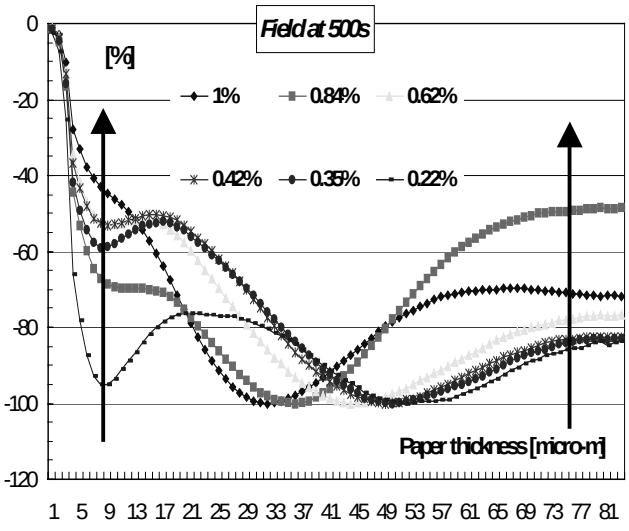

Fig.6 – Internal field profiles at different mineral content.

In our study, two reference limits of the demineralizing process were appointed:

- the lowest value, 0.22% mineral content, considered the technological limit of the demineralizing process, obtained after a chemical reaction lasting 70min, and
- the highest value, 1% mineral content, i.e. the maximum reference value accepted for normal papers for electrical purposes.

When the mineral content value decreases till about 0.35 %, the increase of space charge is expected in the vicinity of the cathode, due to the diminishing of cationic content of cellulose material. But, once the mineral content becomes less than 0.30%, the positive effect of demineralization process may be progressively lost, due mainly to the presence of the residual Cl^- anion, and the space charge may reach saturation. Hereby, another phenomenon provides an important additional contribution, i.e. the acid attach at the cellulose fibers surface (seen as a chemical aging process, consequent to the long demineralization duration and/or the

increased concentration of acid reactive), related directly to the decrease of the cellulose equivalent degree of polymerization. Consequently, a peculiar evolution of space charge may occur by the inferior limit of the mineral content.
At a first view, the difference between patterns in Figs. 5 and 6 is remarkable.
For low mineral content, i.e. 0.22%, a homocharge process was noticed in the vicinity of the cathode, causing an increased field value. When the mineral content increases, the charge close to cathode becomes negative (likely due to injection) and interfacial cathode field decreases, Figs. 5 and 6. As a consequence of field effect and continuous charge injection, charge penetrates in the insulation, reaching the anode and giving rise to hetrocharge formation. Progressively, the field value increases towards the bulk of specimen when mineral content increases.
In order to convert the PEA-derived parameters towards valuable information for quality assessment, the following parameters were appointed:
- the relative value of charge at 15 micro-m [%], obtained from Fig.5, and respectively
- the spatial position of the maximum field [micro-m], obtained from Fig.6.
The correlation between these characteristics and mineral content is presented in Fig.7, providing a reasonable confidence value.
Secondly, the absolute (integral) value of stored charge at the beginning of the depolarization, $\mathbf{Q_0}$, was tested for different mineral content, because particularly this characteristic was appointed to reflect very accurately the charge mobility and the chemical structure, consequent to a certain manufacturing process of the material, [4,5].
The correlation with mineral content involving $\mathbf{Q_0}$ characteristic is presented in Fig.8.

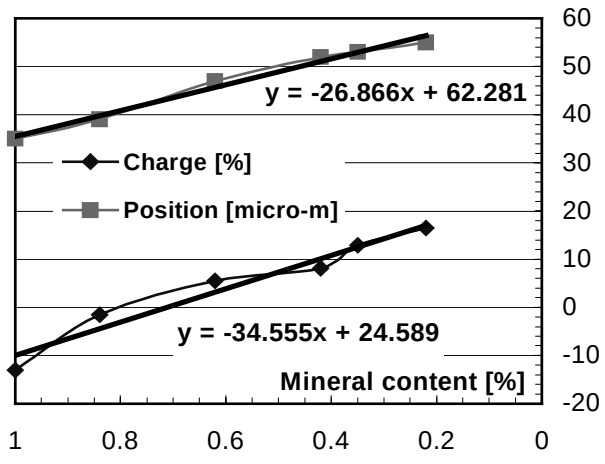

Fig.7 – Example of correlation between PEA parameters and mineral content.

The observation from Figs. 5 and 6 are reconfirmed by Fig. 8, i.e. the higher the mineral content, the shorter the heterocharge process, the higher the stored charge and the lower the field in the vicinity of the cathode.

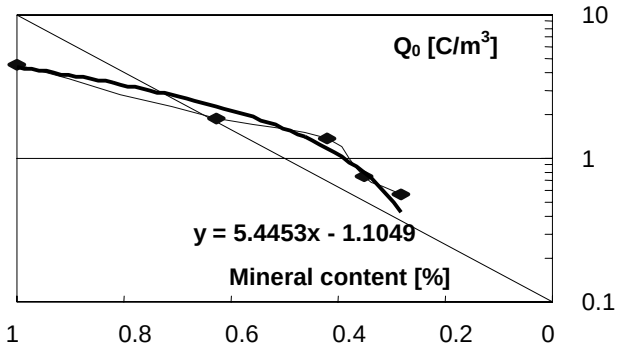

Fig.8 – Example of correlation between $\mathbf{Q_0}$ and mineral content.

4. CONCLUSIONS

Some PEA derived parameters, even not traditionally connected with the dielectric area of interest, can be successfully used towards promoting criteria for the best choice of materials structure, and further, towards evaluating the quality of the applied technology. The problem lies only in planning and performing short time PEA measurements, under well-defined poling conditions, i.e. up to 500s poling time for paper materials (moment when the process of internal charge transport is supposed to be established).

The correlation between the dedicated PEA characteristics and mineral content in paper provided a reasonable confidence value, being considered a valuable procedure towards quick evaluation of paper material performance and/or of the parameters limits of demineralizing process.

ACKNOWLEDGMENT

The very precious scientific and technical support assured by Prof. G.C. Montanari - IEEE Fellow and LIMAT, University of Bologna, Italy, and the periodical sponsorship from the A.v.Humboldt Foundation, Bonn, Germany, are equally thankfully acknowledged.

REFERENCES

[1] G.C. Montanari, G. Mazzanti, L. Dissado and D.K. Das Gupta, "Space charges as a trigger for electrical aging of polymeric insulation", in *Charges in Solid Dielectrics*, The Dielectrics Society Ed., UK, 1998, pp. 259-272.
[2] P. Morshuis and M. Jeroense, "Space charge measurements on impregnated paper: A review of PEA method and a discussion of results", in *IEEE Electrical Insulation Magazine*, 1997, pp. 13-26.
[3] G.C. Montanari, "Electrical degradation threshold of polyethylene investigated by space charge and conduction current measurements", in *IEEE Trans. on DEI*, Vol. 7 no.3, 2000, pp. 309-315.
[4] R. Ciobanu and G.C. Montanari, "Space charge behavior in cellulose materials", in *Proc. of the 5th Conf. on CSC*, Tours, France, July 2001, in press.
[5] G.C. Montanari and D. Fabiani, "Evaluation of dc insulation performance based on space charge measurements and accelerated life tests", in *IEEE Trans. on DEI*, Vol. 7 no.3, 2000, pp. 322-328