

## Method for measuring the dielectrics charging potential under ion irradiation using shifting the bremsstrahlung edge

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A method is proposed for measuring high-voltage charging potentials of dielectrics under ion irradiation by shifting the boundary of the bremsstrahlung X-ray spectrum. Since there is no bremsstrahlung output during  $\text{Xe}^+$  ion irradiation, it was proposed to use a probing electron beam to generate bremsstrahlung X-rays. To eliminate the effect of charge compensation on the surface, the value of the current of the probing probe of electrons was selected. The values of the equilibrium charging potentials of  $\text{Al}_2\text{O}_3$  ceramics,  $\text{Al}_2\text{O}_3$  sapphire,  $\text{SiO}_2$ , and Teflon are obtained at different ion irradiation energies. The data obtained are compared with the results of spectrometric studies.

**Keywords:** dielectric charging, ion irradiation, FIB-SEM, surface potential measurement.

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### Introduction

An active search for new radiation- and temperature-resistant materials [1,2] for the development of modern nuclear systems has led to an increase in the number of works both in the field of research of the materials themselves and in the field of radiation physics of dielectrics [3,4]. Thus, when dielectric samples are studied by standard methods in two-beam scanning electron microscopes with a focused ion beam (FIB-SEM), the dielectrics are strongly charged under both electron and ion irradiation [5,6]. If at this moment a significant number of papers are devoted to the problem of dielectrics charging under electron irradiation, then the problem of dielectrics charging under ion irradiation is not fully understood. At the same time, the vast majority of few publications relate to the study of the charging process for targets consisting of thin dielectric films [7–11] on a conductive grounded substrate. Despite the fact that the depth of path of the irradiating ions is smaller than the thickness of the films under study, for such samples, the significant effect belongs to leakage currents through the materials of the film samples, which reduce the surface potential. Another important factor that reduces the equilibrium value of the surface potential is the capture of secondary electrons by the positively charged surface, the electrons are generated on diaphragms or parts of the chamber and equipment surrounding the sample. The importance of accounting and the effect of this problem are demonstrated in the papers [12,13]. For example, in the publication [14], the process of charging a massive (not film) quartz sample under irradiation with  $\text{He}^+$  ions with energies of 1 to 4 keV was studied. In this case, the indicated

equilibrium surface potential  $V_S$ , determined experimentally, did not exceed several tens of volts.

One of the methods for measuring high-voltage charging potentials in the course of the experiment is the ion-mirror method described in [15]. Here, the equilibrium surface potential  $+V_S$  was calculated from the value of the accumulated charge  $Q_t$  determined from the experiment. But this method gives only estimated results (error 50%). Another method for measuring the dielectric kinetics  $+V_S$  during irradiation with ion beams is the spectrometric method [12,13]. However, this method is difficult to implement in modern commercial FIB-SEM installations and requires additional equipment.

In this paper, a method for estimating the surface charging potential under ion irradiation, which is easily implemented in FIB-SEM units equipped with an attachment for X-ray spectral microanalysis is suggested. It also allows us to confirm the correctness of the conclusions made in the papers [12,13]: the value of the surface potential for bulk dielectrics is close to the accelerating potential of the ion source, and a significant decreasing of the surface potential is the result of the capture of secondary electrons by a positively charged sample.

### 1. Experimental procedure

In the case of a negative charge of a dielectric, when it is irradiated with an electron beam the bremsstrahlung X-ray spectrum edge shifts towards lower energies by the amount of charging  $-qV_S$  due to the energy decreasing of the incident electron beam  $E_0 - qV_S$ . This method is

also used to measure high-voltage charging potentials of dielectrics under electron beam irradiation [16].

When dielectric materials are irradiated with ion beams (PIXE method), the intensity of both characteristic and bremsstrahlung X-ray is much lower than during electron irradiation [17]. In this paper we propose a method for studying the high-voltage positive charge of dielectrics under ion irradiation, based on the shift of the bremsstrahlung X-ray spectrum. In this case, to generate bremsstrahlung X-ray an electron probe is used with a current that is several orders of magnitude lower than the ion charging current. The ion beam of greater density than the probing electron current charges the region of the sample under study and subsequently provides a dynamic equilibrium state of charging. After the sample is charged, the electron probing beam is switched on with the minimum possible current to obtain the bremsstrahlung spectrum. Since during ion irradiation the sample is charged to a potential  $+qV_S$ , the energy of the incident electron probe will increase in a positive charging field to  $E_0 + qV_S$ , and the corresponding bremsstrahlung X-ray spectrum will shift to higher energies. And it is also possible to determine the positive charging potential based on this shift. The disadvantage of this method is the partial discharge of the sample under test by the probing current. To assess the influence of this effect, the value of the probing current was varied, which made it possible to choose the ratio of the incident and probing current, which gives the maximum value of the measured potential. But in this case, the value of the probing current can not be too small, since the chosen spectrum accumulation time may not be enough to obtain statistics sufficient to determine the high-energy edge of the bremsstrahlung X-ray spectrum.

The experimental study of the process of charging by  $\text{Xe}^+$  ions was carried out on a Helios G4 PFIB electron-ion scanning microscope equipped with an EDAX Elite X-ray detector installed in the Advanced Imaging Core Facility laboratory at the Skolkovo Institute of Sciences and Technology. At the same time, the method was tested and charging was studied on the following dielectric samples: single-crystal  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$ , as well as degussite ( $\text{Al}_2\text{O}_3$  ceramics) and Teflon. First of all, the selected sample was charged by a defocused  $\text{Xe}^+$  ion beam with energy of  $E_{i0} = 8$  or  $16$  keV and current of  $I_{i0} = 5$  nA for 5 min. In this case, the charging area was  $100 \times 100 \mu\text{m}$ . After that, the probing electron beam with the energy of  $E_{e0} = 1$  keV and current of  $I_{e0} = 0.4$  nA was switched on, and the accumulation of the X-ray spectrum was started simultaneously. The spectrum accumulation time was 200 s. In this case, the  $\text{Xe}^+$  charging ion beam continued to irradiate the specified region during the entire process of recording the X-ray spectrum. To estimate the value of discharge by the probing electron beam, the study was carried out on a single-crystal  $\text{SiO}_2$  with currents of  $I_{e0} = 0.2, 0.4, 0.8$  and  $1.6$  nA.

## 2. Experimental results and discussion

Fig. 1 shows the bremsstrahlung X-ray spectra recorded on  $\text{SiO}_2$  charged with defocused beam of  $\text{Xe}^+$  ions with  $E_{i0} = 8$  keV and current of  $I_{i0} = 5$  nA for different values of the probing electron beam current  $I_{e0}$ . As our experiments have shown, the bremsstrahlung spectrum edge at the current  $I_{e0} \leq 0.4$  nA practically does not shift, and already at high currents the edge shift is significant.

Let us consider an error arising due to the discharge of the charged dielectric surface by the probing electron beam. When the  $\text{SiO}_2$  sample under study is charged by  $\text{Xe}^+$  ion beam with energy  $E_{i0} = 8$  keV, the maximum possible charging potential is limited by 8 kV. In this case, if such charged surface is irradiated with electrons with energy  $E_{e0} = 1$  keV, then the maximum allowable value of the X-ray photon energy will be  $E_{i0} + E_{e0} = 9$  keV. From the assumption that the maximum possible charging potential will be 8 kV, it can be found that the reduced error will not exceed the value

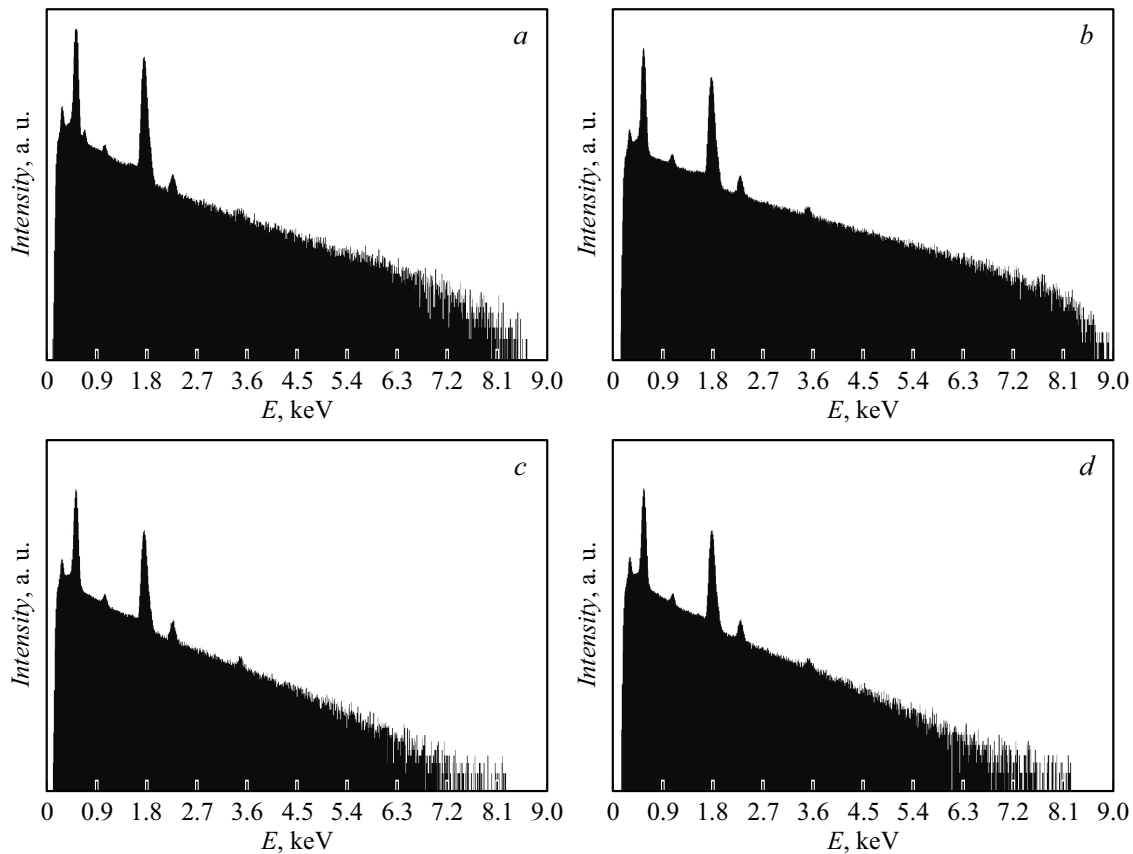
$$\gamma = 1 - \frac{eV_{S0}}{(E_{i0} + E_{e0})}, \quad (1)$$

where  $V_{S0}$  is the equilibrium value of the surface potential, measured from the shift of the bremsstrahlung X-ray spectrum. The reduced value of the surface potential vs. the ratio of the ion beam current to the probing beam current is shown in Fig. 2.

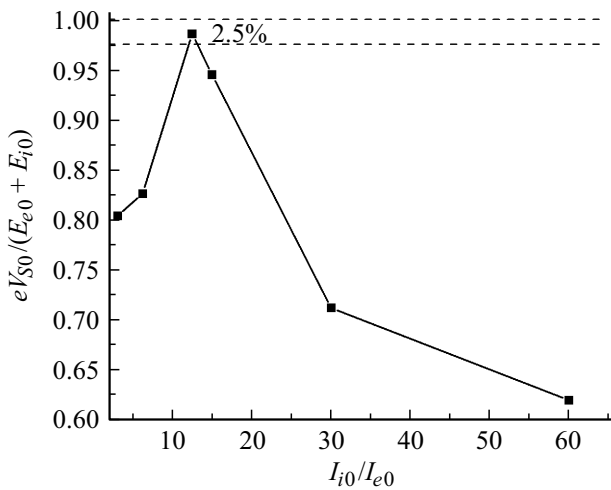
As can be seen from Fig. 2, the measurement error of the surface potential at the ratio  $I_{i0}/I_{e0} \sim 12.5$  can be 2.5% maximum, which is acceptable for the presented method. In this case, if the value of the probing electron current is reduced to the value  $I_{i0} \leq 200$  pA, then the counting rate of the spectrometer greatly decreases, and it is not possible to accumulate a sampling sufficient for determining the high-energy edge of the spectrum for the chosen spectrum accumulation time. This leads to increasing the error in determining this spectrum edge, and thus in the determination of the surface potential of the sample. Thus, to estimate the equilibrium value of the surface charging potential of dielectrics under ion irradiation, one can choose the ratio of the charging and probing current densities. And it is also seen from this dependence that at the ratio  $I_{i0}/I_{e0} > 12$  (provided that the probing current  $I_{i0} > 200$  pA) the probe current does not significantly discharge the sample under study. The disadvantage of this method is the relatively low accuracy of determining the edge of the bremsstrahlung X-ray spectrum due to low statistics at the high-energy edge of the spectrum.

Fig. 3 shows the surface potential  $V_S(E_0)$  vs. the energy of primary ions, measured from the shift of the spectrum of secondary ions during charging (from paper [12]) and according to the presented methodology.

Ion-spectroscopic studies were carried out for samples of  $\text{SiO}_2$ , sapphire and Teflon by irradiating them with  $\text{Ar}^+$  ion beam formed by an Ardenne duoplasmatron. When determining the values of the surface potential according to



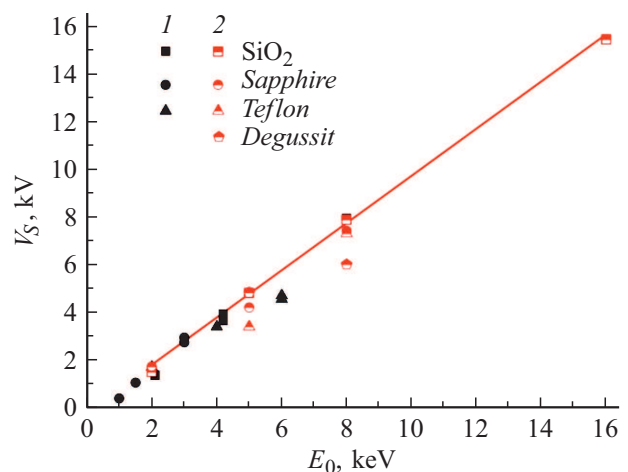
**Figure 1.** Shift of bremsstrahlung spectrum edge for SiO<sub>2</sub> charged by defocused Xe<sup>+</sup> ion beam with  $E_{i0} = 8$  keV at different values of the probing electron beam current  $I_{e0}$ : a — 200 pA, b — 400 pA, c — 800 pA, it d — 1600 pA. Energy of the probing electron beam is  $E_{e0} = 1$  keV.



**Figure 2.** Reduced value of the surface potential  $(eV_{S0})/(E_{e0} + E_{i0})$  vs. ratio of the ion beam current to the probing electron beam current  $I_{i0}/I_{e0}$ . The dashed lines show the relative error range 2.5%.

the presented method, the energy of the probing current of electrons — 1 keV is subtracted from the measured value of the bremsstrahlung spectrum edge. Despite the fact that, in the presented comparison, charging was carried out under

irradiation with ions of different inert gases, the resulting values of the surface potential  $V_S$  fit well into one straight line, close in value to the ion gun potential  $E_0/e$ . As can be seen from Fig. 3, upon the correct selection of the probing



**Figure 3.** Equilibrium value of surface potential for different samples vs. kinetic energy of the primary beam of inert gas ions  $V_S(E_0)$  measured in two different ways: by secondary ion spectrometry [12] during the samples irradiation with Ar<sup>+</sup> ions (1) and by the shift of the edge of the bremsstrahlung X-ray spectrum when the samples are charged with Xe<sup>+</sup> (2).

current both methods give similar results. But unlike the spectrometric method, this approach gives only the value of the equilibrium state of charge, but not kinetic results. It can also be concluded that the value of the equilibrium charging potential for inert gases depends on the energy of the incident ion beam, but not on the chemical composition of the inert gas.

## Conclusion

A new method is proposed for determining the equilibrium charging potential under ion irradiation, which makes it possible to conduct studies in standard FIB-SEM systems equipped with an energy-dispersive X-ray spectrometer. This method, if the correct selection of the value of the probing current is made, makes it possible to obtain the values of the equilibrium charging potential with an acceptable error of maximum 2.5%. The presented method does not allow one to obtain the kinetic characteristics of charging, but allows value measurement of the equilibrium potential of the dielectric surface.

The accuracy of determining the high-energy edge of the bremsstrahlung X-ray spectrum has a great influence on the accuracy of measurement by this method. During the new method testing, it was shown that the value of the equilibrium charging potential of dielectrics under ion irradiation is close to the accelerating potential of the ion source. In other words, for ion irradiation in the kiloelectronvolt range of energy there is no mechanism that regulates the equilibrium value of the surface potential. It can be seen from the comparison of the method of secondary ion spectrometry with the proposed method that the equilibrium value of the surface potential during charging with argon and xenon ions is practically the same.

## Conflict of interest

The authors declare that they have no conflict of interest.

## References

- [1] S.J. Zinkle, G.S. Was. *Acta Mater.*, **61**, 735 (2013). DOI: 10.1016/j.actamat.2012.11.004
- [2] J. Knaster, A. Moeslang, T. Muroga. *Nature Phys.*, **12**, 424 (2016). DOI: 10.1038/NPHYS3735
- [3] F.G. Ferré, A. Mairov, M. Vanazzi, Y. Serruys, F. Leprêtre, L. Beck, L.V. Brutzel, A. Chartier, M. Beghi, K. Sridharan, F.D. Fonzo. *Acta Mater.*, **143**, 156 (2018). DOI: 10.1016/J.ACTAMAT.2017.10.011
- [4] A. Baggott, M. Mazaheri, B.J. Inkson. *J. Europ. Ceramic Soc.*, **39**, 3620 (2019). DOI: 10.1016/j.jeurceramsoc.2019.05.012
- [5] K. Ohya, T. Yamanaka, D. Takami, K. Inai. *Proc. SPIE*, **7729**, 77290V (2010). DOI: 10.1117/12.853488
- [6] Yu.V. Petrov, A.E. Anikeva, O.F. Vyvenko. *Nucl. Instrum. Methods Phys. Res. B*, **425**, 11 (2018). DOI: 10.1016/j.nimb.2018.04.001
- [7] K. Ohya. *J. Vac. Sci. Technol. B*, **32**, 06FC01 (2014). DOI: 10.1116/1.4896337
- [8] J. Shi, M. Fama, B. Teolis, R.A. Baragiola. *Nucl. Instr. Meth. Phys. Res. B*, **268** (19), 2888 (2010). DOI: 10.1016/J.NIMB.2010.04.013
- [9] T. Nagatomi, T. Kuwayama, Y. Takai, K. Yoshino, Y. Morita, M. Kitayama, M. Nishitani. *Appl. Phys. Lett.*, **92**, 084104 (2008). DOI: 10.1063/1.2888957
- [10] S. Yogev, J. Levin, M. Molotskii, A. Schwarzman, O. Avayu, Y. Rosenwaks. *J. Appl. Phys.*, **103**, 064107 (2008). DOI: 10.1063/1.2895194
- [11] R.A. Baragiola, M. Shi, R. Vidal, C. Dukes. *Phys. Rev. B*, **58**, 13212 (1998). DOI: 10.1103/PHYSREVB.58.13212
- [12] K.F. Minnebaev, E.I. Rau, A.A. Tatarintsev. *Phys. Solid State*, **61** (6), 1013 (2019). DOI: 10.1134/S1063783419060118
- [13] E.I. Rau, A.A. Tatarintsev, E.Yu. Zykova, K.E. Markovets, K.F. Minnebaev. *Vacuum*, **177**, 109373 (2020). DOI: 10.1016/j.vacuum.2020.109373
- [14] F. Lefebvre, J. Vigouroux, J. Perreau. *J. Appl. Phys.*, **65**, 1683 (1989). DOI: 10.1063/1.343420
- [15] F. Croccolo, C. Riccardi. *J. Microscopy*, **229**, 39 (2008). DOI: 10.1111/j.1365-2818.2007.01866.x
- [16] X. Meyza, D. Goeuriot, C. Guerret-Piccourt, D. Treheux, H.-J. Fitting. *J. Appl. Phys.*, **94** (8), 5384 (2003). DOI: 10.1063/1.1613807
- [17] M. Nastasi, J.W. Mayer, Y. Wang. *Ion Beam Analysis. Fundamentals and Applications* (CRC Press, London, 2015)