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Use of zeolites in the capture of charged particles from plasma

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The zeolites NaA and 13X were introduced to a coplanar discharge cell to investigate the behavior of charged particles from plasma. The zeolite crystals were attached to the surface without blocking their nanopores. The memory margin related to the accumulated charged particles on the surface indicated that the zeolites absorb charged particles. This phenomenon was also observed at the displacement and discharge current plots. Zeolites with a different window size cause abnormally high displacement and discharge current plots. Note in particular that NaA seems to not only absorb charged particles but also capture gas molecules. © 2008 American Institute of Physics. [DOI: 10.1063/1.2973158]

Zeolites can trap a large number of gas molecules in their cages at a high pressure and temperature if the gas has a slightly larger kinetic diameter than the zeolite window size.¹⁻⁵ According to research reports, 1.8 atoms of xenon (Xe) were encapsulated in the α -cages of NaA at 525 K and 40 bar, and then released by exposing the zeolite to small polar molecules or heating.^{4,6}

Xe as a discharge gas is widely used in plasma display panels (PDPs) consisting of millions of microcoplanar discharge cells. The surface phenomenon caused by ionic Xe or activated Xe species from discharge space is crucial for determining the firing voltage needed to initiate the discharge and the operational voltage margin needed to sustain the discharge of the cells, and for understanding or interpreting the physical phenomena on the surface, such as the striations⁷⁻⁹ and the secondary electron emissions (SEE).^{10,11} With regard to striations, the results of one study suggest that interactions between electron and ion wall charges lead to these striations.⁷ Theoretical research on SEE from a porous surface shows a marked increase in SEE due to the accumulation of charged particles near the pores.^{10,11} From these viewpoints, we are eager to separately analyze electrons or positively charged gas ions from the surface and to inspect a visual signal that occurs on the surface.

In this paper, we attach the following two types of zeolites to the surface of coplanar discharge cells, as described in Fig. 1: 13X (with a window size of 0.8–1 and 1.2 nm super cages) and NaA (with a window size of 0.4–0.42 and 1.1 nm α -cages). The structure used in this experiment is one of the popular discharge cell structures of ac-PDPs. The sawtooth electrode was made of sputtered indium tin oxide (ITO) film with a thickness of 130 nm. A 35 μm transparent dielectric layer was formed on the patterned ITO by means of a screen printing method. To avoid blocking the pores with melted frit glass in a dielectric material during the firing, we fired the dielectric layer after printing it twice, and then printed a thin dielectric layer to fix the zeolite crystals. After drying, we deposited zeolite crystals of 13X (13X molecular sieves, Aldrich) or NaA (4A molecular sieves, Aldrich) on

the dried dielectric film by spin coating with 5 ml of a solution (2.5 ml ethylene glycol and 2.5 ml butyl carbitol acetate) that contained 0.1 and 0.4 g of the zeolites, respectively. Finally, the panels were fired at 823 K for 1 h.

The prepared panel was set in a vacuum chamber and activated at 573 K for 10 h under a high vacuum of 1×10^{-6} Torr. A mixed gas of Ne (96%) and Xe (4%) was then injected into the chamber and fixed at 100 Torr for an aging experiment. All the samples were electrically aged for 1 h and then tested at various gas pressures in the range of 100–250 Torr. The electrodes were connected to a square pulse, the frequency and duty of which were fixed at 40 kHz and 20%, respectively.

The surface images in Fig. 2 show well-dispersed crystals on the electrode area. Because the NaA crystals are smaller than the 13X crystals, we observed that the NaA surface has more dark dots than the 13X surface.

Each panel displays a quick elevation of firing voltage before 15 min of aging, which is a normal characteristic of coplanar discharge cells without a protective layer. After 15 min of aging, all the samples were relatively stabilized in terms of firing voltages. As the aging progresses, the operational memory margin (defined as the difference between firing voltage V_f and minimum sustaining voltage V_s) presents very interesting relationships to the number of crystals and the types of crystals, as shown in Fig. 3. Our previous report mentioned that the memory margin is closely related to the porosity and conductivity of the discharge surface.¹² If a surface has high conductivity, the memory margin should be reduced by a reduction in charged particles on the surface. On the other hand, if a surface has numerous pores, the

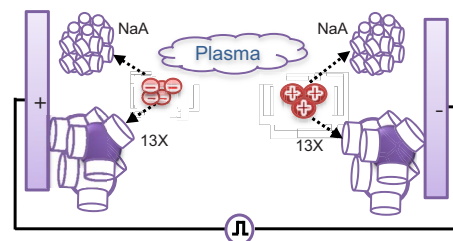


FIG. 1. (Color online) Experimental model for a surface discharge of a cell with NaA or 13X (half way through the cycle of discharge period).

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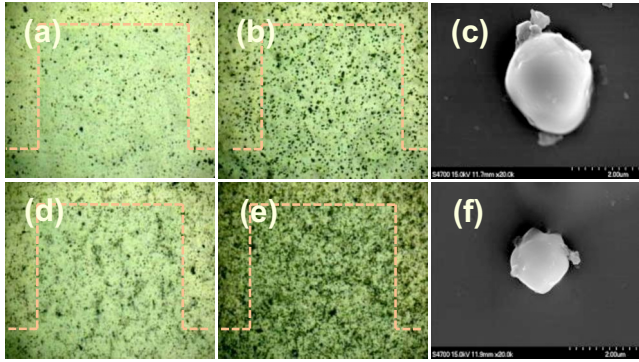


FIG. 2. (Color online) Microscope and scanning electron microscope (SEM) images of the distribution of zeolite crystals (dark dots) on an electrode of a unit cell (dashed lines indicate the boundary of ITO electrode): (a) 13X/0.1, (b) 13X/0.4, (c) SEM image of a 13X crystal, (d) NaA /0.1, (e) NaA /0.4, and (f) SEM image of an NaA crystal.

memory margin should be increased by an increase in charged particles near the pores on the surface. Thus, these results suggest that zeolites capture the charged particles and have difficulty releasing them during discharge. This phenomenon can also be explained by the results of the displacement and discharge currents.

Note that the displacement current in Fig. 4 from the discharged cells with zeolites is remarkably increased regardless of the contents of the crystals. This increase means that the crystals greatly affect the capacitor structures of the discharge cell by adsorbing the charged particles during discharge.

The coplanar discharge cells have three types of capacitors: a discharge space filled with discharge gas, an intergap dielectric, and dielectrics (C_g) between the gas space and the electrodes. Electrically, the cells should show a small change in C_g and, as a result, in the displacement currents. The large change in displacement currents, as shown in Fig. 4, seems to be caused by the adsorption characteristics of the zeolites. As is well known, zeolites of silicoaluminates, such as NaA and 13X, have negatively charged frameworks with ion-exchangeable sites; hence, they tend to favor adsorption of positively charged ions. There are two ways to move charged particles in a plasma space: drift by an electric field and

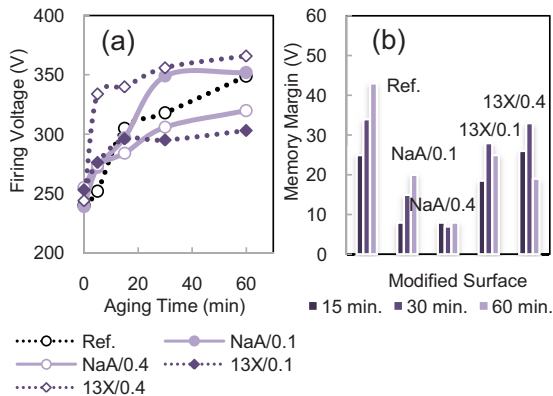


FIG. 3. (Color online) Aging characteristics of test samples operated with a square pulse of 40 kHz and a 20% duty ratio at 100 Torr: (a) Aging time vs initial firing voltages, (b) calculated static memory margin at the aging time of 15, 30, and 60 min, respectively, for samples showing relatively stable operational voltages.

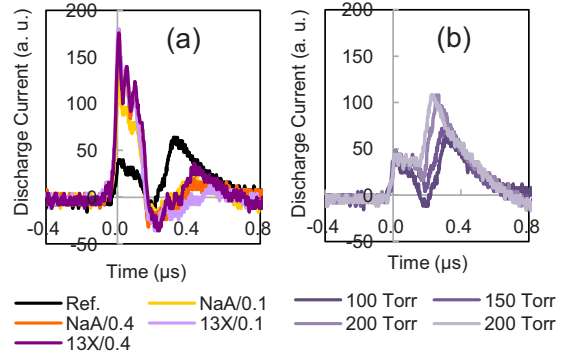


FIG. 4. (Color online) Displacement and discharge currents plotted to the discharge time after 60 min of aging: (a) as measured at 100 Torr and (b) for currents from reference cells vs gas pressure.

diffusion by the concentration gradients described in a drift-diffusion equation with several assumptions.¹³

$$\Gamma_p = \text{sgn}(q_p)\mu_p E n_p - D_p \nabla n, \tag{1}$$

where Γ_p is the flux of particle p , E is the electric field, q_p is the particle charge, μ_p is the mobility, and D_p is the diffusion coefficient. Assuming all the samples have a similar electric field between the electrodes, the diffusion of charged particles into the nanosized pores of the crystal can cause a large change in the displacement current. In fact, the relative permittivity of NaA lies between 2.3 and 5.22, which is a lower range of values than that of the dielectric material in the cells.¹⁴ Thus, when we apply electric fields, some points of the surface with zeolite crystals should have a lower electric field than those points without crystals.

In the case of the discharge current in Fig. 5, the cells with crystals show remarkably lower values than the reference cell. This result suggests that charged particles to be accelerated are greatly reduced, especially the gas ion species, which is the main source of power consumption during a discharge. As the gas pressure increases, the discharge cur-

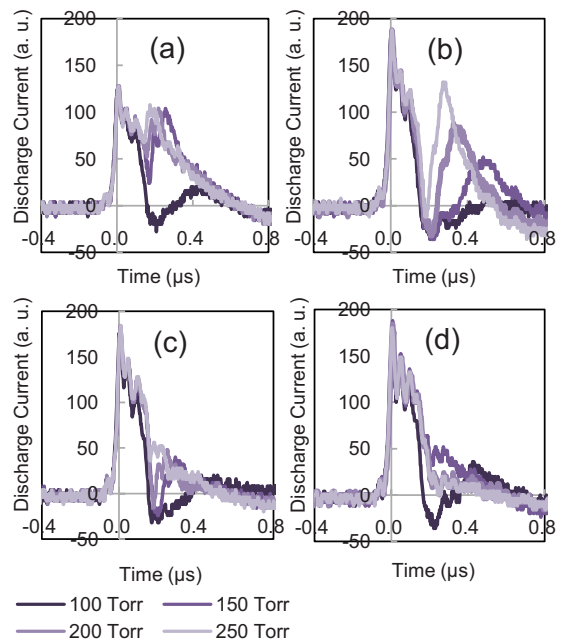


FIG. 5. (Color online) Plots of the displacement and discharge currents of (a) NaA/0.1, (b) 13X/0.1, (c)NaA/0.4, and (d) 13X/0.4NaA/0.1 dependent on gas pressure.

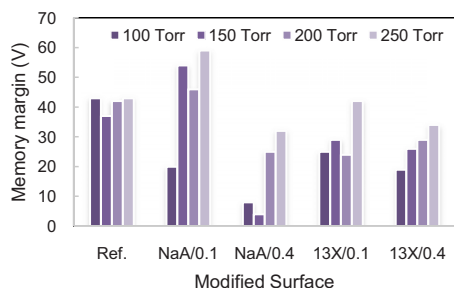


FIG. 6. (Color online) Trend of memory margin of test samples in relation to gas pressure after 60 min of electrical aging.

rent in Fig. 5 from 0.1 NaA quickly reaches a near-saturation state, while the current from 0.1 13X increases linearly.

With regard to the memory margin, Fig. 6 indicates considerable aspects of the discharge characteristics of the samples with zeolite crystals. At the saturation point of the discharge current, 0.1 NaA and 13X have similar or higher memory margins than the reference. Compared to normal discharges, this phenomenon is very strange and has not previously been reported.

Experiments and simulations indicate several kinds of ionic species in a discharge space, namely, Ne^+ , Xe^+ , Xe_2^+ , and NeXe^+ . Of these, Ne^+ disappears quickly during the

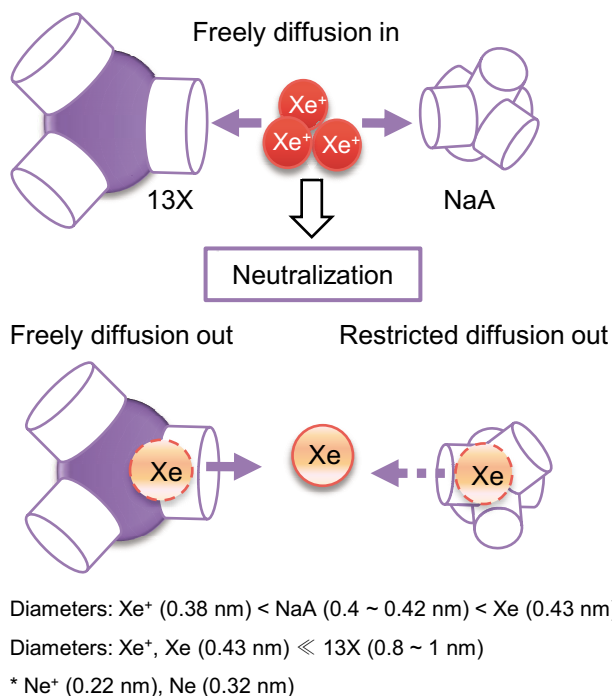


FIG. 7. (Color online) Ideal figure for Xe^+ ion and Xe diffusion into and out of channels of NaA or 13X on surface of discharge cell.

discharge; Xe^+ subsequently disappears; and Xe_2^+ eventually becomes the most important ion species.^{13,15} Note that NaA seems to freely capture most of the ionic species. However, if it captures ionic species, the diffusion of molecules is restricted, except for Ne^+ . The restriction is likely due to the preoccupied gas molecules and probably the neutralized Xe molecule, the size of which is larger than the window size of NaA, as shown in Fig. 7. Given that most of the electrical energy to be discharged is consumed in the process of accelerating the charged particles and gas ions, the adsorption characteristic of the zeolites is the likely reason why the discharge current from 0.1 NaA is quickly saturated when gas pressure increases. For 0.1 13X, the adsorption capacity is similar but the 1 nm entrance window is larger, which enables the charged particles or gas atoms to freely move in and out regardless of the ionic species. These free diffusions can explain the linear increase in the discharge current and the gas pressure in Fig. 5. In the case of 0.4 NaA and 13X, the adsorption capacity seems too high to present the diffusion selectivity of zeolites.

All experimental results confirm that charged particles from plasma can be absorbed by zeolite crystals. In addition, the memory margins of the NaA samples suggest that the diffusion of ionic Xe can be restricted by preoccupied Xe molecules whose size is larger than the window and channel sizes of NaA. Another important result is the high value of the displacement current, which is generally ignored. This experiment confirms that the displacement current can be greatly changed by the charged particles on the surface.

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