The distribution of the condensate surface potential was studied during condensation of a polarized molecular beam onto a substrate with a one-dimensional temperature gradient. It was found that on the free surface of the condensate layer, electric regions of different sign appeared: electric domains. Estimation of the surface charge density within a single domain in the case of a poly-domain structure of the condensate gives considerably smaller values than that obtained for the mono-domain condensate with a uniform polarized structure.

# Electric domains created in polarized molecular condensates during condensation onto a substrate with a one dimensional temperature gradient

J. Chrzanowski and B. Sujak

#### Key words: cryogenic, condensate, electric domains

Due to development of low temperature electrical engineering in the last years, the working conditions of many electrical devices changed significantly. The conditions created by high vacuum and low temperatures generate an unknown environment in which new physical processes may be the important factors affecting the reliability of those electrical devices.

The process of residual gas condensation onto some essential constructional cold parts may induce real changes in the electrical properties of the solid state surface of these parts. A series of papers concerning practical applications or fundamental research,<sup>1-6</sup> published in the last decade has been devoted to electrical surface phenomena appearing at the condensation of polar gases and vapours. These papers showed an active menace of cryo-electrical devices which occurred as the result of the strong electrical fields generated by electrically polarized cryocondensates of residual gases. The condensation of a beam of polar molecules onto a substrate cooled down to low temperature, leads to the build-up of an electrically polarized condensate layer.<sup>2-7</sup> The sign of the surface potential of the polarized condensate depends on the kind of the condensed gas and on the respective condensation substrate temperature. It was also shown recently,<sup>6</sup> that in some cases even the condensation of non-polar gas mixtures may lead to similar effects.

The model describing the mechanism where by the condensate is electrically polarized, proposed by us,<sup>7</sup> suggests that in the case of condensation of polar molecules onto a substrate with a one-dimensional temperature gradient parallel to the surface which creates large temperature differences between the adjacent surface regions, the appearance of electrical domains should be possible as a result of the different-sign of electrical charge on various condensate regions. The existence of such domains may also affect the electrical properties of the substrate surface and the surrounding volume. This paper is devoted to the experimental verification of such electrical domains under the conditions mentioned above using a vibrating electrode method. (A. Kruk observed in 1980 the existence of such domains in the case of a long narrow substrate panel using a static movable sonde,<sup>9</sup>).

## **Experimental details**

The wheel shaped condensation panel (substrate and condensation electrode) was made of copper plate with a radius R = 50 mm. To the edge of this panel an electrical heater was attached. The central part of the condensation panel was cooled down with the aid of a continuous flow cryostat.<sup>8</sup> The configuration cold finger (condensation panel) edge heater assured very good heat transport as well as sufficient electrical insulation. This device allowed us to obtain a temperature distribution T = T(R) along the panel radius. The mean gradients used were of the order dT/dR = 5 - 20 K cm<sup>-1</sup>. The temperature at the central part of the condensation panel could be held at about 90 - 100 K, while at the same time the external part of the panel near the edge was about 170 - 180 K. In some experiments the central part of the panel was cooled without heating at its edge. In this case the temperature of the external part near the edge was only by about 20 K higher than at the centre.

For the detection of the surface charges appearing on the condensates a classical arrangement using a vibrating electrode was used. The vibrations of the measuring electrode were induced by an electromagnetic generator supplied with a power oscillator. The fundamental frequency of the oscillations was  $f_0 = 21$  Hz. The mean distance between the vibrating element and the surface of the condensation substrate was fixed to about 0.5 mm. The diameter of the vibrating electrode was  $\phi = 10$  mm. A schematic of the experimental arrangement with the detection circuit is shown in Fig. 1.

After amplification of the signal by a preamplifier, the signal was transmitted to the selective nanovoltmeter, which indicated the alternating current of the second harmonic:

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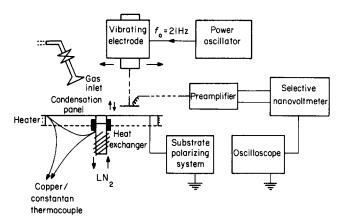


Fig. 1 Block diagram of the experimental system: 1 - vibrating electrode, 2 - power oscillator, 3 - preamplifier, 4 - selective nanovoltmeter, 5 - oscilloscope, 6 - substrate polarizing system, 7 - heat exchanger, 8 - heater, 9 - condensation panel, 10 - gas inlet, TC - copper/constantan thermocouple

 $2 f_0 = 42$  Hz. The value of the surface potential and its sign for a specific element of the condensate layer was determined by polarization of the substrate with the constant potential  $U_c$  (+ or -) to compensate for the local electric field in the region produced by the vibrating element and the specific element of the condensate layer.<sup>10</sup> For condensates thinner than 0.1 mm the externally applied compensation potential  $U_c$  is equal, with good accuracy, to the measured value of the surface potential  $U_c$  but opposite in sign:

$$U_{\rm c} = -U_{\rm s} \tag{1}$$

An oscilloscope connected to the zero-indicator allowed visual control of the harmonic motion of the vibrating electrode's induced oscillation.

The parallel movement of the vibrating electrode along the radius of the condensation panel (substrate) was assured by respectively constructed mechanical devices. Experiments with a clean electrically polarized condensation panel (Fig. 2) show that the unavoidable small deviations of the distance between the laterally moved vibrating electrode and the substrate have only a minor influence on the registered signal. For the following experiments with condensates these deviations in the indicated signal have therefore been neglected.

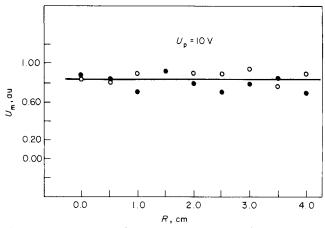


Fig. 2 ac signal observed with the vibrating electrode was moved along the radius of the clean substrate (substrate polarizing voltage  $U_{\rm p}$  = 10 V)

The condensation process takes place at a pressure of the order  $10^{-5}$  Torr.

### **Experimental results**

As a source for the respective gases to be condensed the following three pure liquids were used: water ( $H_2O$ ), acetone (CH<sub>3</sub>COCH<sub>3</sub>) and ethyl alcohol (CH<sub>3</sub>CH<sub>2</sub>OH). These substances were selected because the electrical properties of their condensed phases were sufficiently well known.<sup>4,7</sup>

In Fig. 3 the increase of the surface potential is presented. An increase occurs when a slow condensation process  $(H_2O)$  takes place on the previously ice free panel at a temperature T = 90 K. It was ascertained that the observed potential is negative compared to earth potential. Its value increased linearly with the thickness of the condensate layer. A similar tendency was observed for the condensates of other polar gases. This confirmed the earlier observations.<sup>4</sup> In the following Figs 4 - 6 the dependence of the surface potential magnitude for H<sub>2</sub>O, CH<sub>3</sub>COCH<sub>3</sub> and CH<sub>3</sub>CH<sub>2</sub>OH condensates is shown when along the radius of the substrate surface the temperature difference given above was maintained.

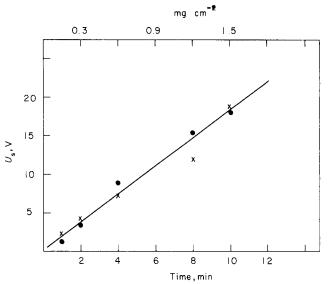


Fig. 3 Voltage curve obtained during slow condensation of an ice condensate onto the substrate at a temperature T = 90 K

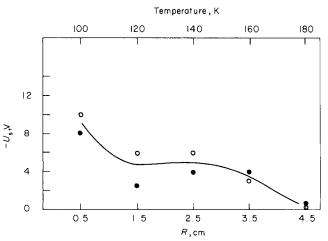


Fig. 4 The surface potential of the ice cryocondensate plotted against the substrate radius. The approximate temperature values along the radius are also indicated

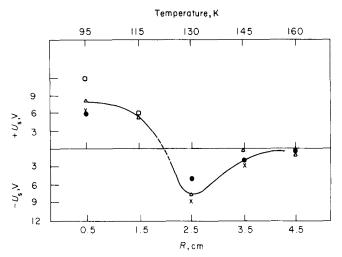


Fig. 5 Two-domain structure observed for acetone cryocondensate with positive (+) region at substrate temperatures T = 90 - ca. 110 K and negative (-) region at higher substrate temperatures

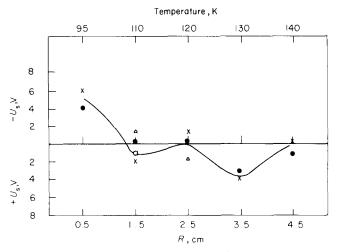


Fig. 6 Poly-domain structure observed for ethyl alcohol cryocondensate

Fig. 4 shows the distribution of the surface potential for ice layers in different regions of the substrate. It can be seen that the magnitude of this potential decreases considerably with increasing temperature of the substrate surface (condensate) up to about T = 120 K, remains almost constant over the next range up to T = 150 K and decreases again at higher temperatures. The next figures illustrate that the condensate surface shows different regions with positive or negative surface charges when acetone or ethanol are condensed.

Repetition of the measurements for a specific condensate yielded in all cases a rather large dispersion of the magnitude of the measured surface potential. This may be a consequence of local changes of the surface charge density distribution or may be provoked by changes of the thickness of the condensate layer due to re-sublimation.

The observed values of the surface potential suggest the existence of strong internal electric fields inside the condensates. To estimate the density of the surface charges the permittivity of the condensates  $\epsilon$  should be known. However, besides some data for ice, CO and SO<sub>2</sub><sup>11</sup> this parameter is not known for the condensate layers. Taking into consideration the fact that most condensates have a similar structure as the above mentioned substances (low

density, amorphic or semi-glassy texture), it may be assumed that the dielectric permittivity of other condensates, in the studied temperature range, to a first approximation is of the same order, that is, can be taken as  $\epsilon \sim 2 \div 3$ . With this assumption the known values of the surface potential and of the thickness of the condensate layer allowed us to estimate the magnitude of the surface charge density.

In the case of the electrical circuit used in this experiment a simple consideration leads to an expression for the surface charge density:

$$\sigma = \epsilon \epsilon_0 \ \frac{U_c}{d} \tag{2}$$

where  $U_{c} = -U_{s}$  is the value of the compensation potential,  $\epsilon$  and  $\epsilon_0$  are the dielectric permittivity of the condensate layer and the vacuum respectively, and d is the thickness of the condensate layer. The latter was determined taking into consideration the condensed mass of the substance per unit area of the condensing substrate. It was assumed, except for the ice condensates, that the density of the condensate layers approaches the density of the liquids used. (For water vapour condensates obtained in this temperature range  $\rho = 0.58 \text{ g cm}^{-3}$ .<sup>12</sup>) The estimations of  $\sigma$ , presented below, should be taken into account with a great caution especially for the poly-domain structures. As was mentioned above, the thickness of the condensate films was determined only as a mean mass thickness. A local thickness of the condensate should be a function of the radius R (local temperature of the substrate).

It is worth while remarking once more that the temperature gradient introduced in this paper is a function of the substrate radius. The local temperature of the substrate along its radius changes non-linearly and therefore dT/dR is also changing with the substrate radius.

The estimation of the surface charge density  $\sigma$  for the condensates studied are presented in the Table 1.

#### **Concluding remarks**

The presented results indicate the existence of electric regions of different sign, electric domains, in the cryocon-

Table 1.	Presenting the estimated surface charge density $\sigma$			
for the cryocondensate studied				

	$\sigma$ C cm <sup>-2</sup> Uniformly pol structure	$\sigma_{\rm max} ~\rm C.cm^{-2}$	
Substance		Mono-domain structure	Poly-domain structure
H <sub>2</sub> 0 (water)	(–) 10 <sup>-7</sup> – 10 <sup>-8</sup> at 90 K	(–) 3.10 <sup>-9</sup> at 100 K	
CH <sub>3</sub> COCH <sub>3</sub> (acetone)	(+) 5.10 <sup>-9</sup> at 90 K (–) 2.10 <sup>-10</sup> at 130 K	(+) 10 <sup>-9</sup> - 10 <sup>-10</sup> at 90 K (-) 10 <sup>-10</sup> - 10 <sup>-11</sup> at 130 K	(+) 10 <sup>-10</sup> at 95 K () 10 <sup>-11</sup> at 130 K
CH <sub>3</sub> CH <sub>2</sub> OH (ethyl alcohol)	() 2.10 <sup>-9</sup> at 100 K (+) 5.10 <sup>-10</sup> at 130 K	(—) 10 <sup>-9</sup> at 100 K —	(+) 10 <sup>-10</sup> at 95 K (-) 10 <sup>-10</sup> at 130 K

densed layers of polar molecules deposited onto a substrate with the studied, one-dimensional, temperature gradient. This will also hold for a two-dimensional gradient.

The surface charge density estimated for mono-domain as well as for poly-domain condensates is considerably lower than the  $\sigma$  value obtained for a regular condensate which was cryocondensed onto a panel of uniform temperature. This may be connected with electric currents (ions ?) flowing in the condensate induced by the temperature difference and by electrical fields appearing between the adjacent domains.

We therefore conclude that the surface potential differences between adjacent electric domains developing in some cryocondensates may reach tens of volts. Uncontrolled growth of the condensate thickness may therefore lead to generation of potential differences of the order of thousands of volts between the near regions of solid surface. The mean linear size of the domains is of some centimeters or shorter. This may lead to the appearance of strong local electric fields on the respective cryocondensate surface. The hypothetical currents (ions?) flowing between the adjacent domains may also provoke changes of the conductivity of the condensation substrate surface. The existence of the postulated currents in the condensates may be a source of specific electrolytic processes affecting the substrate surface or inducing in the condensate itself some chemical reactions leading to production of free ions, radicals or even UV radiation. The postulated currents could be manifested

by subtle magnetic effects which might be detectable with SQUID techniques. The poly-domain structure of the condensates in question may be a reason for electrical breakdowns along the condensate surface which in turn can cause a local increase of temperature leading to gas pressure enhancement. This may be a source of gaseous characteristic discharges as in the case of surface discharge phenomena on ferroelectrics.<sup>13</sup>

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