

## NUCLEATION OF VAPOURS OF BENZOIC ACID IN THE DIFFUSION CLOUD CHAMBER

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Critical supersaturation of vapours of solid benzoic acid was measured in the diffusion cloud chamber in the temperature interval from 270 to 310 K. The dependences of critical supersaturation on temperature were calculated according to the classical and Lothe-Pound equations for the rate of homogeneous nucleation and the Volmer and Russel equations for the rate of heterogeneous nucleation on gaseous ions. The experimental results correspond, in comparison with the theoretically calculated data, to the homogeneous nucleation of vapours in the form of crystal nuclei. At temperatures above 320 K the heterogeneous nucleation on gaseous ions was observed.

In processes in which it is necessary to condense the vapours of the evaporated matter from the inert gas the problem of formation of fog is met. The fog can form if a certain supersaturation of vapour mixture is reached. At condensation of supersaturated vapour, nuclei of condensed phase must form at first inside the bulk of the vapour phase. The nuclei can form themselves spontaneously due to the fluctuation of density of the vapour phase (homogeneous nucleation) or on condensation centres which are usually particles of dust or gaseous ions (heterogeneous nucleation). Critical supersaturation for nucleation from the vapour is defined as that ratio of actual pressure of vapour to equilibrium vapour pressure of the bulk of condensed phase at the same temperature for which the nucleation rate becomes appreciable. If supersaturation is smaller than critical supersaturation the nucleation rate is so small that supersaturated vapour phase is relatively stable. Above  $S_{crit}$  the nucleation rate sharply increases and the vapour phase condenses in the form of fog.

If we want to avoid formation of the fog, it is necessary to condense the vapour mixture under the conditions when the possible supersaturation is smaller than the critical one. The necessary critical supersaturation can be estimated from equations for the nucleation rate obtained on basis of theoretical assumptions. But it has been observed that for different compounds different rate equations are suitable<sup>1</sup>. For estimate of critical supersaturation it is also necessary to take into consideration that the vapour can condense in the form of liquid even though the temperature is below the freezing point<sup>2-4</sup>. Temperature at which the change in nucleation mechanism takes place is usually not known. Also the specific effect of different gaseous

ions in heterogeneous nucleation is not known. For these reasons it is in most cases necessary to measure the critical supersaturation experimentally.

Equations for the rate of homogeneous nucleation of supersaturated vapours are usually used for expression of the dependence of critical supersaturation on temperature. Variation of critical supersaturation with temperature can be predicted by use of the classical equation which for formation of liquid nuclei can be written in the form<sup>5,6</sup>

$$J = a(2N^3/\pi)^{1/2} (P_e/RT)^2 S^2(\sigma M/d^2)^{1/2} \exp\{(-16\pi N/3R^3)(M/d)^2(\sigma/T)^3(\ln S)^{-2}\}, \quad (1)$$

where  $a$  is the condensation coefficient,  $J$  nucleation rate ( $\text{cm}^{-3} \text{s}^{-1}$ ),  $N$  Avogadro's number,  $R$  gas constant,  $M$  molecular weight,  $d$  density of liquid,  $\sigma$  surface tension,  $P_e$  pressure of saturated vapours at temperature  $T$  and supersaturation  $S$ . The nucleation rate of crystal nuclei from supersaturated vapour can be at the assumption of spherical nuclei, isotropic specific surface free energy, and the "molecularly rough"<sup>7,8</sup> surface of crystal nuclei, described according to the classical theory by the same equation<sup>8,9</sup>. For a general form of crystal nuclei it is necessary to introduce the corresponding shape factors in the exponential and preexponential terms<sup>9,10</sup>. As the specific surface free energy of crystal nuclei is in general anisotropic, the mean specific surface free energy<sup>9</sup> can be used in Eq. (1). For the "molecularly smooth" surface of crystal nuclei it is necessary to consider the two-dimensional nucleation on the surface<sup>9,11</sup>. Lothe and Pound<sup>12,13</sup> have introduced the correction term which is increasing the nucleation rate according to the classical theory for a factor  $10^{17-20}$ . Nucleation rates according to the classical theory  $J_{\text{clas}}$  and according to the Lothe-Pound theory  $J_{\text{LP}}$  are related at the assumption that the "replacement factor" is of the order of magnitude<sup>13</sup>  $10^{-3}$  by the relation

$$J_{\text{LP}} = (0.02897(MTi)^4/dP) 10^{-3} J_{\text{clas}}, \quad (2)$$

where  $i$  is the number of molecules in the critical nuclei and  $P$  the actual vapour pressure (Torr). For an estimate of the dependence of critical supersaturation of vapours on temperature at heterogeneous nucleation on gaseous ions, again the corresponding equation for the nucleation rate can be used. The equation according to Volmer<sup>5</sup> has the form

$$J = (a/R\pi)(N/2)^{1/2} (P_e/Td) SM^{1/2} N_a [4\pi\sigma - (1 - \epsilon^{-1}) e^2/r_k^3]^{1/2} \times \\ \times \exp\{-2N/3R[2\pi(\sigma/T)(r_k^2 - r_a^2) + e^2(1 - \epsilon^{-1})(r_k^{-1} - r_a^{-1})/T]\}, \quad (3)$$

where  $N_a$  is concentration of gaseous ions,  $\epsilon$  dielectric constant of liquid,  $e$  elementary charge ( $4.8 \cdot 10^{-10}$  aec) and  $r_k$  and  $r_a$  the radii of the drop which is in an unstable or metastable equilibrium with the vapour. The radii  $r_k$  and  $r_a$  are obtained as the largest and the next smaller roots of equation

$$-(RTd/M) \ln S + (2\sigma/r) - (e^2/8\pi r^4)(1 - \epsilon^{-1}) = 0. \quad (4)$$

Russell<sup>14</sup> has obtained the following equation for the nucleation rate by application of the Lothe-Pound theory on condensation on gaseous ions

$$J = (a/R\pi)(N/2)^{1/2} (P_e/Td) SM^{1/2} N_a (r_k/r_a)^{12} [4\pi\sigma - (1 - \epsilon^{-1}) e^2/r_k^3 - 18RT/Nr_k^2]^{1/2} \times \\ \times \exp\{-2N/3R[2\pi(\sigma/T)(r_k^2 - r_a^2) + e^2(1 - \epsilon^{-1})(r_k^{-1} - r_a^{-1})/T]\}, \quad (5)$$

where  $r_k$  and  $r_d$  are obtained as the largest and the next smaller root of equation

$$-(4\pi RTd/M)r^4 \ln S + 8\pi\sigma r^3 - 12(RT/N)r - (e^2/2)(1 - \epsilon^{-1}) = 0. \quad (6)$$

In order to study nucleation of condensed phase in the supersaturated vapour it is necessary to produce supersaturated vapour of known supersaturation. For this purpose several experimental techniques have been developed which are summarized and critically evaluated in studies<sup>1,15-17</sup>. The most applied method is cooling the vapour mixture by a rapid adiabatic expansion in the expansion cloud chamber or by its flow through a supersonic nozzle. In another method the saturated hot vapour mixture is cooled at turbulent mixing with a cooler gas<sup>17-20</sup>. Lately a method is also used for study of nucleation of vapours of liquids in which the supersaturation of vapour mixture is reached at nonisothermal diffusion flow in the diffusion cloud chamber<sup>21-25</sup>.

In principle, the diffusion cloud chamber consists of two metal plates connected by a glass cylinder. The bottom plate with the studied liquid is heated and the upper one is cooled.

The chamber is designed so that at high degree of approximation one-dimensional diffusion takes place from the heated pool of liquid through the inert carrier gas to the upper plate where the vapour molecules condense. The chamber is constructed so that the condensing liquid is flowing downward the walls back to the bottom plate and the apparatus can be thus operated continuously. By solution of one-dimensional equations for heat and mass transfer on basis of temperatures of both plates and the corresponding saturated vapour pressures the profiles of partial pressure and temperature in the chamber are obtained. As both these profiles vary approximately linearly with the height in the chamber while the pressure of saturated vapour is varying with temperature approximately exponentially, the vapour mixture is in the chamber supersaturated. By increasing the temperature gradient between the plates, supersaturation can be arbitrarily increased with liquid drops appearing in the chamber at the sufficient supersaturation. By a suitably selected temperature gradient the nucleation rate  $\sim 1$  can be reached. The critical supersaturation corresponds to the maximum supersaturation in the chamber at the observed nucleation rate. The critical supersaturation in dependence on temperature of water vapour<sup>25</sup> and of numerous vapours of organic liquids<sup>21-24</sup> was measured by this method.

Though it is assumed that the diffusion cloud chamber is suitable for studies of nucleation of vapours with boiling points of their liquids being substantially greater than the tripple-point<sup>23</sup>, we have made an attempt to arrange the operation of the diffusion cloud chamber for measurements of condensation of supersaturated vapours

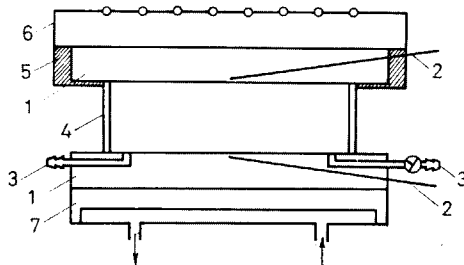


FIG. 1  
Diffusion Cloud Chamber

of solid organic matters. For this purpose a simple model of the diffusion cloud chamber has been constructed and its operation has been tested by measurements of critical supersaturation of vapours of the solid benzoic acid.

## EXPERIMENTAL

**Apparatus.** Bottom and top of the chamber (Fig. 1) is formed by two dural circular plates 1 of the diameter 180 mm and 20 mm thick. In plates the channels are drilled for the measuring thermocouples 2 and holes for filling the chamber by inert gas and for measurement of pressure 3. The plates hold together by six screws situated on their circumference. The walls of the chamber are formed by a glass Simax cylinder 4 with the inside diameter 140 mm, wall thickness 4 mm and 43 or 21 mm high which is in contact with both plates sealed by Teflon. The upper plate is insulated by Lukopren 5. The upper plate is heated by a coil 6 made of resistance body situated on a copper plate 210 mm in diameter and 20 mm thick which distributes heat from the heated body on the whole contact area. Heating is controlled by a variable voltage transformer. The bottom plate of the chamber is cooled by the plate 7. This cooling plate is a closed copper cylindrical vessel 180 mm in diameter and 22 mm high. The bottom of the vessel which is in contact with the bottom plate of the chamber is 15 mm thick so that a constant temperature on the whole contact area is obtained. The cooling plate is in contact with the liquid thermostat. The contact areas of heating and cooling plates and of both chamber plates are ground so that they are in close contact resulting in good heat transfer. A 15 W lamp with biconvex lens was used to produce a beam of light illuminating the interior of the chamber. The forward scattering of light by the particles was visible at small angles (10 to 30°) of the beam axis. For perfect visibility of particles part of the glass cylinder was covered from the external side by black paint and from the internal side it was matted in order to remove the light reflexes.

Temperature of both plates was measured by jacketed NiCr—Ni thermocouples 1 mm in diameter connected to the line recorders EZ 4 and EZ 10. For the used sensitivity of recorders the measurements were performed with a  $\pm 0.1$  K accuracy. Thermocouples were situated in channels 2 with their ends in the centres of plates about 0.5 mm below the surface. By measuring the temperature of the plate surface in various points by a contact thermocouple and by comparison with the measured temperature in the channels, it was found that both temperatures differ in average by  $\pm 0.2$  K, *i.e.* in the range of accuracy with which the temperature is measured. To find out how sensitive is the thermocouple in the channel to changes in temperature of the surface, the plate was covered with the amount of about 1 g of benzoic acid in the point above the end of the thermocouple and the plate was heated with the rate 5 K/min. Temperature was recorded. At the melting point of benzoic acid there appears on the record a retention which corresponds to the melting point of benzoic acid with an accuracy of  $\pm 0.2$  K. Pressure in the chamber was measured by a mercury manometer.

**Compounds.** The compound used for testing the whole equipment was chosen on basis of two requirements: for comparison with the theoretically obtained values it is necessary to know some of physical properties of the measured compound and its temperature dependence and the compound must be stable in the studied range of temperatures. For these reasons benzoic acid was chosen (analytical grade reagents, Lachema, Brno). The possible decarboxylation of benzoic acid can be neglected (at 400 K and concentration corresponding to the vapour pressure of saturated vapours at this temperature the initial reaction rate of decarboxylation of benzoic acid<sup>26</sup> is  $r^0 \sim 10^{-24}$  mol l<sup>-1</sup> s<sup>-1</sup>). Physical properties of benzoic acid and of helium are: Benzoic acid:  $M = 122.05$  (g mol<sup>-1</sup>);  $\log P = 12.2937 - (4530 \pm 80.988)/T$ ;  $290.4 - 315.5$  K (Torr)<sup>27</sup>;

$\log P = 30.172 - 4714/T - 6.720 \log T$ ;  $395.7 - 523.2 \text{ K}$  (Torr)<sup>28</sup>;  $\sigma = 45.8 - 0.11 (T - 273.15)$  (dyn cm<sup>-1</sup>); \*  $d_1 = 1.19335 - 0.0009107(T - 273.15)$  (g cm<sup>-3</sup>), \*\*  $d_s = 1.316$ ; 300°K (g cm<sup>-3</sup>)<sup>28</sup>;  $\lambda \approx 0.4 \cdot 10^{-3}$  (cal cm<sup>-1</sup> s<sup>-1</sup> grad<sup>-1</sup>), \*\*\*  $C_p(g) \approx 30$  (cal mol<sup>-1</sup>), \*\*\*\*  $\Delta H_{\text{subl}} = 20$  (kcal mol<sup>-1</sup>)<sup>28</sup>;  $D_{\text{He-benzoic acid}} = 0.268$  (cm<sup>2</sup> s<sup>-1</sup>); 300 K, 1 atm<sup>29</sup>; Helium:  $\lambda = 0.000341 + 0.7164 \cdot 10^{-7}(T - 273.15)$  (cal cm<sup>-1</sup> s<sup>-1</sup> grad<sup>-1</sup>). \*\*\*\*\*

Helium, nitrogen and methane were taken from pressure cylinders with water removed by freezing in the freezer by a mixture of solid CO<sub>2</sub> and ethanol.

*Experimental procedure.* As far as the evaporated and condensed phases are solid, the diffusion cloud chamber cannot be operated continuously and the steady state cannot be reached. Direct measurement of the condensation surface is difficult. But the surface temperature of the plate can be measured and a correction on the thickness of the condensing layer can be made. For these reasons it is desirable that the layer would grow very slowly. The diffusion flows are in the majority of cases small due to large molecular weights of majority of subliming organic matters and their very low saturated vapour pressures below the tripple point. As the nucleation rate is practically independent of the total pressure and the type of inert gas, the diffusion flow can be decreased by a large partial pressure and by the type of inert gas. The contribution of the evaporated matter to the total density of the mixture vapour-inert gas is then small. To avoid convection, the upper plate of the chamber must be heated and the lower cooled so that the total density of the vapour mixture is greater at the lower plate.

For evaluation of both profiles the steady state has been approximated at various rates of setting of the temperature gradient between the plates and by their extrapolation to the zero velocity.

On the heated plate benzoic acid was melted and the molten acid was subcooled by up to 15 K. The subcooled benzoic acid solidified into the compact uniform layer stuck to the plate surface and was 0.2 to 0.3 thick. The chamber was filled by inert gas by a continuous inlet. The temperature gradient between the plates was fixed so that at the same initial temperature of both plates the bottom plate was kept at constant temperature and the upper was heated at a constant rate. The inside of the chamber was observed under the side light and at the moment the first particles have appeared, temperatures of both plates were recorded together with the total pressure in the chamber. Particles formed in the same plane about in one tenth of the height from the lower plate and were falling directly toward the lower plate.

## RESULTS AND DISCUSSION

The measurements were made at various temperatures of the bottom plate. The effects of the total pressure, of inert gas and of the chamber height on supersaturation were studied at various heating rates of the upper plate. With respect to low partial pressures of benzoic acid and its large molecular weight in comparison to the inert gas, the molecular diffusivity of the mixture vapour-inert gas is smaller than the

\* The temperature dependence of surface tension of benzoic acid was obtained from available data<sup>28,30</sup> and from the dependence of surface tension of *m* and *o* toluyl acid on temperature<sup>31</sup> according to the publication<sup>32</sup>; \*\* obtained from the available data<sup>28,30</sup>; \*\*\* temperature dependence of thermal conductivity of solid benzoic acid in the published paper<sup>33</sup> differ considerably, thus the mean value was chosen and its temperature dependence was not considered; \*\*\*\* estimated by the method of Rihany and Doraiswamy<sup>34</sup>; \*\*\*\*\* obtained from the published data<sup>35</sup>.

thermal diffusivity and it can be expected that during fixing of the temperature gradient between the plates the concentration profile will be steadied at a smaller rate than the temperature profile. In agreement with this statement the resulting temperature of the upper-heated plate corresponding to formation of the first visible particles inside the chamber was larger at larger heating rates. The heating rate was in the range from 0.3 to 5 K min<sup>-1</sup>. The found dependence of the resulting temperature of the upper plate on the heating rate was linear. By extrapolation to the zero heating rate were obtained in individual cases temperatures of the upper and lower plates corresponding at steady state to the formation of first visible particles in the chamber. To verify that both these temperatures are characterizing the critical state in the chamber experiments were performed in which the temperature gradient between the plates was fixed so that the temperature of the upper plate was by about 1 K lower than the temperature obtained from extrapolation to the zero heating rate. Both plates were kept at constant temperature for about 30 min. During this time no condensation in the chamber was observed. From thermal conductivity of the solid benzoic acid and from the thickness of layers on both plates corrections were calculated on temperature of both surfaces of benzoic acid. Correction on temperature of the surface of benzoic acid on the bottom plate has not exceeded in majority of cases the accuracy with which the temperature was measured and was thus neglected. Correction on the surface temperature of the evaporating layer of benzoic acid was lower than 0.3 K.

TABLE I  
Experimental Results

Experiment	$T_1$ , K	$T_2$ , K	$P$ , Torr	Chamber height mm
1	262.2	326.8 ± 1.1	800	43
2	262.0	325.6 ± 0.3	800	43
3	273.6	337.1 ± 0.4	800	43
4	273.6	334.6 ± 0.1	800	21
5	282.3	346.7 ± 0.4	1 200	43
6	282.3	346.5 ± 0.5	800	43
7	282.3	346.1 ± 0.6	400	43
8 <sup>a</sup>	292.5	358.7 ± 0.5	800	43
9	292.5	357.9 ± 0.4	800	43
10 <sup>b</sup>	292.5	357.6 ± 0.5	800	43

Inert used: helium, except for <sup>a</sup> methan, <sup>b</sup> nitrogen.

Experimental results are given in Table I where  $T_1$  is the temperature of the condensing surface of benzoic acid and  $T_2$  extrapolated temperature of the evaporating surface of benzoic acid.

From temperatures of both surfaces of benzoic acid and of the corresponding saturated vapour pressures the profiles of supersaturation in the chamber were calculated for the case when the forming nuclei are crystalline and for the case when benzoic acid forms nuclei in the form of subcooled liquid. We have assumed in the calculation that the temperature profile and profile of partial pressure of benzoic acid are linear. The results of measurements with a chamber as half as high demonstrate that approximation of one-dimensional diffusion and of heat conduction is satisfactory. As the resulting profile of supersaturation depends on the accuracy with which the temperature and partial pressure profiles are evaluated we have in the experiment No 4 considered the temperature dependence of the diffusion coefficient and of thermal conductivity and the heat flow resulting from the diffusion flow<sup>36</sup>. The concentration dependence of both coefficients has not been considered for low concentrations of vapours of benzoic acid and the effect of thermodiffusion was as well considered to be negligible<sup>22-24</sup>. The resulting supersaturation profile was in this case in vicinity of maximum smaller by 3.5%. The same error was due to the inaccuracy in determination of the extrapolated temperature of the upper plate.

Here, an attempt has been made to find out if the diffusion cloud chamber is suitable for measurements of critical supersaturation of vapours of solid matters and for these reasons a simple model has been proposed. It is obvious that the used evaluation method is at the reached accuracy satisfactory.

Profiles of supersaturation in the region of maximum and their comparison with theoretical calculations are plotted in Figs 2 and 3. Benzoic acid forms nuclei from the vapour state in the form of liquid at temperature around 275 K<sup>4</sup> and thus for theoretical estimations we have assumed formations of crystalline as well as of liquid nuclei. In orientation experiments in which, because of some difficulties the quantitative evaluations could not have been made, we have observed in the chamber tracks after passage of highly energetic particles which were able to ionize the gas in the chamber and, therefore, we have also considered the catalytic effect of gaseous ions. For theoretical prediction Eqs (1) to (6) were used with the temperature dependence of physical constants for the nucleation rate  $J = 1$  and the condensation coefficient  $a = 1$ . The change in the nucleation rate or in the condensation coefficient in orders of magnitude is causing only the change of 2% in the calculated supersaturation. In crystal nuclei the spherical shape has been considered and their surface free energy has been approximated from the surface tension of liquid benzoic acid according to relations  $\sigma_s = \sigma_l$ ,  $\sigma_s = 1.1\sigma_l$  (ref.<sup>10,37</sup>). For calculation of the dependence of critical supersaturation on temperature at heterogeneous nucleation on gaseous ions by use of Eqs (3) to (6) we have considered the concentration of gaseous ions of the order  $10^3$  and the dielectric constant of liquid benzoic acid  $\epsilon = 3$

(dielectric constant of crystalline benzoic acid  $\epsilon = 2.3$  (ref. <sup>38</sup>) or 3 (ref. <sup>39</sup>)). Also the possible effect of dimerization of benzoic acid in vapour phase has been considered. Data on dimerization of benzoic acid in vapour phase<sup>40</sup> were used together with the equation for nucleation rate in which association is included<sup>41</sup>. It resulted that at the considered conditions the effect of dimerization is negligible (at 280 K and concentration corresponding to critical supersaturation 3.4% of gaseous molecules are dimerized which is increasing the calculated critical supersaturation by 2%).

From comparison of experimental results with theoretical dependences (Figs 2 and 3) can be stated that benzoic acid forms nuclei in the considered temperature range in the form of crystal nuclei and that the classical equation fits with an acceptable approximation to the specific surface free energy and the shape of nuclei relatively well with the experimental results. The independence of critical supersaturation on total pressure and on the type of inert gas (Table I) is in agreement with the theory of homogeneous nucleation. Gaseous ions are catalyzing only nucleation of vapours in the form of liquid nuclei<sup>2,3,10</sup> when the molecules in the drop are movable and can

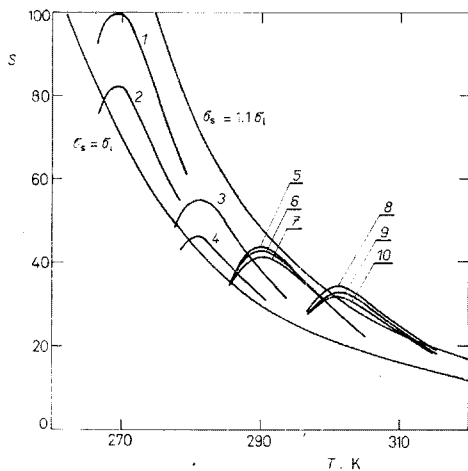


FIG. 2

Comparison of Experimental Data on Critical Supersaturation of Vapours of Benzoic Acid Corresponding to the Origin of Crystal Nuclei with Theoretical Values Calculated According to Eq. (1)

Curves are denoted in the same manner as the experiments according to Table I.

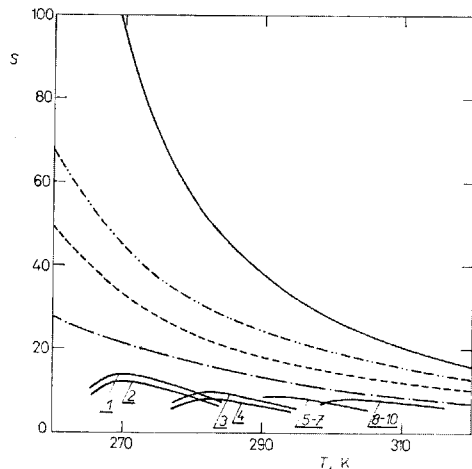


FIG. 3

Comparison of Experimental Results on Critical Supersaturation of Vapours of Benzoic Acid Corresponding to the Origin of Liquid Nuclei with Values Calculated According to Eqs (1) to (6)

— Classical equation, - - - - - Lothe-Pound equation, - · - · - · - Volmer equation, - - - - - Russel equation. Curves are denoted in the same manner as the experiments according to Table I.



be oriented around the central ion<sup>10</sup>. On basis of the made experiments in which the tracks of highly energetic particles have been observed in the chamber can be expected that at higher temperatures (in the point of maximum supersaturation estimated about 320 to 325 K) benzoic acid forms nuclei in the form of subcooled liquid and that nucleation is catalyzed by gaseous ions. By inquiry in the Astronomic Institute of Czechoslovak Academy of Sciences we have been informed that the intensity of cosmic rays was in the day and approximately in the time the measurements were performed by an order of magnitude one to two higher than usual (measured at the earth surface) and thus the observed tracks of particles can be ascribed to cosmic radiation. As these tracks were observed practically simultaneously with the origin of the first diffusively distributed particles in the point of maximum supersaturation we assume that also these particles are condensing on ions and that the possible catalytic effect of condensation centres which are produced by the surface of evaporating benzoic acid is comparable with catalytic effect of gaseous ions. The effect of gaseous ions has been observed according to the tracks after the passage of  $\alpha$ -particles produced by the preparation containing Am 241 installed in the chamber. Partial success has been reached by use of argon as inert gas with a small content of water vapours. Molecules of water vapours are there apparently acting as a stabilizer of originating ions<sup>4,2</sup>.

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