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Kinetic stability of glass states of cryocondensates of organic molecules at low temperatures

Abstract: the article provides a deep understanding of the processes and studies of cryovacuum condensates of glass-forming molecules obtained from the gas phase at low and very low temperatures. This is due to the fact that the resulting cryovacuum condensate films have unique properties that, even at a sufficiently high freezing rate, cannot be achieved by freezing from the liquid phase. In particular, it should be noted that a number of properties depend on the cryopreservation temperature, in particular, on the distance from the transition temperature to the glass. The objects of these ideal studies can be simple organic molecules such as methane, methanol, ethanol, freon 134A and 134, as well as carbon tetrachloride. At T=70 K, the transition from the amorphous glassy state to the liquid-solid cooled phase occurs, after which its crystallization in the temperature range of 75-78 K passes into the plastic crystalline state - a cubic volume-centered structure with a directionally unregulated rotating subsystem. At T = 78-80 K, the transformation of the plastic crystal into a monoclinic crystal begins, which ends at T = 83 K.

Keywords: freon, crystallization, structural rearrangement, cryovacuum condensates, glazing, IR spectra.

Introduction. In the last 10-15 years, the study of thin films of cryocondensates of simple organic molecules, such as methane [1,2], ethanol [3], freons with different compositions [4], and others, has been of interest. Understanding the important experimental and environmental significance of studying the properties of such substances, the international community has begun to study a wide range of properties of these substances in the last decade. At the same time, in this project, we plan to pay attention to the fundamental issues of cryofilm formation and the formation of their properties. In particular, we are talking about the causes that result in the formation of various glassy States of cryocondensates of organic molecules of various degrees of stability. It is known that glassy substances whose relaxation processes proceed according to the Arrhenius law are referred to as "hard" glass-forming substances ("strong" glass formers).

$$\tau = \tau_0 exp(E_a/kT) \tag{1}$$

here, τ and τ_0 are the real and ideal relaxation times, E_a is the activation energy, and T is the temperature, respectively. And processes whose characteristic relaxation time significantly exceeds the "standard" values obeying the Arrhenius law are referred to as "weak" ("fragile" glass formers). In this case, the relaxation time is described by the well-known Vogel-Fulcher-Tammann equation.

$$\tau = \tau_0 exp[DT_0/k(T - T_0)] \tag{2}$$

here T_0 is the temperature corresponding to the relaxation of an ideal gas, or the Kozman temperature. The parameter D is associated with the coefficient of weakness (stability) m by the following relation:

$$m = (DT_0/ln10T_g)(1 - T_0/T_g)^{-2} \tag{3}$$

here T_g is the transition temperature to the glass.

A simple value (m=16 or D= 100) for classic "strong" glasses, for example, for SiO2, GeO2.For organic glass the value of m is in the range of 40 to 60. It also raises questions about the properties of low-temperature glasses in important unregulated States in physics, and the reasons that determine their belonging to a particular group of "strong" or "fragile". Physical and chemical parameters that mainly determine the properties of cryovacuum condensates, including glassy and amorphous States, can be divided into external and internal conditions of cryoposition. External factors include the cryopreservation temperature and the velocity of the gas-solid boundary, i.e., the pressure of the gas phase. It is known that the temperature of the substrate determines the degree of movement of molecules and limits the search time for the minimum energy location on the sample surface [5]. In this case, the pressure determines the time after which the molecules on the surface of the adsorption layer are absorbed by the front of a monolithic sample growing in its current state [6]. However, it should be taken into account that a very high saturation pressure can lead to heating of the condensation surface due to the release of condensation heat and the final value of the layer heat transfer. As for the molecular-kinetic reasons that contribute to the formation of stable glasses, first of all, it is necessary to note the degree of internal molecular freedom of organic molecules, the activity of which increases the barrier to transition to a low-energy state with high stability during cryopreservation. In addition, according to the Ramos model [7], an important condition affecting the formation of stable organic glasses is the molecular structure of glassformers. From our point of view, the above conditions serve as the basis for conducting a comprehensive study of the processes of thermointegrated rotation of glass-forming organic media at the low temperatures planned in the project. Alcohols and freons with different molecular structures were selected as the studied substances [8, 9]. For example, if Freon 134 (CHF2-CHF2) is a symmetric version of the molecule, then its freon isomer 134A (CF3-CH2F) reflects the molecular structure of the molecule. After analyzing the data obtained for these molecules, we can draw certain conclusions about the processes of glazing of these substances in the structure of the molecule. At the same time, if we study the effect of the film growth rate on the glazing and relaxation processes, we can understand the role of moving phase boundaries in the parameters of the transition temperature to glass and relaxation. Thus, this project is intended for experimental study of the properties of thin films of cryovacuum condensates of glass-forming organic molecules and their formation processes, as well as thermointegrated structural-phase transformations and relaxation processes at low sample temperatures. The objects of research are alcohols (methanol, ethanol) and freons (CCl4, CF3-CFH2, CHF2-CHF2). To achieve these goals, we use the method of cryovacuum condensation of samples cooled from the gas phase to a low coating temperature. It is known that this method is one of the most effective ways to obtain various and, importantly, well-controlled structural-phase States of cryoplastics. In Western literature, this method is called physical gas-phase condensation (PVD) and is widely used in the study of the properties of matter, such as density at low and ultra-low temperatures [9], polarization [8,9], optical characteristics [9-10], as well as in the search for solutions to many problems of astrophysical and astrochemical research [11, 12].

Experiment. An additional incentive for comprehensive research is the results of our previous studies of the cryopreservation processes of alcohols (methanol and ethanol) and freons (Freon 10 and Freon 134A). Data of considerable interest have been obtained. In particular, the refractive indices of ethanol were measured. The relationship between these parameters and the temperatures of the structural-phase rotation of these substances is established. Figure 1 shows that in the region of condensation temperatures T=70 K and T=90 K, the random measurement error immediately increases without any external cause, which is reflected in the significant scattering of experimental data.

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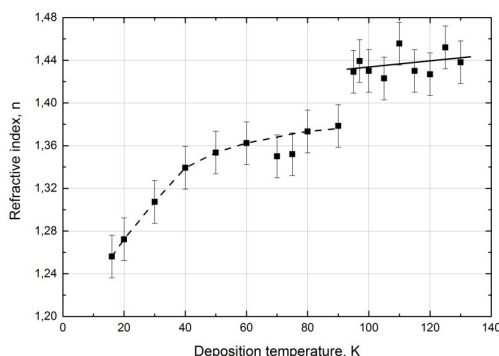50

Figure 1 – Influence of condensation temperature on the refractive index of thin films of cryovacuum ethanol condensates

A further increase in the condensation temperature from T=90 K is accompanied by an increase in the refractive index to the value n=1.430 at T=98 K, the average value equal to n=1.352 at T=90 K.

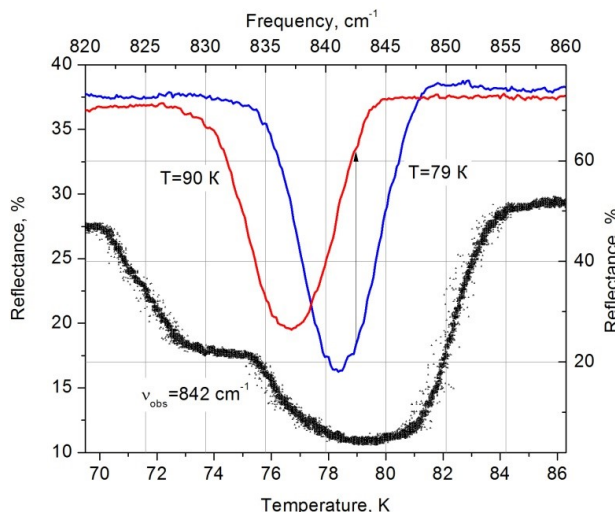Figure 2 – V_{obs} =842 cm-thermogram (lower curve) at the control frequency 1 and the corresponding change in the absorption band V7 modes of the freon 134a molecule when the sample is heated from 79 K to 90 K

Figure 2 shows data for the vibrational mode V7 of the freon 134a molecule. The sample is planted at a condensation temperature T=16 K and then heated to T=90 K. The figure shows the heating thermogram (lower curve) at the control frequency Vobs=842 cm-1, as well as the absorption bands at temperatures T=79 K and T=90 K. The reference point shows a jump-like increase in the spectrometer signal based on the" red " shift of the absorption band when the film is heated from 79K to 90K. These absorption bands, according to our forecasts [13] (according to[8]), reflect the final stage of transformation related to the transition from the state of a plastic crystal to the state of a monoclinic crystal structure. An integrated approach is used to solve these problems with the simultaneous use of several experimental methods: * The method of double-loop laser interferometry is used to determine the growth rate, thickness of the cryocondensed film, and its refractive index; * The IR spectrometric method is used to obtain IR reflection spectra of films and determine the States of cryovacuum condensates based on the analysis of absorption amplitudes and band locations corresponding to the characteristics of the studied molecules in the unbound state; * The thermodesorption method is used for equivalent determination of temperature values of structural-phase transformations. In the course of numerous studies, the sample under study is formed during cryovacuum condensation from the gas phase to the substrate at low temperatures

at a condensation temperature from 10 K to 100 K and a gas phase pressure from 10-8 Torr to 10-2 Torr. The sample thickness varies from 10 microns to 100 microns [14]. The method of obtaining information is based on the analysis of the absorption amplitude of the band corresponding to the vibrations of the molecule in different States. Measurements are carried out on the installation, the scheme of which is shown in figure 3. the Main unit of the installation is a cylindrical vacuum chamber with a diameter and height of 450 mm. The vacuum chamber is pumped by a turbo-V-301 (2) turbomolecular pump connected to the chamber by a CFF-100 (3) slide vacuum cover. the sh-110 dry spiral pump is used as a pre-vacuum pump (not shown in the figure). The maximum vacuum in the chamber is brought to a value not lower than P = 10-8. The chamber pressure sensor is implemented with a broadband pressure sensor with AGC-100 FRG-700 (4). The Gifford-McMahon microcryogenic system is located in the center of the chamber, on the upper flange of which a mirror substrate (6) is mounted, which serves as a condensation surface for a mixture of nitrogen and ethanol. The lining is made of copper, and the work surface is covered with silver. The seal diameter d = 60 mm. The minimum condensation temperature is T=10 K. the temperature is measured by a silicon sensor DT 670-1. 4 using the temperature controller M335 / 20C. The thickness and condensation rate are measured using a two-day laser interferometer based on Photoelectron multipliers P25a-SS-0-100 (7). IR absorption spectra were measured in the frequency range 400 cm-1 -4200 cm – 1. The procedure experimenting is as follows. The vacuum chamber is pumped to a pressure $P = 10^{-8}$ Torr, after which the gasket is cooled from T=10-12 K to T=150 K. Then, using the drain system (10), the test gas is sent to the chamber and its working pressure (condensation pressure) is set in the range of values determined by the current experiment. The operating pressure range is from $P = 10^{-2}$ Torr to $P = 10^{-6}$ Torr. Then the process of cryopreservation of the film, controlled by a two-day laser interferometer, began. Based on the obtained interferograms, the film thickness and refractive index were calculated. When the sample thickness reaches a value from 0.5 to 30 μm, depending on the task, the gas supply is stopped and the pressure in the chamber is again set to the value $P = 10^{-8}$ Torr [9]. Next, the vibrational spectrum of the sample is measured, after which the IR spectrometer is set to the control frequency and the substrate is continuously heated. During the heating process, the interferometer signal was measured at a fixed control frequency, the change of which is a reflection of the rotations of the test sample[15].
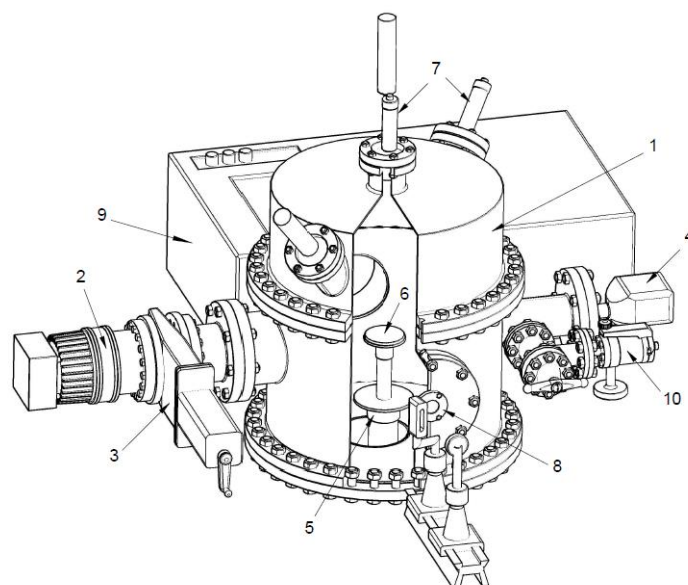

FIGURE 3 – Diagram of the experimental setup. 1-vacuum chamber; 2-Turbo-V-301 vacuum grade; 3 - FFS-100 slide vacuum bag; 4–FRG-700 pressure sensor; 5-Gifford-McMahon refrigerators; 6-toaster; 7-laser interferometer; 8-optical channels of the IR spectrometer; 9-IR spectrometer; 10-ASU system

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Results and Discussion. In figure 4, curves 1 and 2 represent the absorption bands of the mode V15 at the condensation temperature T=16 K (1) and after heating to T=90 K (2), curves 3 and 4 are thermograms at the observation frequencies VOBs=967 cm-1 and VOBs=958 cm-1, respectively. In this case, the thermogram 3 controls the beginning of the band splitting process, and the thermogram 4 is "responsible" for controlling the position of this absorption band. Comparing the data of the position of the band V15 and thermograms 3 and 4, we can come to the following conclusions. An increase in the film temperature from T=16 K to T=70 K is accompanied by a gradual shift of the band V15 to the higher frequency range ("blue" shift). At a temperature in the vicinity of T=71 K, this shift becomes sharp, as evidenced by an upward jump in the thermogram 4 and a slight drop in the values of the thermogram 3. a further increase in temperature from 73 to 77 K is characterized by a relatively stable state of the film [16]. Starting from T=78 K, a sharp shift of the absorption bands in the red region of the spectrum (falling values of thermal 4 arrow 4 down) with the simultaneous beginning of the splitting strip ?15 (increase signal images 3, arrow 3 up). At a temperature in the vicinity of T=83 K, the film state stabilizes (horizontal section of the thermogram 4), while the band splitting continues up to T=87 K (horizontal section of the thermogram 3). It is also interesting to note that the thermogram 3 undergoes a break during growth at T=81 K (insert), which can be interpreted as a change (deceleration) in the dynamics of the splitting of the band V15. At a temperature of about 100 K, the sample begins to evaporate[17].

This difference in the behavior of thermograms 3 and 4 of figure 3 may indicate that isothermal relaxation processes are carried out in the temperature range 78-85 K, one of which leads to mixing of the position of the band V15, and the other to its splitting. The presence of such processes is indicated by the data shown in figure 4, the procedure for obtaining which is as follows. The sample was condensed at T=16 K and then warmed to T =76 K (thermogram 1), i.e., to the temperature of existence, as we assume, of a plastic crystal (figure 1). Then the temperature value was set to T=76 K, and at this temperature, the sample was kept for 20 minutes. As can be seen (arrow marked as t=20 min), the signal decreased during this time, which indicates isothermal relaxations in the sample[18,19]. The question arises: if we have only a plastic crystal in this state, then, by analogy with ethanol [20] and on the basis of General concepts [21], as a result of the cooling, it is possible to transfer the sample to a state with a frozen rotational subsystem - orientation glass. However, the transition from plastic crystal to orientation glass and back should be reversible. As can be seen from figure 4, cooling from T=76 K to T=16 K (thermogram 2) leads, as can be seen from figure 1, to a slight "blue" shift of the band V7. Repeated heating (thermogram 3) differs from thermogram 2 and, in addition, demonstrates residual relaxation phenomena at temperatures above T=70 K. Based on the above, we can assume that the sample in the final state of thermogram 1 and the initial state of thermogram 2 is a mixture containing, as we assume, a liquid supercooled phase and a plastic crystal.
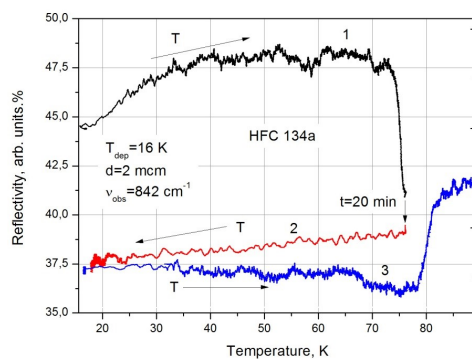

FIGURE 4 – Thermograms of changes in the position of the absorption band V7 during thermal Cycling

Conclusions. The temperature range is 16-60 K at the condensation temperature T=16 K, an amorphous state of freon 134A is formed. this indicates the Gaussian character of the shape of the

absorption bands of the main modes of vibrations of the molecule. An increase in temperature from about 50 to 60 leads to a gradual change in the spectra, which, we believe, is due to the sequential thermostimulated transformation of one amorphous state into another amorphous state, similar to the amorphous States of water [22]. As a result, an amorphous state is formed, which is formed during ultra-fast cooling of the liquid phase. The temperature range from 70 to 90 K is characterized by a number of changes, which is reflected in the position of the absorption bands. Thus, at T = 70 K, there is a sharp decrease in the interferometer signal at the reference frequency V=842 cm-1, which corresponds to the" blue " shift of the absorption band V7. in addition, at T=70 K, there is an increase in the pressure in the chamber associated with heat absorption processes. All this suggests that significant structural changes occur in the 134a freon film near the temperature T=70 k. The nature of these transformations is not yet clear, but based on the totality of the data obtained and our previous experience in studying glass conductors in water and ethanol [23,24], as well as taking into account the data [25], we make the following assumptions about the temperature of the glassy conductor and the nature of subsequent transformations. The glass temperature in thin films of cryovacuum condensates of freon 134A is TD=70 K. In addition, an important reason for this conclusion is the thermal desorption effect T=70 K. This phenomenon always occurs during the transition from the state of glass to the state of a very cooled liquid of various cryocapacitors, in particular water and ethanol.

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Төменгі температурадағы органикалық молекулалар криоконденсаттарының шыны күйлерінің кинетикалық тұрақтылығы

Аннотация. Мақалада төмен және өте төмен температурада газ фазасынан алынған әйнек түзетін молекулалардың криовакуумдық конденсаттарының процестері мен зерттеулері туралы терең түсінік берілген. Себебі, криовакуум конденсатының пайда болған қабықшалары ерекше қасиеттерге ие, оларды мұздату жылдамдығы жеткілікті жоғары болса да, сұйық фазадан мұздату арқылы қол жеткізуге болмайды. Атап айтқанда, бірқатар қасиеттер криоконсервация температурасына, Температураның әйнекке өту температурасынан алыстығына байланысты екенін атап өткен жөн. Осы идеалды зерттеу объектілері метан, метанол, этанол, фреон 134а және 134 сияқты қарапайым органикалық молекулалар, сондай-ақ тетрахлорметан болуы мүмкін. Т=70 К температурада аморфты шыны тәрізді күйден сұйық қатты салқындатылған фазаға ауысу жүзеге асырылады, содан кейін оны 75-78 К температура аралығында кристалдану пластикалық кристалды күйге бағдарлы-реттелмеген айналмалы кіші жүйесі бар кубтық көлемдік-орталықтанған құрылымға өтеді. Т = 78-80 К температурада пластикалық кристалдық моноклиндік кристалға айналуы басталады, ол Т = 83 К температурада аяқталады.

Түйін сөздер: фреон, кристалдану, құрылымдық қайта құру, криовакуум конденсаттары, әйнектеу, ИҚ спектрлері.

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Кинетическая стабильность стеклообразных состояний криоконденсатов органических молекул при низких температурах

Аннотация. В статье дается глубокое понимание процессов и исследований криовакуумных конденсатов стеклообразующих молекул, полученных из газовой фазы при низких и очень низких температурах. Это связано с тем, что образующиеся пленки криовакуумного конденсата обладают уникальными свойствами, которые, даже при достаточно высокой скорости замерзания, не могут быть достигнуты путем замораживания из жидкой фазы. Следует отметить, что ряд свойств зависит от температуры криоконсервации, в частности, от удаленности температуры перехода на стекло. Объектами этих идеальных исследований могут быть простые органические молекулы, такие как метан, метанол, этанол, фреон 134А и 134, а также тетрахлорметан. При Т=70 К осуществляется переход из аморфного стекловидного состояния в жидкостно-твердую охлажденную фазу, после чего его кристаллизация в интервале температур 75-78 К переходит в пластическое кристаллическое состояние, кубическую объемно-центрированную структуру с направленно-нерегулируемой вращающейся подсистемой. При Т = 78-80 К начинается превращение пластикового кристалла в моноклинный кристалл, который заканчивается при Т = 83 К.

Ключевые слова: фреон, кристаллизация, структурная перестройка, криовакуумные конденсаты, остекление, ИК спектры.

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