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Effect of microwave energy on dehydration process of sodium iodide used in single crystal growing

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Data on sodium iodide dehydration by thermal heating and microwave drying in vacuum are compared. The use of microwave energy has been shown to provide a higher dehydration efficiency at reduced energy consumption. The working pressure range providing the best dehydration efficiency has been determined.

Сопоставляются данные по обезвоживанию йодида натрия с помощью термического нагрева и вакуумной СВЧ-сушки. Показано, что применение СВЧ-энергии позволяет увеличить эффективность обезвоживания при сокращении энергозатрат. Определен диапазон рабочих давлений, при которых процесс обезвоживания происходит наиболее эффективно.

The intensive production development of optical and scintillation materials is related closely to wide use thereof in various branches of the national economy and fundamental scientific researches. The modern technology of scintillators on the basis of NaI and CsI crystals is inseparable from the problem of complete dehydration not only of initial raw materials but also of scintillators themselves prior to assembling, since the residual water determines the operational characteristics of the final products and the service lifetime thereof [1]. This problem is especially critical for materials on the sodium iodide basis which exhibit a high hygroscopicity and chemical activity.

The most widespread way to dehydrate the initial raw material is thermal dehydration in vacuum. An essential drawback of the resistance heating is a non-uniform warming of the salt over its volume. To get over that difficulty, the microwave (SHF) heating was proposed. The advantage of the vacuum SHF drying is the uniform material heating, reduced process duration and improved technical and economic parameters. This work deals with the SHF technology application at dehydration of NaJ salt.

The setup for sodium iodide SHF drying is schematized in Fig. 1. The salt sample to be treated (up to 500 g mass) is placed in the measuring cell, the quartz ampoule (4) being used to that end. The rotation frequency of the ampoule during the dehydration is adjusted by the block (2) so that no changes in the salt granulometry and bulk density occur and no electrostatic charge appears due to friction. The energy transfer from an SHF radiation source (6) to the moist material is provided by a waveguide (8) and volumetric resonator (3). The pressure in the system is controlled by the vacuum gauges (10) and (7).

The dehydration of samples in vacuum was studied using a setup [2] schematically presented in Fig. 2. A quartz ampoule was used as the cell. The sample mass was varied amounting to 500 g. The sample to be studied was placed into the ampoule which then was connected to the vacuum system and pumped out by a vacuum pump, simultaneously, the pressure changes in the sys-

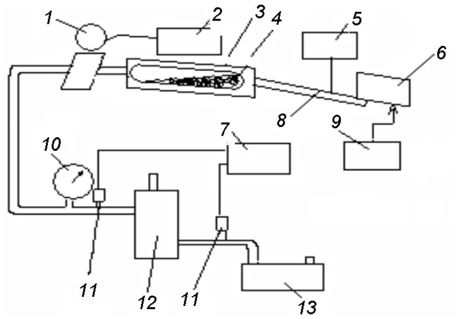

Fig. 1. Schematic view of the setup for SHF drying: 1 – ampoule rotation system; 2 – rotation frequency adjustment block; 3 – volumetric resonator; 4 – ampoule with salt; 5 – SHF measuring block; 6 – magnetron SHF generator with air cooling system; 7 – vacuum gauge; 8 – waveguide; 9 – SHF generator power supply; 10 – vacuum gauge; 11 – manometric tube; 12 – liquid nitrogen trap; 13 – vacuum pump.

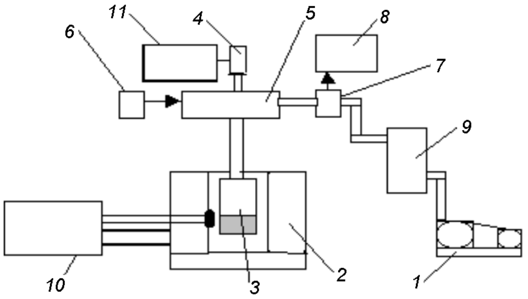

Fig. 2. Schematic view of the setup for examination of pressure change in vacuum system during the exhausting: 1 – vacuum pump; 2 – heating furnace; 3 – cell with the salt sample; 4, manometric tube; 5, absorbing cell; 6 – submillimeter spectrometer; 7 – detector; 8 – computer; 9 – diffusion pump, 10 – temperature control block; 11 – ionization-thermocouple vacuum gauge.

tem were recorded. Before the beginning of experiments, the setup was preliminary calibrated to limiting residual pressure value in the vacuum system with an empty ampoule. The limiting value is a characteristic for the specific vacuum system depending on the system design and on the vacuum pump performance. The residual pressure value remaining unchanged within 30 minutes at continuous exhausting was taken as the limiting residual pressure.

A submillimeter range radiation synthesizer of electromagnetic spectrum (6) was tuned in the step-by-step manner at ~10 kHz steps near to frequency 325152.82 MHz [3] (frequency of a the water absorption line) by a personal computer and after passage through the absorbing cell was registered by the detector (7). The detector signal was processed by computer and displayed (8). Since it is just the probing radiation frequency modulation that is used in that spectrometer to record the absorption lines of the gases under study, not the absorption line but its derivative is displayed. Besides, it is to note that as pressure in the system during the spectrum recording is $(1.0 \text{ to } 1.5)\cdot 10^{-2}$ Torr, the widening character of the water spectral line is of a mixed character and is very close to the Doppler type. In this case, the spectral line width changes insignificantly but it is its intensity that changes mainly. The furnace temperature was raised in 6 to 8°C steps with exposure no less than 3 min at each step. The temperature was measured to within ±2°C.

The heating process of a material by electromagnetic energy during the SHF drying is carried out up to pressure below which there is a SHF discharge occurs. Due to the SHF discharge in the ampoule, various undesirable processes (dispersion of a material, change of its structure, radiation-induced transformations, etc.) may occur in the raw material, therefore, it is very important to know influence such a high-density low-temperature plasmas on the salt under study [4].

In a superhigh-frequency gas discharge, the initial ionization caused by electron motion is the only electron replenishment source, therefore, this type of breakthrough is the main and most probable one. Parameters of such plasma are studied in sufficient detail [5, 6]. So, at the plasma density exceeding 10^{13} cm^3 and electronic temperature of about 5 to 10 eV (1 eV answers to approximately 11,000°C) a potential difference $U \sim 0{,}5U_{SHF}$ may arise near the wall [7]. As this potential difference appears at the plasma-ampoule interface, the ampoule surface destruction process occurs [8] due to sputtering with ions accelerated in this layer up to energy of 2 to 5 eV that will cause increased contamination of the salt with components of the quartz ampoule (according to specifications, the total impurity content should not exceed about 10^{-5} mass %). Besides, at the above-stated parameters of the SHF discharge plasma, heat flows up to about 50 J/cm^2 will arise, that, in turn, will result in local overheating of the near-surface layer of the raw material being dried, its fusion and sintering.

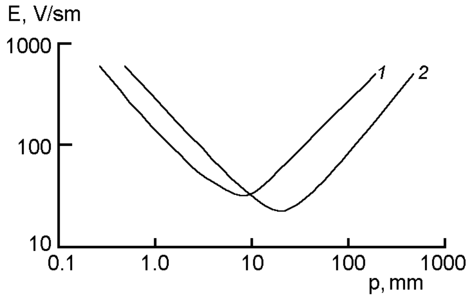

Fig. 3. Electric field strength dependence on residual pressure in the ampoule for dry air (*1*) and for water vapor releasing from NaI (*2*).

Because of poor heat conductivity of sodium iodide, the local overheat temperatures may attain 600 or even 700°. Therefore, it is very important to know conditions of the breakthrough occurrence in the residual gas at pressures near those used in the vacuum-SHF drying process.

In Fig. 3, a typical dependence of electrical (SHF) field strength on air pressure in the ampoule (4) is presented. It is seen from the Figure that the minimal field strength at which the electric discharge in an ampoule arises corresponds to the residual pressure approximately 5 or 10 Torr (curve 1). The $E = f(p)$ dependence for water vapor with a minimum $p \sim 20$ Torr (curve 2) is similar to the former. To prevent the discharge in the ampoule, the pressure ~15–20 Torr is to b maintained. The pressure in the ampoule exceeding 50 Torr results in an elevated temperature of the material being dried, increased total pressure and a rapid moisture release. This may result in a disbalance between the water vapor exhaust rate and the gaseous water release out of iodide, therefore, the salt hydrolysis becomes possible.

The advantages of vacuum-SHF drying listed above make it possible to increase essentially the raw material dehydration efficiency and rate, and also enable to carry out the dehydration at temperatures not exceeding 60–80°C. The comparative diagrams of NaI dehydration using resistance and SHF heating are shown in Fig. 4. Changes in the water contents in the raw material in both cases were determined by weight method. It follows from the data obtained that the application of SHF heating allows to reduce the dehydration process duration by a factor of about 3. Thus, the power consumption is decreased considerably. The data on dehydration in an industrial ampoule containing 10 kg of the raw material are shown in Fig. 5. It is seen from the Figure that the dehydration time of 10 kg raw material using SHF energy is about 3 h and energy consumption about 1.5 kWh.

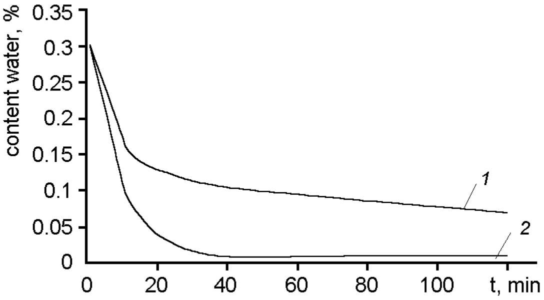

Fig. 4. Change of water content in NaI samples under resistance heating (*1*) and SHF heating (*2*) (m_{NaI} = 500 g; $W_{res} \sim 2$ kW; $W_{SHF} = 0.2$ kW).

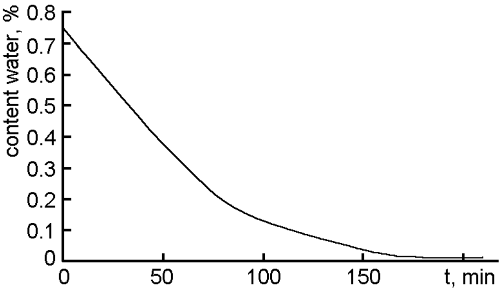

Fig. 5. Sodium iodide dehydration at vacuum-SHF drying (m_{NaI} = 10 kg, $W \sim 500$ W).

For effective drying of sodium iodide, eliminating the moisture condensation in the ampoule volume, the vacuum dehydration key rule (water vapor exhausting rate should exceed speed that of the crystal hydrate decomposition). Besides, it is necessary to have a booster volume for water vapor about 30 % of the total volume. On the other hand, the filling factor of the microwave resonator with dielectric should not exceed 0.7 to 0.8 [9], otherwise, the resonator quality will be deteriorated, thus causing a decreased efficiency of SHF energy use (Fig. 6).

Basing on the results obtained, the following conclusions can be drawn. The existence region of the stationary SHF discharge in a resonator filled with sodium iodide has been determined. Proceeding from the data on working pressure and electric field strength in the resonator, the working pressure range has been determined where the vacuum-SHF drying of sodium iodide proceeds most efficiently and at high quality.

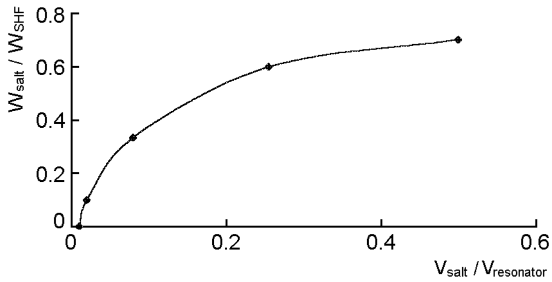

Fig. 6. Efficiency of SHF energy use depending on the resonator filling factor.

The use of vacuum-SHF drying allows to reduce the power consumption and to shorten by 3 to 5 times the time of the salt dehydration down to residual moisture <0.005 %. The dehydration sodium iodide is shown to be possible at temperatures not exceeding 60 to 80°C.

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Дослідження ефективності вакуумного НВЧ-сушіння кристалогідратів лужногалоїдних солей для вирощування монокристалів

А.Ю.Волошко, Б.В.Гриньов, В.І.Горилецький, Д.С.Софронов, О.В.Шишкін, Є.М.Кисіль

Порівнюються дані з зневоднюванню йодиду натрію за допомогою термічного нагріву та вакуумної НВЧ-сушки. Показано, що застосування НВЧ-енергії дозволяє збільшити ефективність зневоднювання при скороченні енерговитрат. Визначено діапазон робочих тисків, при яких процес зневоднювання відбувається найбільш ефективно.