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Effect of growth atmosphere composition on heat and mass transfer at continuous feed growth of scintillation crystals from melt

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Effect of the furnace atmosphere composition on the heat and mass transfer has been studied when growing CsI(Na) crystals of 300 mm dia. under control of the crystal-melt interface (CMI) and the crystal immersion depth into the melt at the fixed cylindrical part height (60 mm). The most intense heat transfer from the crystal has been found in argon atmosphere, when the deposit amount on the upper crystal butt is the smallest that is of critical importance for the increased CMI immersion depth as compared to other atmospheres. An opposite result has been obtained in helium atmosphere both in the heat transfer from the crystal to the furnace walls and in the CMI immersion depth. In the mixed gas atmosphere, the result was found to be, as expected, intermediate between two preceding cases in all the parameters measured.

Рассмотрено влияние различного состава атмосферы в печи на тепло-массоперенос при выращивании монокристаллов CsI(Na) диаметром 300 мм при контроле формы фронта кристаллизации (ФК) и измерении его глубины погружения в расплав на фиксированной высоте цилиндрической части кристалла — 60 мм. Установлено, что в атмосфере с аргоном наблюдается наибольший теплоперенос от выращиваемого кристалла при наименьшем количестве осевшего на верхнем торце кристалла конденсата расплава, что является определяющим в увеличении глубины погружения ФК в расплав в сравнении с другими вариантами атмосфер. При выращивании кристаллов в атмосфере с гелием результат оказался противоположным как по теплоотдаче от кристалла к стенкам печи, так и по глубине погружения ФК. Результат в атмосфере из смеси двух газов, как и следовало ожидать, оказался усредненным по двум предыдущим вариантам по всем измеряемым параметрам.

The gas medium composition is an important factor in heat transfer process at continuous growth of large alkali halide crystals [1–3]. Alkali halide crystals are used in scintillation (CsI, CsI(Na), CsI(Tl) and NaI(Tl)) and optical devices (KCl). Nowadays, scintillation elements for high energy physics have dimensions up to 300 mm, IR-optical windows attain the diameter 500 mm and more [1–3]. This requires growth of large crystals. The process of growth takes several days. The growth from melt is accompanied by intense evaporation of melt components and deposition thereof on the crystal and inner parts of growth furnace. The evaporation intensity depends on the growth atmosphere composition. The deposit layer thickness during the growth can reach several millimetres. The melt evaporation impedes visual control of the crystal-

lization; moreover, the deposit affects strongly the heat transfer conditions.

The melt evaporation can be suppressed using an excess pressure. Such conditions provide a decreased melt evaporation and prevent atmosphere air inflow. However, alkali halide crystals can be grown only in inert gas atmosphere (Ar, He, N) in a forevacuum. Increase of gas pressure above 35 Torr results in a more intense evaporation (NaI(Tl)), or in strong convection flows in the melt (CsI) making stable growth impossible. Thus, growth at excess pressure is impossible for alkali halides, versus many other types of crystals.

At present, CsI(Na) crystals are grown in ROST type units in atmospheres of pure Ar or He at 30 Torr pressure. The aim of this work is to study the growth atmosphere influence on deposit formation, heat and mass transfer conditions in the furnace, as well as equilibrium crystal-melt interface (CMI) shapes at growth in different atmospheres: conventionally used of argon/helium mix at a ratio of 1:2.5 (argon partial pressure (PP_{Ar})≈10 Torr, PP_{He}≈20 Torr); argon (PP_{Ar} = 30 Torr); helium (PP_{Ar} = 30 Torr). Nitrogen influence was not studied in this work due to the gas insufficient purity and its specific interaction with the melt.

Two series of crystals of 300 mm dia. and 60 mm length were grown in the abovementioned atmospheres at growth speeds of 2 and 4 mm/h. The grown ingots were torn off from the melt to save the current equilibrium shapes of crystal-melt interface. The plates of 10 mm thickness were cut from the crystals to evaluate the CMI shape and the undermelt crystal volume. The deposited condensate layer was harvested separately from crystal and different parts of furnace inner surface, and weighted. The crystal surface temperature was measured using a thermocouple and a pyrometer [4]. Temperatures of bottom (t_{bot}) and side (t_{side}) heaters were registered simultaneously. The t_{bot} parameter is varied by control system to maintain the constant crystal diameter. It decreases by 50–70°C at the start of crystal growth, and then increases by about the same value during the growth till 500 mm crystal length. The crystals were torn off from the melt at H_{cyl} = 60 mm, i.e., just after t_{bot} minimum [5]. The equilibrium CMI shapes were registered. The constant pressure in the furnace was controlled by vacuum gauge.

The t_{bot} dynamics in time (τ) reflects the heat transfer intensity between the growing

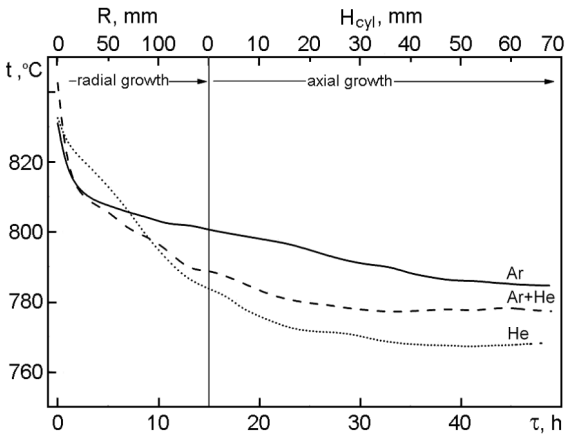

Fig. 1. Temperature vs. growth time and crystal size (upper scale) at CsI(Na) growth of 300 mm dia. at various atmosphere compositions. Pulling rate 2 mm/h.

crystal and furnace walls. The $t_{bot}(\tau)$ dependences for crystals grown in different gaseous atmosphere compositions are presented in Fig. 1. The t_{bot} values and their dynamics were found to be substantially different. These differences were attributed before to the deposit amount on the crystal upper butt [5, 6]: the more the condensate amount, the more intense is the heat transfer, and the more heat should be supplied to provide stability of the CMI.

It is seen that t_{bot} at the growth completion in Ar atmosphere exceeds by ≈20°C that in He atmosphere. Correspondingly, the smallest deposit amount is observed in Ar (see Fig. 1). The results obtained for mixed atmosphere were found to be intermediate as compared to the pure gases. The observed trend of the curves is an experimental evidence of the deposit influence on heat transfer in the furnace at radial growth. Herein, the $t_{bot}(\tau)$ trend becomes identical at cylinder growth, because the deposit amount on side surface is the same in all the atmospheres, and heat transfer therethrough increases identically. Thus, the atmosphere composition influences the heat transfer from the crystal, and $t_{bot}(\tau)$ curve trend. The condensate amount on different surfaces under different atmospheres can be evaluated by the collected deposit mass per unit area (Table 1).

The largest unit area mass of the deposit is observed in He on the "hot" surfaces (crystal upper butt and crystal holder), in contrast to the "cold" furnace walls. The limited area of the crystal side surface made it impossible to observe a strict de-

Table 1. Deposit mass distribution in the furnace at CsI(Na) growth in various atmospheres

Atmosphere	Unit area weight of deposit on the crystal upper butt, g/cm^2	Unit area weight of deposit on the furnace walls, g/cm^2	Unit area weight of deposit on the crystal holder, g/cm^2	Averige unit area weight of the condensate, g/cm^2
Ar	0.081	0.017	0.029	0.018
Ar + He	0.137	0.016	0.054	0.019
He	0.364	0.0119	0.238	0.024

Table 2. Deposit weight fractions on various surfaces at CsI(Na) growth in various atmospheres

Atmosphere	Crystal upper butt (%)	Crystal holder (%)	Furnace walls (%)
Ar	9.1	3.1	87.8
Ar + He	14.6	5.8	79.6
He	31.53	20.27	48.2

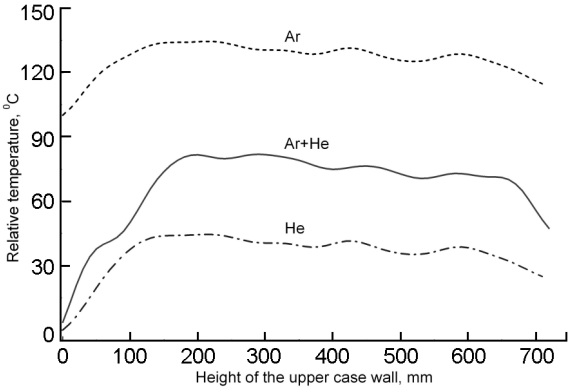

Fig. 2. Relative temperatures of the furnace wall at H_{cyl} = 50 mm at CsI(Na) growth in various atmospheres, growth rate – 2 mm/h.

pendence of the deposit amount on the growth rate. The deposit amount can be supposed to decrease as the growth rate increases. This is confirmed indirectly by the observed fact of the total deposit weight decrease by 5 %. The deposit mass distribution inside the furnace is shown in Table 2. The fraction of deposit on the "hot" crystal surface and crystal holder substantially increases when passing from Ar to He (from 12 to 52 %), and proportionally decreases on "cold" furnace walls (from 88 to 48 %).

Thus, the heat transfer in the furnace is more intense in Ar atmosphere at lesser deposit density on the crystal surface. This result also testifies that the deposit density on the furnace walls is too small to influence heat transfer thereto. Moreover, we can conclude that the heat conductivity of the gases is not of a great importance here (heat conductivity of He is 9 times higher than in Ar: $\lambda_{He} = 0.152$ W/(m·K), $\lambda_{Ar} = 0.0177$ W/(m·K)). Temperature measurements of furnace inner walls confirm this conclusion. The experiment was carried out

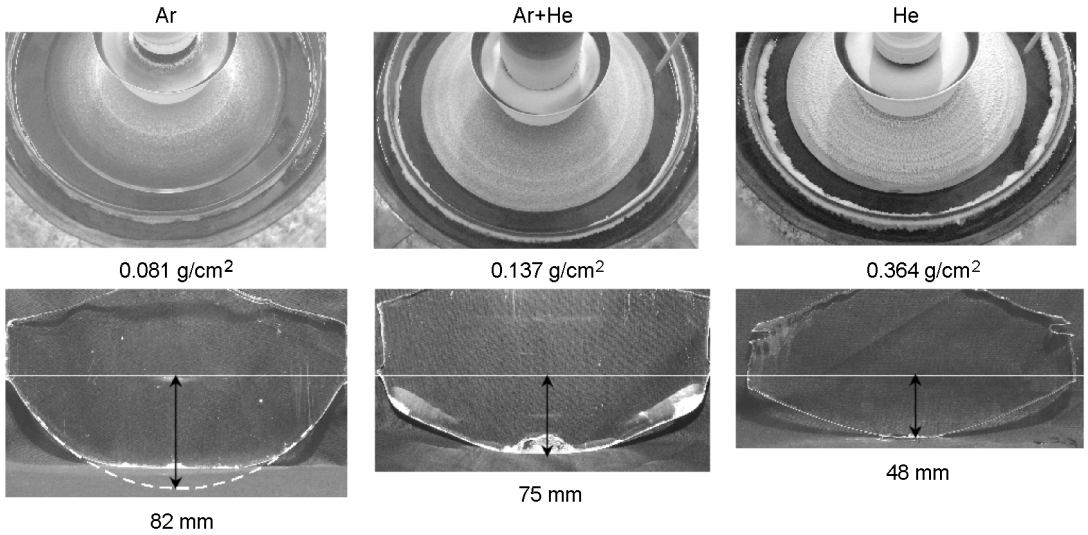

Fig. 3. Photos of the crystal upper butt and CMI equilibrium shapes in different atmospheres. The corresponding values of the deposit unit area weight and CMI depths are also presented.

at H_{cyl} = 50 mm, the zero point corresponds to the height 60 mm above the melt surface. It is seen that the highest temperatures are observed in Ar, while the lowest, in He. The difference reaches 50–100 degrees. So, the heat flux from the melt to crystal is the largest in Ar, in contrast to He. The trend of the temperature vs. height curves is similar in all the atmospheres — the lower part of the furnace wall is colder, because it is not irradiated directly by hot melt and crystal upper surface.

It is clearly seen that the CMI convexity is inversely proportional to the deposit amount on the crystal (photos in Fig. 3). The atmosphere composition influences substantially the deposit thickness on the crystal upper butt and heat transfer intensity from the crystal and to the crystal (see the $t_{bot}(\tau)$ parameter trend in Fig. 1). Consequently, the crystallization isotherm shape can be controlled by the growth atmosphere composition.

Thus, the heat transfer intensity from the CMI depends mainly on the deposit thickness on the crystal surface, and does not depend substantially on that at the furnace walls. The reciprocal dependence of CMI depth vs. deposit amount on the crystal surface has been shown experimentally. The deposit redistribution in the furnace in different growth atmospheres is probably connected with variations in convection flux pattern in these atmospheres.

The main conclusion here is that the crystallization isotherm shape can be controlled by the growth atmosphere composition, and formation of macrodefects in crystals can be prevented using this factor.

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Вплив складу газової атмосфери у ростовій печі на тепло-масопереніс при вирощуванні сцинтиляційних кристалів із розплаву безперервним методом

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Розглянуто вплив різного складу атмосфери у печі на тепло-масоперенесення при вирощуванні монокристалів CsI(Na) діаметром 300 мм при контролі форми фронту кристалізації (ФК) та вимірюванні її глибини занурення у розплав при фіксованій висоті циліндричної частини кристала (60 мм). Встановлено, що в атмосфері з аргоном спостерігається найбільше теплоперенесення від кристалу, що вирощують, при найменшій кількості конденсату розплаву, що осів на верхньому торці кристала, і це є визначальним для збільшення глибини занурення ФК у розплав порівняно з іншими варіантами атмосфер. При вирощуванні кристалів в атмосфері з гелієм результат виявився протилежним, як за тепловіддачою від кристала до стінок печі, так і за глибиною занурення ФК. Результат в атмосфері із суміші двох газів, як і слід було очікувати, виявився проміжним між обома попередніми варіантами за всіма виміряними параметрами.