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Growing of NaI:Tl crystal plates in garnissage

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A method is proposed to obtain large area crystal plates using direct crystallization in garnissage that makes it possible to omit the deformation stage of cylindrical crystal obtained by other techniques, thus shortening considerably the process duration. The technology provides the growing of crystal plates with any perimeter shape and of other substances.

Предложен метод получения кристаллических пластин больших площадей непосредсвенно кристаллизацией в гарнисаже, что позволяет исключить стадию деформации цилиндрических кристаллов, полученных другими методами и значительно сокращает время технологического процесса. По данной технологии можно получить кристаллические пластины любой формы по периметру.

Recently, there is an increased need for large area crystal plates, e.g., plate scintillators shaped as large size screens ($500\times600\times10$ mm^3) for gamma cameras. In general, the prior art technology of such scintillators includes growing of cylindrical single crystals using, for example, the technique described in [1] followed by deformation thereof [2] to obtain a large area plate. The deforming process requires complicated equipment and the crystal growing process needs the use of precious metals, thus increasing the production costs of the scintillators. The multistage process results in a high rejection rate both at the single crystal growing and deforming. It is to note also that the crystal deforming requires as much time as its growth.

To obtain the large area crystal plates immediately by directional crystallization, the axial heat removal from the crystallization front should predominate during the crystal growth. In such conditions, it is possible to provide a flat crystallization front necessary to grow a high-quality crystal and to attain a homogeneous distribution of dopant over the cross-section of the crystal in growth. The crown crystal should also be easily removable out of container. The existing techniques of crystal scintillator production using containers do not provide the desired result, since all the construction materials, including graphite being used to make container, are wettable with alkali halide melts, thus causing the adherence of the grown crystal to the container walls. During the cooling of a crystal (especially a large-area one), thermal stresses arise resulting often in the crystal failure. To remove the crystal out of container, it is necessary either to flash the crystal using a special technology (that is very difficult at large area crystals) or to break the container that is uneconomical.

An attempt was done to obtain the scintillator crystal plates by molding [3] where a liner was used to prevent the crystal adherence to the container walls. The process includes the selection of the mould, lining it, the melt pouring into the mould, cooling the plate and its removal out of the mould together with the liner adhered thereto. The method drawback consists in thet the melt crystallization rate cannot be controlled during the whole process.

In this work, the manufacturing technology of large area crystal plates in a garnissage (a "cold" crucible) is studied taking

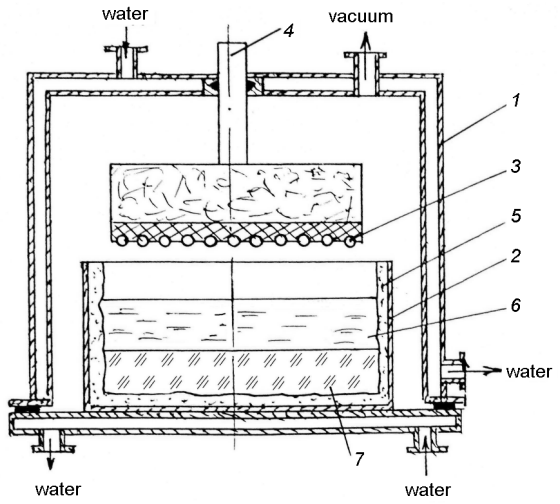

Fig. 1. Schematic view of the unit for plate growing.

the plate-shaped thallium doped sodium iodide crystal scintillators as an example. The garnissage methods are in use already for a long time both in metallurgy [4] and in crystal growth [5–7] to obtain various high-purity materials reacting with the crucible walls in melted state. In particular, the garnissage technique is used widely in single crystal growth by the Czochralsky technique. We have proposed a method to grow alkali halide crystals that makes it possible to organize the heat removal from a flat crystallization front of practically any area and to remove the crystal plate out of the container. The garnissage layer is water-soluble and is removable easily from the plate. The crystallization is carried out in a vacuum chamber where the container is placed with a heater positioned above it. The heater area should be smaller than the container one and provide a 5 to 10 mm thick garnissage layer near the container walls and bottom.

To grow a plate, the substance is placed into container having a perimeter of any shape and melted in a manner providing some unfused dispersed substance remaining near the container walls. The dopant is introduced together with the substance to be crystallized. The container can be made of a metal or other material and its walls are cooled down to a temperature lower than the construction material melting point. In our case, the container is made of aluminum having the melting point equal to that of sodium iodide. An example of the NaI:Tl plate manufacturing is presented in Fig. 1.

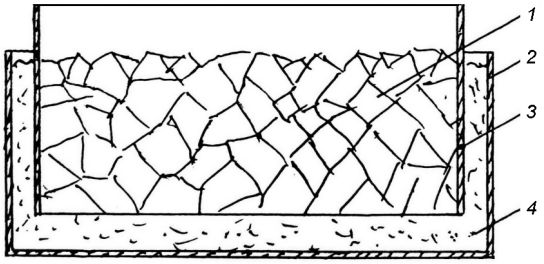

Fig. 2. Container loading with crystal debris.

The raw material (thallium doped sodium iodide powder) to be melted is loaded into an aluminum container 2. The container with the raw material is placed in the vacuum chamber 1 with cooled walls and bottom. The chamber is evacuated down to forevacuum. The salt drying and organic impurities burning-out is carried out using the known procedure. The resistance heater 3 is positioned at a distance about 10 mm above the raw material surface and its temperature is raised gradually up to the raw melting point. Since the heater 3 has a smaller surface area than the container 2, a garnissage layer 5 of about 5 mm thickness is formed at the container walls, thus hindering the melt 6 contact therewith. It is possible to provide the fusion of a thicker raw material layer by vertical displacing the heater 3 fixed to the rod 4 at a constant heater power.

After the melt 6 is formed within the garnissage 5, the heater temperature is lowered gradually at a rate providing the optimum crystallization speed for the specific substance (2–5 mm/h in our case), thus causing the crystallization of the melt 6 starting from the cooled bottom of the vacuum chamber 1. The whole crystallization process occurs in a forevacuum, thus favoring the melt de-gassing and lowering the bubble formation probability in the crystal. As the whole melt 6 is crystallized, the obtained plate 7 is annealed by lowering the heater 3 temperature at a specified rate (5–10 deg/h in our case). After the plate 7 is cooled down to room temperature, the container 2 is removed from the vacuum chamber 1. The plate 7 together with the garnissage 5 is withdrawn out of the container 2 and the garnissage layer is removed.

When a crystal debris is used as the raw material, the powdered raw 4 (see Fig. 2) with grain size of maximum 0.5–1.0 mm is first placed onto the container 2 bottom forming a layer of at least 20 mm thickness. A cowling 3 is then placed onto that

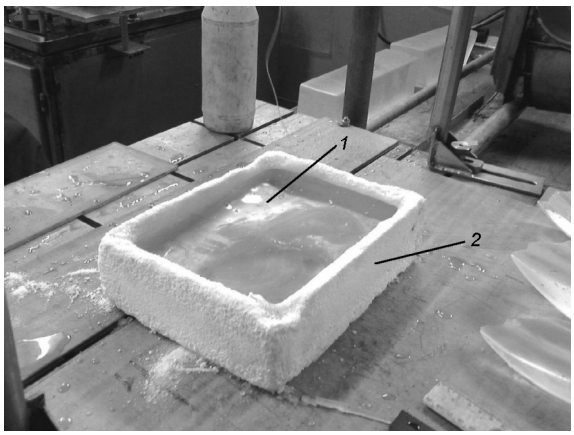

Fig. 3. Plate with garnissage.

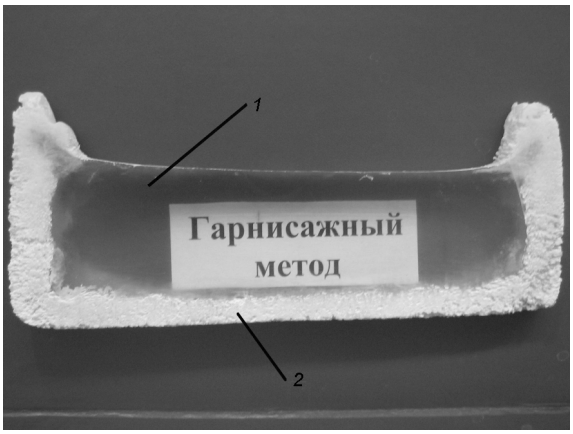

Fig. 4. Plate cross-section.

layer and loaded with the crystal fragments 1; the gap between the cowling 3 and container 2 is filled with powdered raw material 4 providing the layer thickness at least 20 mm. After the loading, the cowling 3 is removed and the raw material is melted as above. In Fig. 3, a plate in garnissage is shown and in Fig. 4, its cross-section (1, the crystal; 2, the garnissage).

Using a pilot plant including a 380 mm diameter vacuum chamber, scintillator plates of 180×270×40 mm^3 have been obtained. To determine the scintillation characteristics of the plate, detectors of 25×30 mm^2 were manufactured from its various parts. It is seen from Table that the energy resolution of those detectors is at the level of the best samples grown by pulling from melt [1] and the light yield is considerably higher. The resolution over the 180×270 mm^2 area is homogeneous to within 0.1 % and light yield, to within 0.3 conditional units (CLYU). The thallium distribution over the 40 mm plate thickness is homogeneous to within 0.001 %.

Thus, the manufacturing of large area scintillator plates by direct crystallization makes it possible to omit the deformation of cylindrical crystals, thus reducing considerably the process duration and its cost, including the rejects rate. The method proposed provides a maximum simplification of the growth unit design and minimizes the control and measuring apparatus, thus simplifying the process control. Reduced is the number of parts made from precious materials such as platinum. The technology makes it possible to manufacture the scintillators of any perimeter shape as well as plate crystals of other substances. The process provides the de-gassing of a gas-enriched melt as well the overheating of the melt in the same unit that is impossible when the scintillator is produced by pulling from melt. At the scintillator manufacturing, a considerable amount of wastes is formed, but about 90 % thereof can be used in the subsequent growth operations without any complex pretreatment.

Table . Scintillation characteristics of a plate

Sample No.	Resolution, %	Light yield, CLYU	Conc. Tl, %
1	6.3	4.4	0.056
2	6.3	4.1	0.057
3	6.2	4.1	0.056
4	6.2	4.3	0.056
Scintillation characteristics of crystals manufactured according to [1]			
1	6.5	2.5	—
2	6.3	2.6	—
3	6.2	2.7	—
4	6.1	2.9	—

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Вирощування кристалічних пластин NaI:Tl у гарнісажі

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Запропоновано метод отримання кристалічних пластин великих площин, безпосередньо кристалізацією у гарнісажі, що дозволяє виключити стадію деформації циліндрових кристалів, отриманих іншими методами та значно скорочує час технологічного процесу. За даною технологією можна отримати кристалічні пластини будь-якої форми за периметром.