The Review of Physical Chemistry of Japan Vol. 27 No. 2 (1957)

PREPARATION OF SINGLE CRYSTAL OF THALLIATED SODIUM IODIDE

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Introduction

In the course of study of the compacted scintillators[1], the single crystal of the thalliated sodium iodide was required for comparison. For this reason, the method of the preparation of the single crystal of the sodium iodide was examined and the single crystal of the thalliated sodium iodide was prepared as well as the pure crystal. In the previous paper[2], we reported on the color centers in these crystals. Now, we will report on the preparation of the single crystal and the distribution of thallium ions in the crystal.

Sodium iodide is hygroscopic and easily decomposed by heating in the humid air. On the other hand, thallium iodide volatilizes preferentially at the melting point of the sodium iodide. These points must be taken into consideration for crystallization.

R. Hofstadter[3] prepared, for the first time, the thalliated sodium iodide crystal in the form of several large grains in the quartz tube under vacuum and found that such crystals are extremely efficient phosphors.

We prepared the transparent single crystal in the pyrex glass crucible in the atmosphere of nitrogen gas by Stockbarger's method.

Experimentals and Results

About 200 grams of NaI powders or mixed powders of NaI and TlI (0.15%, 0.30%) were funnelled into the 5 cm pyrex glass crucible** with conical bottom to a depth of 13 cm and were heated slowly from the room temperature under vacuum and kept at 400°C for 1 hour. After drying, the crucible was filled with nitrogen gas of 15 cm Hg and sealed off. In order to make the crystal grow from only the point bottom, the crucible, except the point bottom, was enveloped with the tube having large heat capacity. The furnace was divided into two parts by the baffle of an insulator at which the temperature was the melting point of NaI and the vertical temperature gradient was 10~20°C/cm at the outside of the wall of the crucible. The crucible was made to descend continuously toward the part of the lower temperature with the constant rate (1.5 mm/hour) and the molten salt was crystallized. After crystallization, the descent of the crucible was stopped and the crystal was annealed for 12 hours and then cooled to the room temperature with

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1) R. Kiyama, *This Journal*, 26, 38 (1956)

2) K. Shimizu, *ibid.*, 26, 48 (1956)

3) R. Hofstadter, *Phys. Rev.*, 75, 796 (1949)

** The contamination of the crystal by Si, B and Al from the crucible could not be observed by means of the spectro-analysis.

the rate of 50~60°C/hour. The single crystal obtained by this method was 5 cm in diameter and 5 cm in total height. The crystal was transparent except at the part of 2~3 mm in thickness below the free surface. In most cases, the crystals had a plane of cleavage that was inclined at about 70 degrees to the wall of the crucible.

The concentration of Tl ions in the NaI (Tl) crystal was determined by the gravimetry of TlI insoluble in water. As shown in Fig. 1, the height of the crystal was divided into five parts (h_1~h_5) along the oblique plane of cleavage. The crystal of about 7 grams, 2 × 1 × 1 cm, was cleaved from each part, dissolved by adding water and then Tl ions were precipitated as TlI. After drying, the precipitates of TlI were weighed. The result is shown in Table 1. In the table, h_i is the average height of each part from the starting point of crystallization and H is the total height of the crystal as shown in Fig. 1. Here, C is the concentration of TlI in the part corresponded to the height, h_i and C_0 is the initial concentration of TlI. The concentration of Tl ions in the crystal increased as the height from the starting point of crystallization increased. The concentrations of Tl ions h_1~h_4 were lower than the initial concentration. On the other hand, the concentration of Tl ions in h_5 increased rapidly and exceeded the initial concentration. The fractional concentration, C/C_0, as related to the fractional height, h_i/H, in two cases (initial concentration of TlI=0.15%, 0.30%) are shown in Fig. 2. Each point in both cases was plotted on the same curve. This curve shows the distribution of Tl ions in the crystal as mentioned above. It is considered from the figure that the concentration of Tl ions in the crystal will be kept within the certain limits, if the rate of growth is diminished gradually as the crystal grows and, for this purpose, it will be useful to obtain preliminarily the curves of C/C_0~h_i/H corresponded to the various definite rates of growth.

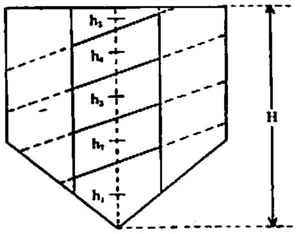

Fig. 1 Cutting of the crystal for the determination of the concentration of Tl ions in the crystal

Table 1

C_0 %		0.15		0.30	
h_i cm	h_i/H	C %	C/C_0	C %	C/C_0
h_1…0.75	0.15	0.032	0.21	0.083	0.27
h_2…2.0	0.40	0.056	0.37	0.13	0.43
h_3…3.0	0.60	0.099	0.66	0.185	0.62
h_4…4.0	0.80	0.134	0.89	0.261	0.87
h_5…4.75	0.95	0.374	2.5	0.680	2.3

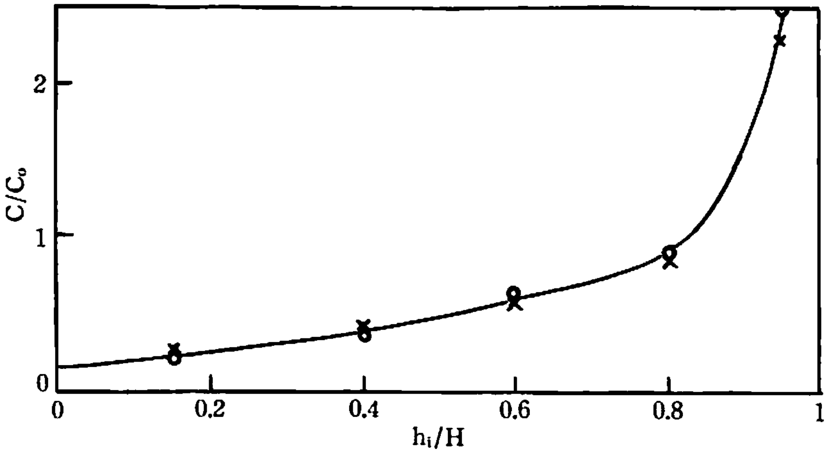

Fig. 2 Distribution of Tl ions in NaI crystal
○: 0.15% initial conc. of TlI
×: 0.30% initial conc. of TlI

The single crystal of NaI (Tl), as compared with the compacted scintillators, have much better efficiency of luminescence, and the decrease of transparency with the lapse of time does not occur, but it is difficult to prepare a large single crystal containing Tl ions at constant concentration and also a transparent single crystal containing Tl ions at high concentration.

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