# **Feature Article**

#### 50th Anniversary Product

## **Nal Scintillation Detector**

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[The Development Team (in Japan)] Back row from the left: Hiroshi Ito, Yasuo Baba Front row from the left: Yoshio Yanagida, Kyoshiro Imagawa

#### [The Development Team (overseas)]

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HORIBA started manufacturing infrared optical crystals in 1954. In the following year, development of the scintillation detector began. This detector, in which Sodium Iodide (NaI) was used, was used for radiation measurements. The infrared optical crystal is a principal part of the technology on which infrared gas analyzers are based. Gas analyzers are the company's prime products. But on the other hand, the growing of large-sized crystals has also become very successful. We grow some of the largest Sodium Iodide crystals in the World. So here we introduce crystal growth, manufacture of the scintillation detector and gamma plate, as examples of applications related to scintillation detectors for radiation measurements that have been built on the foundation of Horiba's radiation technologies.

#### Introduction

In 1948, it was reported that there was a large scintillation phenomenon caused by adding a trace quantity of Thallium (Tl) into a crystal of Sodium Iodide (NaI). HORIBA started manufacturing and development of the NaI (Tl) crystal shortly after NaI (Tl) was discovered. The development of the NaI (Tl) plate for gamma cameras started in the 1970s. In the beginning, crystals with diameters of 1 inch to 3 inches were used as detectors for nuclear physics experiments, environmental radiation measurements at nuclear power plants, or radioimmunoassay. Additionally, mosaic-type crystals were also developed to correspond to large-sized crystals in addition to the crystals with a diameter of approximately 5 inches. After the large-sized gamma camera was developed, larger and larger crystals have been demanded every year, and we have been trying very hard to get ever larger sized NaI (Tl) crystals in recent years.

### Nal (TI) Crystal

The development of the NaI (Tl) crystal for gamma cameras goes back 40 years. Currently, crystals with a diameter of 5 inches to 30 inches are in commercial use. HORIBA also succeeded in manufacturing a large-sized crystal with a diameter of 31 inches and a weight of 500 kg, and has entered into the market (Figure 1). Figure 2 shows Arizona factory where Sodium Iodide crystals are manufactured.



Figure 1 Crystal with a Diameter of 31 Inches (787 mm) and a Weight of 1100 lbs (500 kg)



Figure 2 Arizona Factory where Sodium Iodide Crystals are Manufactured

Furthermore, new crystals, NaI (Tl), Csl (Tl), and recently  $Bi_4Ge_3O_{12}$  are being developed according to requirements, but NaI (Tl) is still predominant as nothing exceeds NaI (Tl) with respect to sensitivity.

 Table 1 shows the performance of a typical crystal for a scintillation detector.

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Material	Nal(TI)	Csl(Tl)	Bi <sub>4</sub> Ge <sub>3</sub> O <sub>12</sub>
Density (g/cm <sup>3</sup> )	3.67	4.51	7.13
Time constant (ns)	230	1000	300
Luminescence wavelength (nm)	420	565	480
Relative light intensity	100	45	7-10
Refractive index	1.85	1.79	2.15

## Manufacture of Nal (TI)

After the NaI (Tl) crystal was developed in 1948, the development of the gamma plate has been active since around 1970s. For the past 30 years, enlarging crystals has been the main challenge, mainly in connection with enlargement of the gamma plate. The NaI (Tl) crystal is manufactured by the Bridgeman-Stockbarger method also known as the pulling-down method in the same manner for other alkali halides such as NaCl. This method is to fill a crucible with the particulate raw materials and melt them at a high temperature, then to gradually solidify the melt by slowly lowering it into the cooling section. Figure 3 is a diagram of the manufacturing principle, and Figure 4 shows the appearance of the furnace.



Figure 3 Diagram of the Manufacturing Principle



Figure 4 The Furnace

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Materials are particulate materials of a grade suitable for compounded scintillations. Especially, <sup>40</sup>K as it exists in nature is manufactured, paying special attention to keep it no more than 2 ppm to distinguish from background levels. Sodium Iodide has a remarkable deliquescent property; therefore, it must be stored in completely dry conditions.

So, a crucible is carefully filled with the prepared raw materials, and a trace quantity of Thallium, which is an activator of the scintillation phenomenon, is added. The crucible is placed in the crystal growing furnace, and firstly vacuum drying and dewatering are performed. Because of the deliquescent property of Sodium Iodide, this process is very important for determining the crystal's optical quality. The next process is the temperature raising process where the furnace is heated to approximately 700 °C to fully melt the materials in the crucible. Using a lift, the crucible is slowly lowered to the lower (cooling) section of the furnace where it solidifies from the tip of the crucible so as to crystallize the whole. This is the most important process, which requires prudent temperature management technology, as well as a processing period of at least 30 days.

The created ingot has a large heat capacity (mass: 240 kg to 500 kg), and it becomes a crystal ingot after a further month of annealing.

# Modifications to Function as a Radiation Detector

To improve the scintillation performance, approximately 0.1% (mole ratio) of Thallium (Tl) is added to the NaI crystal as an activator. The mechanism is that electrons created by the gamma ( $\gamma$ ) rays that enter the NaI crystal are caught by the ionized activator, and the light is emitted at the transition of the excited state. In the case of NaI (Tl), the conversion efficiency of the gamma rays is thought to be approximately 13%, and the wavelength of the light at this point is 420 nm. The light is too feeble and cannot be processed as a signal as it is, therefore the light emitted from this detector is collected by a photomultiplier to measure the radiation intensity by counting pulses. Figure 5 shows a typical scintillation detector, and Figure 6 shows the spectrum data.



Figure 5 Typical Scintillation Detector



#### Gamma Plate

Given that radioactive nuclides and their compounds when injected into the body, gather into cancer cells etc., the gamma plate detects the radioactivity ( $\gamma$  rays) emitted from those parts. The U.S. physicist Anger designed the gamma camera using this principle in the 1950s, and it is therefore called an Anger camera. Radioactive medication gathers into specific internal organs or specific cells, and the gamma rays emitted from these parts are detected and then image processing is performed. Therefore, not only abnormal parts can be detected, but also transient conditions can be observed such as cerebral blood flow or the motion of the heart.

The gamma camera components consist of a radioactive medication that emits gamma rays, a collimator, scintillation detector, photomultiplier, and signal-processing circuit (Figure 7).



Figure 7 Configuration of Gamma Camera

In the signal-processing section, the generated positions of the output signals from each photomultiplier are calculated according to the strength of the signals, and the image is processed based on the calculated data to provide diagnostic information.

#### Manufacture of the Gamma Plate

The gamma plates manufactured and used now are large; from a diameter of 220 mm and thickness of 8 mm, up to  $500 \text{ mm} \times 600 \text{ mm}$  and thickness of 9.5 mm or 25 mm. Figure 8 shows an example of a gamma plate.



Figure 8 Example of Gamma Plate (480 mm x 550 mm x 9.5 mm)

The manufacturing processes of the gamma plate is as follows:

Mechanical processing is by cutting the NaI ingot, processing the form, then grinding and polishing the surface. Assembly follows by enclosing the NaI plate in a case, and so the gamma plate is complete.

All these processes are performed in a dry room where humidity is adjusted to the low humidity or dew point of -30 °C or less because the NaI crystal material is deliquescent (Figure 9).



Figure 9 Work in the Humidity-adjusted Dry Room

JIS or U.S. NEMA standards precisely specify the basic capabilities of the gamma plate. Not only 9.5% (with <sup>57</sup>Co) or less of the energy resolution as a requirement specific to the crystal manufacturer but also in surface uniformity, spatial resolution, etc., which are required for the gamma camera system to be able to accurately conform to the user's requirements.

#### Conclusion

NaI (Tl) is used for gamma cameras, measuring environmental radiation including for radiation emission levels from nuclear reactors, analysis equipment that has been used for radiation measurement, and so on. Recently HORIBA has succeeded in purifying materials with ultralow background emission levels while paying attention to the purity of NaI, and it has turned out that the ultra-low background scintillation detector has become very useful. The time is drawing near for the NaI (Tl) scintillation detector to be used for industrial CT in addition to space physics, due to cost reductions, by using a small a multisegmented structure. Furthermore, the application development in the near future is expected for super-high performance radiation measurement systems that can measure tiny quantities of radiation, as related to the decommissioning demolition technology that will surely be needed in the nuclear power generation field.