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Characteristics of YAG Single Crystals for Electron Scintillators of STEM

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Scintillators made of powder phosphor have been usually used for the detection of signal electrons in SEM and STEM. Single crystals of YAG, developed and applied to scintillators of SEM, were studied experimentally at the accelerating voltage of 100 kV and 1 MV for scintillators of STEM and sequential EELS. Measurements were carried out for the evaluation of scintillation characteristics of YAG single crystals and conventional powder phosphors on the light intensity guided to a photomultiplier tube, resistance to electron irradiation, spatial uniformity in light intensity and the afterglow in the seconds to minutes range. The characteristics of YAG single crystals were found to be preferable to those of powder phosphors at 100 kV and 1 MV.

***Key words*: electron scintillator, YAG single crystal, P46 powder phosphor, STEM and sequential EELS, electron energy of 100 keV and 1 MeV**

Scintillator-photomultiplier detectors are most often used for the detection of signal electrons in SEM and STEM. Powder phosphors have been used as scintillators over a wide range of electron energies from about 10 keV in SEM to 1 MeV in the high voltage STEM. Single crystals of yttrium aluminum garnet activated by cerium (YAG, $Y_3Al_5O_{12}:Ce^{3+}$), on the other hand, were developed and applied to scintillators of secondary and back-scattered electrons in SEM.[1–3] It was confirmed that the YAG single crystals have a high DQE (Detective Quantum Efficiency) and a high resistance to electron irradiation up to an electron energy of about 20 keV, in addition to the general features of high transparency to light, mechanical strength to be machined into a variety of shapes, easy processing of surfaces, for example, for forming reflecting and conducting layers, unnecessary additional material such as binders for the powder phosphor, and so on.

The characteristics investigated so far give us expectations that the YAG single crystals would be applied to scintillators of STEM for a wide range of accelerating voltage up to 1 MV. The YAG single crystals, therefore, were investigated at 100 kV and 1 MV and compared with conventional powder phosphors.

EXPERIMENTS

Experiments were carried out with a conventional detection system adopted in STEM. The primary electron beam excites the scintillator of disc shape directly, the light from the base surface (surface opposite to the electron incidence surface) of the scintillator is guided by a straight light guide and approaches a photomultiplier tube (PMT, Hamamatsu Photonics R268) arranged in the axis of the electron beam. Since the PMT showed a nonlinear behavior for an intense incident light, the performance of scintillators was evaluated for light incident to the PMT obtained by calibrating the output of the PMT according to the gain characteristics of the tube.

Two kinds of shapes of YAG (single crystal) discs were prepared. A disc of 0.8 mm thickness and 10 mm diameter has polished surfaces both for electron incidence and for light output. The surface of electron incidence is covered with an aluminum foil. The other discs, of 1, 2 and 3.2 mm thickness and 10, 10 and 12 mm diameter, respectively, have polished surfaces covered with dielectric reflecting layers and indium tin oxide (ITO) conducting layers on the electron incidence side, and the base surfaces for light output are matted. Scintillators of powder phosphor were prepared with P46 phosphor which had the same composition as the YAG. The powder was deposited on a glass plate with a small quantity of collodion and the surface was metal-backed with an aluminum foil. The thickness of the powder phosphor layer is 0.07 mm and 1 mm for an accelerating voltage of 100 kV and 1 MV respectively, so that the maximum light intensity is obtained.[4]

RESULTS AND DISCUSSION

1. *Light intensity*

A high conversion factor from an electron to photons reaching the photocathode of the PMT is important for noiseless detection. The light intensity, measured and shown in Fig. 1, corresponds to the relative value of the conversion factor. The YAG with a matted surface gives a higher light intensity than the polished YAG and the powder phosphor both at 100 kV and 1 MV. The YAG of 1 mm thickness gives the highest light intensity, 4 times at 100 kV and 10 times at 1 MV, as large as the intensity of powder phosphor.

It is explained by the high refractive index of the YAG ($n = 1.84$) that the matted YAG gives a higher light

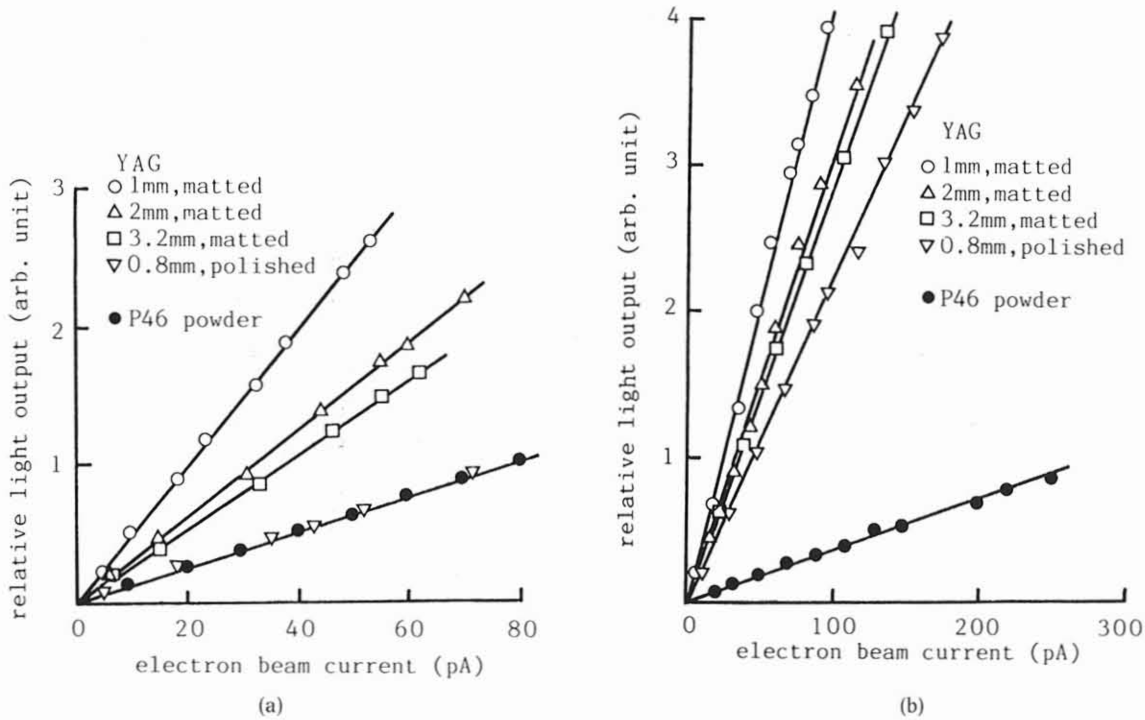

Fig. 1. Relative light output as a function of the electron beam current at the accelerating voltage of (a) 100 kV and (b) 1 MV.

intesity than the polished YAG. A part of the light incident on the polished base surface which is in contact with the material of the low refractive index is transmitted through the surface and the remainder is reflected back into the crystal. When the YAG is in contact with a vacuum, the angle within which the light is transmitted is about 33°. Therefore the light transmitted through the base surface is the sum of the lights reaching the base surface within this angle, both directly from the luminescence area and after reflection at the electron incidence surface, and is 14% of the total luminescent light.[5] For matted base surfaces, on the other hand, surfaces are randomly oriented locally and therefore the chance that the light is transmitted through the matted surface directly or after reflection from other surfaces is high, increasing the light intensity.

The light intensity obtained with the YAG of the matted base surface depends on the thickness. The effect of the thickness on light intensity is evaluated using a simplified model. It is assumed that the light is emitted isotropically at the center of the disc by irradiation of the electron beam and the light is not absorbed within the YAG. It is also assumed that the surface of electron incidence reflects all the light incident on it.

A part of the light emitted is transmitted through the side surface of the disc directly and after reflection at the surface of the electron incidence, as is indicated by the shaded area in Fig. 2. The ratio (p) of the light transmitted through the side surface to the emitted light can be evaluated from the geometry of the disc shape. The value of p calculated is 0.19, 0.34 and 0.40 for the discs of 1, 2 and 3.2 mm thickness, respectively. The light which is not transmitted through the side surface reaches the matted base surface. When the transmission rate for the base surface is q, the ratio of the light transmitted through the base surface to the emitted light is $(1-p)q$. Although q is not evaluated accurately for the matted surface, it is estimated to range between 0.25 and 0.5. It is assumed as well that the light reflected into the crystal from the base surface is transmitted through the side surface and the base surface with the same ratio as that for the light emitted at the center of the disc, and this process is repeated for the reflected light.

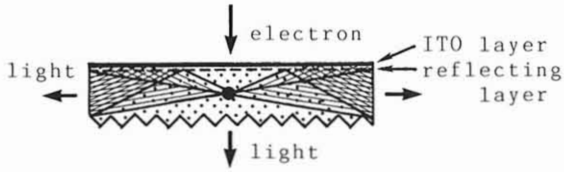

Fig. 2. Schematic diagram showing the transmission of the light through the side surface (shaded area) and the base surface (dotted area), for the YAG disc of the matted base surface.

Table 1. Ratio(T) of the light transmitted through the matted base surface to the emitted light for the transmission rate of a matted surface of $q=0.25$ and 0.5.

Disc size		T	
Thickness	Diameter	$q=0.25$	$q=0.5$
1 mm	10 mm	0.52	0.68
2 mm	10 mm	0.31	0.47
3.2 mm	12 mm	0.27	0.43

Then the total ratio (T) of the light that is transmitted through the base surface to the emitted light is given by

$$T = \frac{(1-p)q}{1-(1-p)(1-q)} . \quad (1)$$

The value of T evaluated as $q=0.25$ and 0.5 is given in Table 1. The ratio of light intensity for the YAG of 2 and 3.2 mm thickness to the light intensity for the 1 mm thick YAG is therefore 0.60 and 0.52, and 0.69 and 0.63 for $q=0.25$ and 0.5, respectively. These values are in close agreement with 0.63 and 0.54 obtained in the experiment at 100 kV. The values of 0.75 and 0.69 obtained at 1 MV are larger than the calculated values. This may be explained as follows. The 1 mm thick YAG is not thick enough to absorb sufficient energy of the 1 MeV electrons, resulting in the increased values of ratio comparatively for the YAG of 2 and 3.2 mm in thickness.

It is also possible to discuss the light intensity obtained for the YAG of a polished surface. The ratio (T) of the light transmitted through the base surface to the emitted light is 0.14 for the YAG of the polished surface. This value is 0.27 and 0.21 times the value calculated for the 1 mm thick YAG of a matted surface for $q=0.25$ and 0.5, respectively, and agrees well with the value of 0.25 obtained in the experiment at 100 kV.

It may be instructive as well to discuss the dependency of the ratio of the light obtained for the YAG to that for the powder phosphor on the accelerating voltage. Since the YAG is highly transparent to light, the light emitted in the whole area of the crystal contributes to the scintillating performance. For the YAG of a sufficient thickness within which a large part of the energy of incident electrons is absorbed, the light reaching to the base surface is proportional to the energy of the incident electrons. On the other hand, the powder phosphor has low transparency to light due to the scattering of the light at powder surfaces. The light emitted only in the area within a limited depth from the base surface, therefore, contributes to the scintillating performance. This limited depth does not depend on the electron energy. Hence the ratio of the energy absorbed in this area to that of incident electrons is small for high accelerating voltage where the electron range is large. The ratio of light intensity for the YAG to that for the powder phosphor is, therefore, more marked at 1 MV.

2. *Deterioration in luminescence*

The decrease in luminescence or the damage is experienced for STEM scintillators and CTEM imaging screens for powder phosphors. This is remarkable especially in high voltage electron microscopes.

The time variation of the light intensity was, therefore, measured for continuous irradiation of the electron beam. An example is shown in Fig. 3 for a 100 pA electron beam of 100 μm diameter at 1 MV. The light intensity decreases to 87% of the initial value in 26 min for the powder phosphor. The YAG, on the other hand, does not show a recognizable change in light intensity in 22 min. The decrease in light intensity for the powder phosphor may be caused by an additional absorption of the light due to the change of collodion used as binders.

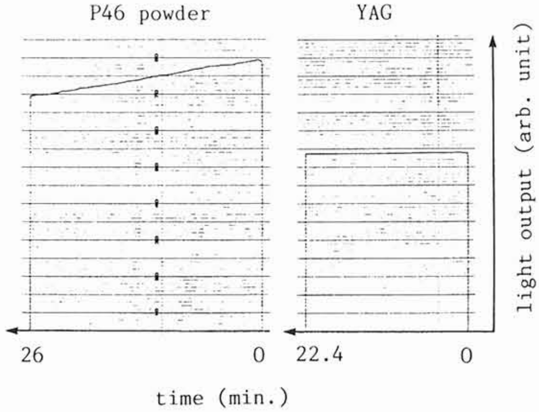

Fig. 3. Time-variation of the light output for irradiation of the 100 pA electron beam of 100 μm in diameter at the accelerating voltage of 1 MV.

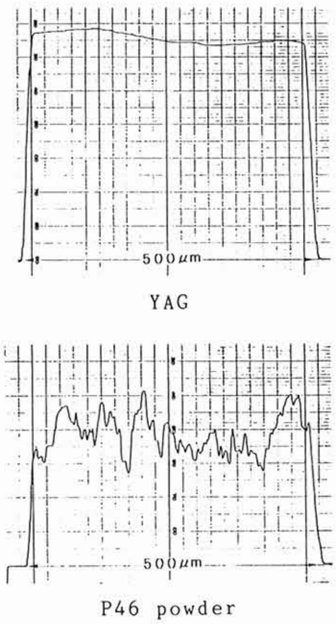

Fig. 4. Spatial variation of the light output obtained by scanning the electron probe of 10 μm in diameter on 500 μm area of the scintillator.

3. *Uniformity in light intensity*

Spatial uniformity in light intensity is sometimes required for a variety of detection modes of signal electrons. An electron beam of 10 μm diameter was scanned on the scintillator and the spatial variation of the light intensity was measured in order to study the uniformity of light intensity. An example is shown in Fig. 4. Although the powder phosphor shows a local variation of light intensity of about 25% due to the granularity of the powder, the YAG shows only a slight variation.

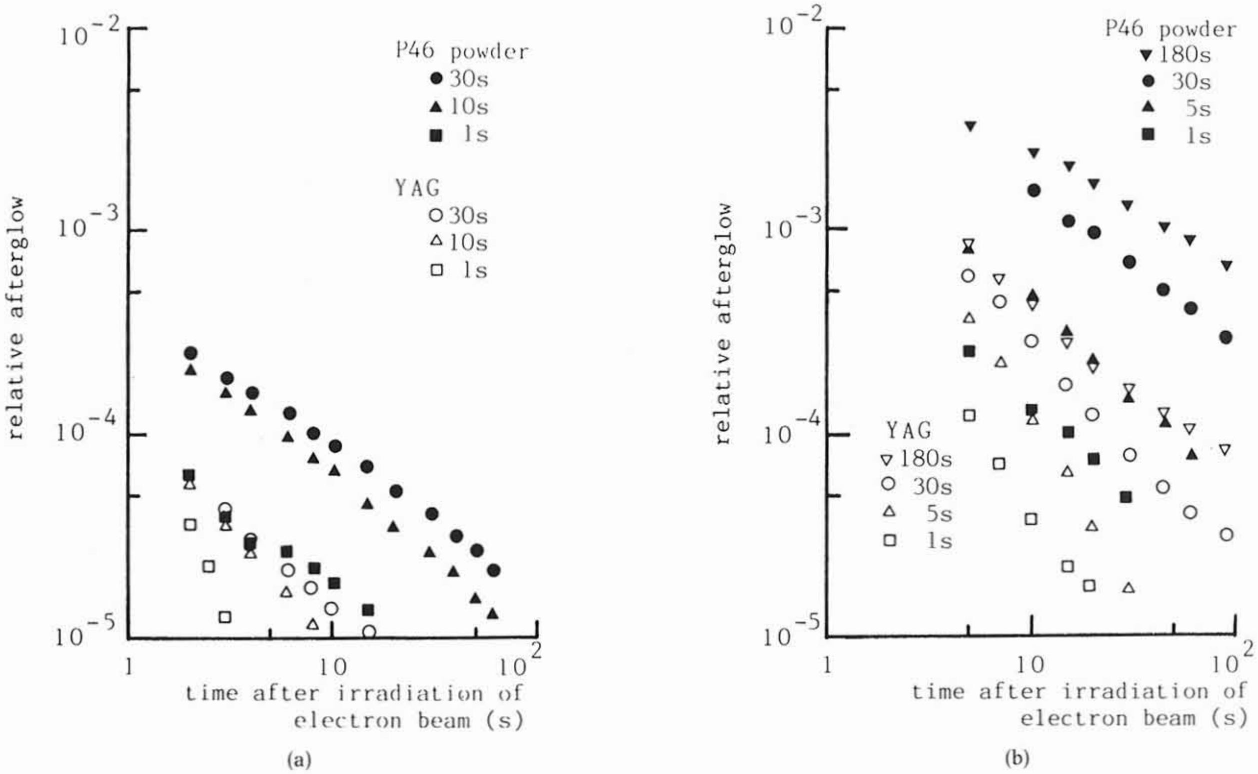

Fig. 5. Relative afterglow after irradiation of a 30 pA electron beam for various periods at (a) 100 kV and (b) 1 MV.

4. Afterglow

The afterglow excited by electron beam irradiation forms the background on the spectrum of sequential electron energy loss spectroscopy (EELS). The influence of the afterglow is large at high voltages, leading to a reduced peak to background ratio of the spectrum. The afterglow, therefore, lowers the accuracy of qualitative and quantitative analysis.

The relative intensity of the afterglow to the light intensity under electron irradiation was measured as a function of the time after the irradiation of the electron beam had ceased. In Fig. 5 we have shown the afterglow for the irradiation of the 30 pA electron beam for the periods of time indicated. It is seen from the linear relation in logarithmic plots that the decay of the afterglow follows the power-law both for the YAG and the powder phosphors. The mechanism of the afterglow, therefore, may be of the recombination type; electrons released from luminescent centers are captured in traps, released again into the conduction band thermally and then recombine with the excited luminescent centers with the emission of photons.

The relative afterglow is less for the YAG compared with the powder phosphor by a factor of 1/2–1/10, largely depending on the time after the irradiation of the electron beam has ceased. This is due to the difference of the power of decay property and the factor is less the longer the time after irradiation has ceased. The polycrystalline boundaries of the powder phosphor may act as defects which are origins of electron traps for the afterglow of recombination types and therefore the afterglow for the powder phosphor is larger than the YAG.

It is also found in Fig. 5 that the relative afterglow at 1 MV is ten times or more as high as that at 100 kV, both for the YAG and the powder phosphor. This may suggest that the irradiation of high energy electrons produces some kind of crystal defect, resulting in the increased traps.

CONCLUSION

Discs of YAG single crystals were investigated for scintillation of electrons at 100 kV and 1 MV and were compared with the conventional P46 powder phosphor. The experimental results are summarized as follows.

(1) The interface between the YAG (single crystal) and the light guide is important for the light to be guided to the outside of the YAG, since the YAG has a high refractive index. When the matted base aurface is used for the light output, the YAG gives a high light intensity. The YAG of 1 mm thickness gives a light intensity, 4 times at 100 kV and 10 times at 1 MV respectively, as large as that of the powder phosphor.

(2) The YAG shows a high resistance to electron irradiation even at 1 MV where the damage or the deterioration in light intensity is considerable for the powder phosphor.

(3) The YAG shows better spatial uniformity in light intensity compared with the powder phosphor which gives a variation of light intensity of about 25% due to

the granularity of the powder.

(4) The YAG shows less afterglow than the powder phosphor by a factor of 1/2 to 1/10, depending largely on the time after the irradiation of the electron beam has ceased. The scintillator of less afterglow is specially preferable for sequential EELS at high voltages where the influence of the afterglow is large.

Thus the YAG single crystal shows the characteristics for scintillation of electron beams which are preferable to a conventional scintillator made of powder phosphor, both at 100 kV and 1 MV.

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