LUMINESCENCE CHARACTERISTIC OF AlN CERAMICS UNDER REACTOR IRRADIATION

Quanli Hu, Satoru Tanaka and Toshiaki Yoneoka
(Department of Quantum Engineering and Systems Science)

Abstract

A broad emission peak (around 370 nm), which had a different position for AlN samples with different oxygen concentration, was found. The intensity of luminescence emission had closed relation with reactor power (neutron flux) and temperature. Influence on emission behavior from radiation defects (vacancies and interstitial) and intrinsic defects (oxygen-related-complex-defects) was discussed.

1 Experimental

The polycrystalline samples of AlN (AlN-a (99 wt% AlN + 0.9 wt% O), AlN-b (94.9 wt% AlN + 4.3 wt% Y_2O_3)) were provided by Tokuyama Co., LTD of Japan. Before irradiation the sample was cut into pellets (diameter: 10mm, thickness: $1 \sim 10$ mm), and then annealed at 973K for one hour in an air. The sample was irradiated in a research reactor YAYOI of the University of Tokyo with 1.3 MeV fast neutron at room and high temperature. The ^{60}Co γ-ray source was used to irradiate AlN ceramics with the dose rate of 0.72$\sim$4.50 KGy/h in the air. Under irradiation, the luminescence from the sample was guided through an electrically polished aluminum tube, a lens (diameter of 2 cm) and an optical fiber, then was received and treated by a monochromator and a photon-counting system. The detail about measurement of thermoluminescence after γ-irradiation was explained elsewhere [1].

2 Results and discussion

Fig.1 shows that two broad peaks of luminescence emission are observed at 370 nm and 350 nm for AlN-a and b under reactor irradiation (power: 250 W, or neutron flux: 3×10^{10} n/(cm$^2\cdot$ s)), respectively. The intensity of 370 nm emission peak increases proportionally with the reactor power (or neutron flux). Fig.2 shows the emission intensity variation with temperature (300$\sim$900 K) for AlN-a under reactor irradiation (power: 1000 W). The intensity of luminescence emission peak (around 370 nm) decreases with the increasing of temperature from 300 to 900 K. AlN-b exhibits the similar behavior of luminescence emission with reactor power and temperature. The luminescence emission is produced during the electronic de-excitation of the electronically excited level at intrinsic and radiation defect sites. Here the intrinsic defect mainly means the oxygen-related-complex-defect produced by manufacturing. During fast neutron irradiation, the electrons trapped in N ion vacancies are excited through obtaining energy, and return to ground state emitting energy partly in the form of photon. In this case, the increase of reactor power (flux) causes an enhancement of density of irradiation damage and production rate of electronic excitations in AlN, and finally leads to increase of an intensity of the luminescence emission. This kind of emission characteristic has strong temperature dependence under reactor irradiation. The higher temperature increases vibration of atoms of AlN lattice, and decreases the value of cross point for two energy band curves (ground and first exciting states). Therefore, the non-radiative decay from first excited state to ground state is enhanced. This decreases the possibility of excited electrons returning back to ground state accompanied by a photon emission [2]. It is possible to get useful information about intrinsic defect though in-situ luminescence measurement underγ-ray irradiation by ^{60}Co source and thermoluminescence afterγ-irradiation. From the results of in-situ luminescence measurement

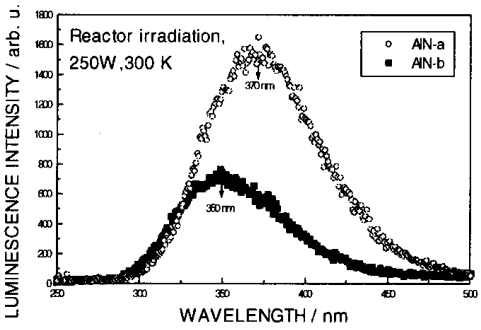

Fig.1: Luminescence spectra of AlN-a and b under reactor irradiation at 300 K.

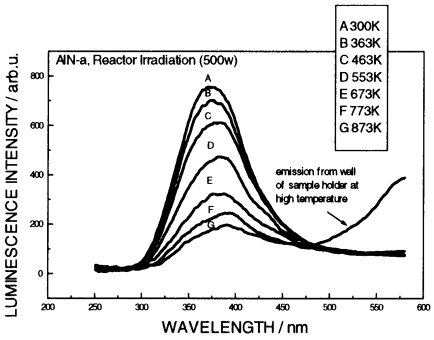

Fig.2: Luminescence spectra of AlN-a at high temperature (300 - 873 K).

underγ-irradiation (0.72 kGy/h), we can find apparent difference in peak position between AlN-a (two emission peaks at 405 nm and 620 nm) and AlN-b (weak broad band from 200 to 600 nm). We consider this difference would be reflected in the emission spectra of reactor irradiated AlN-a and b. In another word, in Fig.1, the broad emission band for AlN-a should be a complex band at least including an influence from intrinsic defects. And the position of emission band for AlN-b should mainly come from the radiation defects and would not or slightly be changed by the weak broad emission from intrinsic defects. In order to study the intrinsic defects further in AlN-a and b, the thermoluminescence measurement was conducted. The detailed works were done by M. Benabdesselam et al. [3] and by us [1]. The general description about the spectra of thermoluminescence is as follows. For AlN-a, two emission peaks (at 450 and 600 nm) are found, and the intensity of 450 nm peak is higher than that of 600 nm peak. The two different intrinsic defects are assigned to two oxygen-related-complex-defects: V_{Al}-$2O_N$-2N and V_{Al}-O_N-3N (V_{Al}: aluminum vacancy; O_N: oxygen anti-site; N:nitrogen). In another word, V_{Al}-$2O_N$-2N and V_{Al}-O_N-3N dominate in AlN-a and b, respectively. The further works will be continued focusing on quantitative relation of the above process.

3 Conclusions

A luminescence peak around 370 nm was observed for AlN ceramics with in-situ luminescence measurement under reactor irradiation. A difference of emission peak position was found for two kinds of AlN ceramics with different oxygen concentration. The intensity of luminescence emission for reactor irradiated AlN ceramics increases with reactor power, but decreases with the increase of temperature from 300 to 900 K. This emission spectra is suggested to be a combined emission band originated from radiation defect and intrinsic defect (oxygen-related-complex-defect).

References

[1] S. Tanaka, Q. Hu, V. Grishmanov and T. Yoneoka, Nuclear Instruments and Methods in Physics Research B, 141 (1998) 547.

[2] B. Henderson and G. Fimbusch, Optical Spectroscopy of Inorganic Solids, Clarendon Press, 1989.

[3] M. Benabdesselam, P. Iacconi, A. Lapraz, P. Grosseau and B. Guilhot, J. Phys. Chem. 99 (1995) 10319.