

Crystal Structure Analysis of the Cordierite Ceramics by X-ray Diffraction Method using Synchrotron Radiation

Hiromi Seki* (4468), Katsumasa Yasukawa (4308), Shimotsuma Yasuhiko (4687), Etoh Kazuya (5016), Kenichi Kato¹ (4097), Eiji Nishibori² (3112), Masaki Takata² (3167), Makoto Sakata² (4097)

R&D Center Kagoshima, Kyocera Corporation, ¹JASRI, SPring-8,
²Department of Applied Physics, Nagoya University

Introduction

There are two cordierite ($2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$) phases in nature; cordierite (high form cordierite) and indialite (low form cordierite). The cordierite is well known as a low expansion material, but the relation between the crystal structure and material properties for dense stoichiometric ceramics has not been clarified yet. Thermal expansion properties are very important subjects to study, correlating with the crystal structure. In the preceding experiment (2000A0272-ND-np), the peak split originated from the phase transition was clearly observed by the synchrotron radiation. In this work, our purpose is to investigate the mechanism of the low expansion property of the cordierite material from a viewpoint of crystal structure, using the synchrotron radiation.

Experimental

The powdered specimens were placed in quartz capillaries of $0.3\text{mm } \phi$, and applied monochromatic radiation of wavelength about 1.0 \AA . The diffraction data were taken by using a large Debye-Scherrer Camera with Imaging Plate (IP) at BL02B2 station. The measurement temperatures were several points between minus 150°C and 700°C .

Results and Discussion

We have distinguished X-ray diffraction (XRD) patterns of high form cordierite between 100°C and 700°C , and performed Rietveld analysis with RIETAN program. The Rietveld analyses were carried on the basis of hexagonal (space group: $P6/mcc$ symmetry [1]). We have evaluated axial thermal expansions from obtained lattice constants. Figure 1 shows a-axial and c-axial thermal expansions of high form cordierite. The

thermal expansions of both axes exhibited small values. As the temperature increases, a-axial thermal expansion increases. On the other hand, that of c-axis remains stable near zero.

These results clearly show an expansion anisotropy. The behavior is a widely known characteristic of a low thermal expansion material.

The phases of high and low form cordierite of dense ceramics were distinguished by the synchrotron radiation, and then the temperature dependence of the lattice constants of high form cordierite was accurately evaluated. Further analyses are in progress.

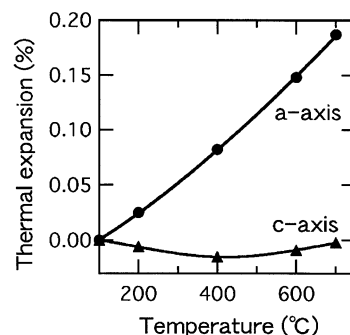


Fig.1 Axial thermal expansions of high form cordierite.

Reference

[1] E. P. Meagher, G. V. Gibbs, Canadian Mineralogist., 15, 43-49(1977).

Molecular Orbital Ordering in Ammoniated Alkali Fullerides

T. Takenobu¹/4910, M. Miyake¹/5326, Dam. H. Chi¹/4671, T. Itou¹/6378, Y. Iwasa¹/4907,
E. Nishibori²/3112, M. Takata²/3167 and M. Sakata²/3119

¹JAIST, Tatsunokuchi, Ishikawa 923-1211 Japan

²Department of Applied Physics, Nagoya University, Nagoya 464-8062, Japan

Recently, we have succeeded in synthesis of isostructural series of $(\text{NH}_3)_A\text{C}_{60}$ type compounds, where A site is the mixture of K and Rb ions. They are synthesized by intercalation of neutral ammonia molecules into the $fcc\text{-}A_3\text{C}_{60}$, which are well known as superconductors. The structure of $(\text{NH}_3)_A\text{C}_{60}$ is very similar to that of $A_3\text{C}_{60}$ except for a slight orthorhombic distortion induced by ammoniation. Interestingly, despite the similar crystal structure, the electronic ground states of ammoniated samples are an antiferromagnetic insulator with transition temperature, T_N , which varies from 40K to 76K, depending on the Rb concentration from 40 to 76 K. This is the highest T_N among molecule based magnets without magnetic elements.

$(\text{NH}_3)_A\text{C}_{60}$ type materials show not only magnetic transitions but also structural phase transitions at T_S , where the rotation of $A\text{-NH}_3$ group and C_{60} simultaneously stops, associated with a doubling of this unit cell. T_S is almost 150 K for all ammoniated samples, in striking contrast with T_N . Importantly, across T_S , both C_{60} molecules and $A\text{-NH}_3$ rotors are ordered in an antiferro(electric) fashion, which is directly correlated to the spin structure below T_N . In other words, the magnetic structure of the AF state is controlled by the orientational ordering of C_{60} .

Recent magnetic study in this type of materials reveals interesting phenomenon. T_N of $(\text{NH}_3)\text{KRb}_2\text{C}_{60}$ drops from 76 K to 40 K under quenching protocol. Similar reduction of T_N under rapid cooling has been observed in all ammoniated samples except $(\text{NH}_3)\text{K}_3\text{C}_{60}$. This result indicates that molecular orientation

is disordered in the case of rapid cooling. In this work, we have investigate cooling rate dependence of crystal structure in $(\text{NH}_3)_A\text{C}_{60}$ compounds and clarify C_{60} ordering state in crystal.

Because the cell doubling is one of the evidence for the orientational ordering of C_{60} , we paid attention to superlattice reflections under quenching protocol. Figure shows the x-ray diffraction patterns of $(\text{NH}_3)\text{Rb}_3\text{C}_{60}$ at room temperature, 90 K with slow and rapid cooling procedures. Absence of strong superlattice reflections under quenching processes suggests a random orientational state of $A\text{-NH}_3$ group and C_{60} molecules. These results, combined with the reduction of T_N under rapid cooling, provide evidence for the correlation between orientational ordering of C_{60} and intermolecular magnetic interactions. Detailed analysis is in progress.

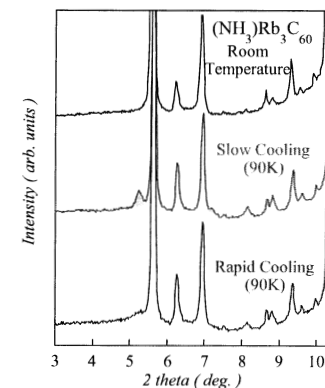


Figure. Diffraction patterns of $(\text{NH}_3)\text{Rb}_3\text{C}_{60}$ at each temperature and cooling protocol.