Structural origin of the anisotropic thermal expansion of Ruddlesden-Popper type oxides

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Ruddlesden-Popper type oxides consist of the alternate stacking of the rock-salt AO- and the perovskite ABO3-layers. Here A and B are larger and smaller cations, respectively. The Ruddlesden-Popper type oxides exhibit a variety of interesting electrical and magnetic properties. Due to these properties, Ruddlesden-Popper type materials have attracted considerable attention as electrode materials in solid oxide fuel cells (SOFCs), and oxygen separation membranes. Because these materials are usually used at the high-temperature, the thermal expansion behavior is one of the most important properties of these materials. Especially, understanding the anisotropy of the thermal expansion is very important.

In order to understand the structure origin of the thermal expansion behavior, it is necessary to determine the precise crystal structures including the position of the oxygen atoms. Therefore, crystal structure analysis based on the X-ray diffraction data is not enough and crystal structure analysis based on the neutron diffraction data is a key for this purpose. In the present study, high-temperature and high-resolution powder neutron diffraction measurements were carried out for the several Ruddlesden-Popper type oxides, which show anisotropic thermal expansion, in order to reveal the structure origin of their anisotropic thermal expansion behaviors.

High resolution neutron powder diffraction measurements have been carried out for $R_2SrAl_2O_7$ (R : rare-earths) at Echidna, ANSTO. Each sample was introduced into a vanadium can and the measurement was carried out in the transmission mode. The measurements were carried out from room temperature to high temperature (maximum 1200 °C) at 200 °C intervals. Each measurement time was few hours. The structural analyses for these data are carried out by Rietveld method using the program RIETAN-FP. The result of the Rietveld refinement of $La_2SrAl_2O_7$ at the room temperature 23 °C is shown in Figure (R_{wp} = 7.395 %, R_B = 1.592 %).

In this structure, La and Sr atoms show positional disorder and they share two different cation sites. The distribution of cations is important to understand the anisotropy of the thermal expansion. The occupancy factors are determined by the structure refinement of the neutron diffraction data. To have more reliable information, now the synchrotron X-ray diffraction analysis and theoretical calculations are also carried out for these materials and we carefully determine the occupancy factors of the cations. The structure analysis of the high-temperature data are also ongoing.

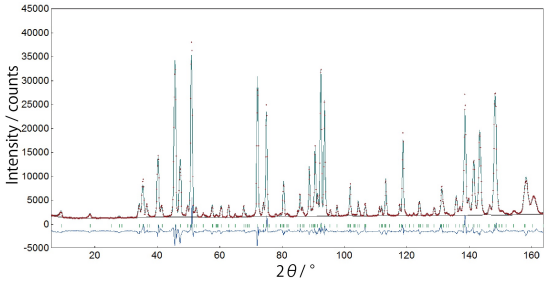

Fig. 1. Rietveld plot of the neutron powder diffraction data of $La_2SrAl_2O_7$