


(※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。)

 <b>MLF Experimental Report</b>	提出日 Date of Report February 1, 2013
課題番号 Project No. 2013B0178  実験課題名 Crystal Structure Analysis of Novel $AA'BO_4$ -Based Mixed and Ionic Conductors 実験責任者名 Name of principal investigator Masatomo Yashima 所属 Affiliation Department of Chemistry and Materials Science, Graduate School of Science and Engineering, Tokyo Institute of Technology	装置責任者 Name of responsible person Toru Ishigaki 装置名 Name of Instrument/(BL No.) iMATERIA / BL-20 実施日 Date of Experiment March 31-April 2, 2014

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)  
 Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.  $AA'BO_4$ - and $A_3B_{11}O_{20}$ -based materials (A: large cations such as rare earth La, Pr, Nd, Y, Ho, Yb and Ba, Sr, Ca; B: smaller cations as In, Ge, Ga, Ta, W).
---

2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。) Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.  <b>Experimental methods</b> Time-of-flight (TOF) neutron powder diffraction data of $AA'BO_4$ - and $A_3B_{11}O_{20}$ -based materials were measured at room temperature by a high-resolution neutron powder diffractometer iMATERIA installed at the beam line BL20 of J-PARC facility, Japan. The sintered or the powder samples were put into 6 mm $\phi$ vanadium sample holders and were used for the diffraction measurements. The diffraction measurements were carried out with double frame mode.  <b>Experimental results</b> TOF neutron diffraction pattern of an $ABaInO_4$ is shown in Figure 1. Recently we reported $NdBaInO_4$ as an oxide-ion conducting material belonging to a new structure family ( <i>Chem. Mat.</i> , 26(8), 2488-2491 (2014)). The structure refinement of the $ABaInO_4$ is now carrying out based on the obtained TOF neutron data using the program Z-Code and it indicates that the $ABaInO_4$ belongs to the same structural type as that of $NdBaInO_4$ . The structure analysis of the $ABaInO_4$ will continue in order to determine its precise crystal structure.
--

## 2. 実験方法及び結果(つづき) Experimental method and results (continued)

TOF-neutron data of doped NdBaInO<sub>4</sub> were also analyzed using the program Z-Code. As shown in Figure 2, it gave good quality of fitting and  $R$ -factors;  $R_{wp} = 4.52\%$  and  $R_B = 6.53\%$ . In the mother material NdBaInO<sub>4</sub>, all cations (Nd, Ba and In) are ordered and there is one independent site for each cation. In doped NdBaInO<sub>4</sub>, the dopant was found to exist not only at the Ba site but also at Nd site from the refined occupancy factors of the cations. We already found that NdBaInO<sub>4</sub> and doped NdBaInO<sub>4</sub> are oxide-ion conducting materials and doped NdBaInO<sub>4</sub> has higher oxide-ion conductivity and lower activation energy for oxide-ion conduction than NdBaInO<sub>4</sub>. This would be due to the presence of larger amount of carrier (oxygen vacancy) and larger bottleneck size for doped NdBaInO<sub>4</sub>. The doping enables to make larger bottleneck size because the the dopant size is larger than host cation. Further detailed structure analysis will be carried out and we will report the mechanistic aspects of oxide-ion conducting behavior of these series of materials in the paper.

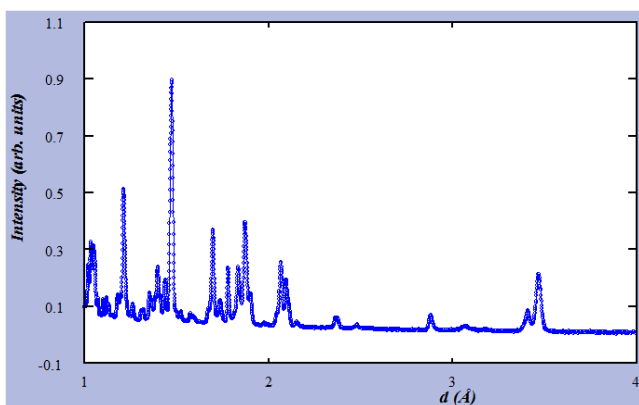


Figure 1 : TOF neutron powder diffraction pattern of ABAInO<sub>4</sub>.

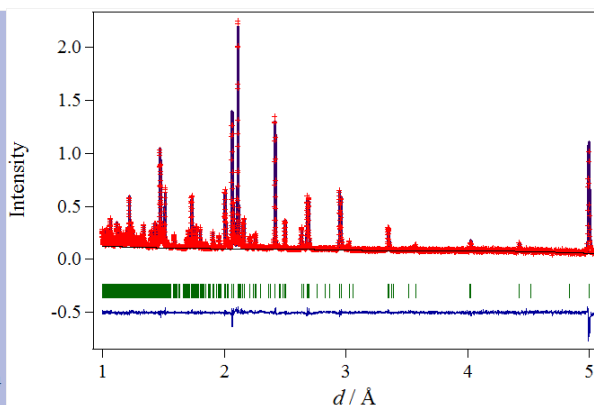


Figure 2 : Rietveld plot for doped NdBaInO<sub>4</sub>.