Oxide and nitride ion distribution effect in crystal structure of new oxynitride superconductor

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Oxynitrides are new materials having interesting chemical and physical properties, because they have characteristic between oxides and nitrides. Unexpected electronic/optical properties are appearing due to a variation in cation-anion covalency in coexistence of oxide and nitride ions. Recently, our research group have reported that Nb-Al oxynitride having the rock salt type crystal lattice showed superconductivity with Tc = 15 K [1]. After annealing at 1100 oC in evacuated sealed tube, its rock salt crystal improved the crystallinity and its superconducting volume fraction increased above 30%. In this study, we investigated the crystal structure and oxide/nitride ionic distribution in the Nb-Al oxynitrides before and after thermal annealing.

Nb-Al oxynitride was prepared by a gel nitridation method [1]. As nitrided powder was post annealed at 1500 oC for 3 h in 0.5 MPa of nitrogen atmosphere. Neutron diffraction measurements at room temperature were carried out with the diffractometer HERMES installed at the JRR-3M reactor in Japan Atomic Energy Agency, Tokai, Japan. Program RIETAN-2000 [2] was used for the structure refinement.

The observed, calculated and difference neutron diffraction profiles for the post annealed Nb-Al oxynitride having the starting composition of Nb:Al = 0.75:0.25 are shown in Fig. 1. Small amount of impurities was observed in the diffraction profile. The structure refinement and oxygen/nitrogen analysis showed the composition of the rock salt type Nb-Al oxynitride in the post- annealed products was refined to be (Nb0.89(1)Al0.11(1))(O0.16(1)N0.84(1)) independent of their starting compositions. Cation sites were randomly occupied by both Nb and Al. Both oxide and nitride ions were also randomly distributed on anion sites in the oxynitride. Recently we have obtained single phase of Nb-Al oxynitride at Nb:Al = 0.89:0.11 and its showed Tc = 17 K and 91% of superconducting volume fraction after its thermal annealing.

References

[1] S. Yamamoto, et al., J. Alloys Compd., 482 (2009) 160-163.
[2] F. Izumi and T. Ikeda, Mater. Sci., Forum, 198 (2000) 321-324.

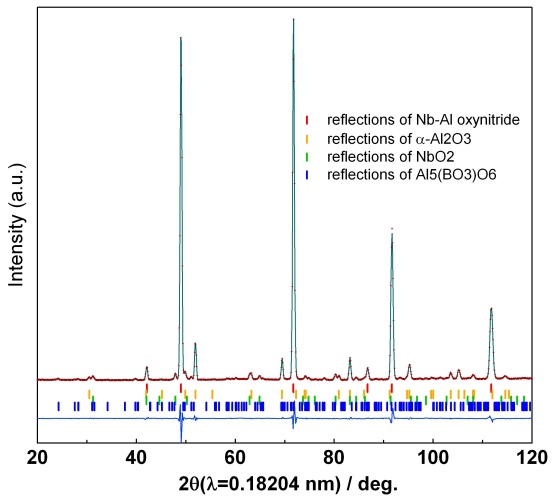

Fig. 1. Neutron diffraction profiles for the post annealed Nb-Al oxynitride at Nb:Al = 0.75:0.25.