

Processing and Thermoelectric Properties of Highly Textured Na-Co-O Ceramics Fabricated by the Reactive Template Grain Growth (RTGG) Method

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反応性テンプレート粒成長 (RTGG) 法による高配向Na-Co-Oセラミックスの合成と熱電特性

金属材料研究室 田島伸

The NaCo_2O_4 system has been a p-type candidate material for thermoelectric applications¹⁻³⁾. The single crystal shows large in-plane thermoelectric power due to the layered structure¹⁾, and thus attains a dimensionless figure of merit ($ZT = S^2\sigma T/\kappa$) greater than 1³⁾, where S is the Seebeck coefficient, σ is the electrical conductivity, κ is the thermal conductivity, and T is the absolute temperature. NaCo_2O_4 polycrystalline ceramics with a preferred c-axis orientation would have in-plane thermoelectric properties as good as the single crystal.

Tani developed a RTGG method for highly-textured polycrystalline ceramics using reactive template particles with an anisotropic shape and lattice matching with a product material⁴⁾. Co_3O_4 has a similar crystal structure to NaCo_2O_4 and thus it is a candidate material as a reactive template. Therefore, c-axis oriented NaCo_2O_4 ceramics could be synthesized by the RTGG method if plate-like Co_3O_4 particles were obtained⁵⁾.

Plate-like Co_3O_4 particles were prepared by a precipitation method. The Co_3O_4 template particles were mixed with Na_2CO_3 powder, a binder and a plasticizer. The slurry was tape-cast by a doctor-blade technique, and then the dried monolayer sheet was cut and laminated to produce a multilayer sheet. The multilayer sheets were heat-treated and then sintered.

Figure 1 shows the XRD profiles of NaCo_2O_4 ceramics prepared by the RTGG method and the JCPDS card. The in-situ reaction was completed during the sintering, and the sintered specimen was identified as NaCo_2O_4 as designed. The parallel surface of the RTGG-processed specimen exhibited strong diffraction peaks from the {001} planes. This result indicates that oriented NaCo_2O_4 grains were formed through a topotaxy or topotaxy-like reaction on the oriented Co_3O_4 particles, and highly textured NaCo_2O_4

ceramics were eventually fabricated by the RTGG method. **Figure 2** shows a SEM photograph of the oriented NaCo_2O_4 ceramics for a section perpendicular to the preferred {001}-oriented plane. The textured ceramics exhibited a unique microstructure in which plate-like grains with a $1\mu\text{m}$ thickness and $10\mu\text{m}$ width were aligned parallel to the casting plane. The σ was larger than that of conventionally sintered ceramics²⁾ due to the texture (600S/cm at 300K). However, the σ values were smaller than expected from the high conductivity of a single crystal (5000S/cm at 300K¹⁾). The power factor ($S^2\sigma$, $4\text{--}5 \times 10^{-4} \text{ W/mK}^2$ above 600K) was not as great as the value reported for conventionally sintered ceramics^{2, 3)}, because the S was not high enough (60-120 $\mu\text{V/K}$). The thermoelectric properties of the c-axis oriented NaCo_2O_4 ceramics could be improved with a more precisely controlled composition.

References

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(Report received on Apr. 23, 2002)

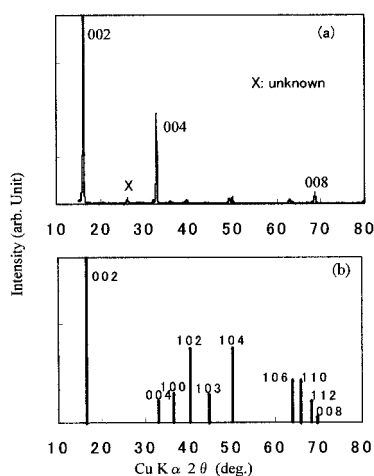


Fig. 1 XRD patterns for (a) NaCo_2O_4 ceramics prepared by RTGG method and (b) JCPDS card of NaCo_2O_4 .

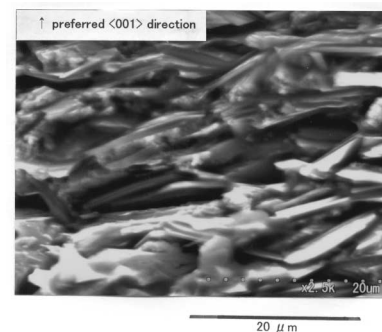


Fig. 2 SEM photograph of NaCo_2O_4 ceramics prepared by RTGG method for perpendicular fracture surface to the original sheet plane.