

## Electronic Supplementary Information

### Extraordinary High Ba<sup>2+</sup> Ion Conducting Solid

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## Additional Experimental Results

### 1. Chemical analyses

The amounts of Ba, N, and O in the 0.75(Gd<sub>0.4</sub>Nd<sub>0.6</sub>)<sub>2</sub>O<sub>3</sub>-0.25Ba(NO<sub>3</sub>)<sub>2</sub> solid were analyzed by using the X-ray fluorescence spectrometer (for Ba) and the nitrogen / oxygen analyzer (for N and O), and the results are listed in Table S1.

In the case for Ba, its amount is constant for all samples. For the nitrogen and oxygen, although both N and O amounts in the sample at 550°C decreased a little compared with those in the mixture, considerable amount (ca. 80%) of N and O were remained in the sample. In contrast, for the sample calcined at 800°C, most of nitrogen disappeared from the sample, which supports the idea that the Ba(NO<sub>3</sub>)<sub>2</sub> decomposed to BaO.

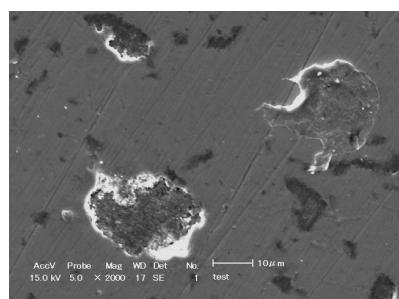
Table S1. The amounts (mass%) of Ba, N, and O in the 0.75(Gd<sub>0.4</sub>Nd<sub>0.6</sub>)<sub>2</sub>O<sub>3</sub>-0.25Ba(NO<sub>3</sub>)<sub>2</sub> solid.

element	Mixture	Calcined at 550°C	Calcined at 800°C
Ba	15.0	14.9	14.7
N	2.15	1.77	0.40
O	18.40	14.24	12.20

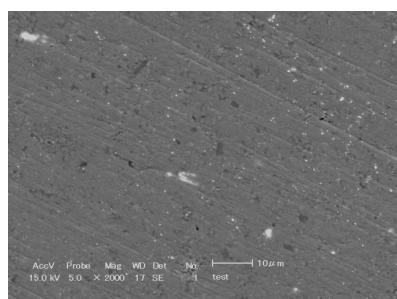
### 2. SEM observations

The SEM photographs of the mixture of (Gd<sub>0.4</sub>Nd<sub>0.6</sub>)<sub>2</sub>O<sub>3</sub> and Ba(NO<sub>3</sub>)<sub>2</sub> (molar ratio: 0.75 : 0.25) and the 0.75(Gd<sub>0.4</sub>Nd<sub>0.6</sub>)<sub>2</sub>O<sub>3</sub>-0.25Ba(NO<sub>3</sub>)<sub>2</sub> solid calcined at 550°C, and also at 800°C are displayed in Figure S1.

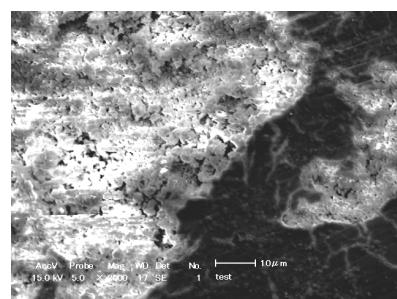
For the mixture, the Ba(NO<sub>3</sub>)<sub>2</sub> particles were clearly observed in the sample. However, such particles were not observed at all for the sample calcined at 550°C. On the other hand, large pores were recognized in the sample, which results from the decomposition of Ba(NO<sub>3</sub>)<sub>2</sub>.



(a)



(b)



(c)

Figure S1. SEM Photographs of the mixture of (Gd<sub>0.4</sub>Nd<sub>0.6</sub>)<sub>2</sub>O<sub>3</sub> and Ba(NO<sub>3</sub>)<sub>2</sub> (molar ratio: 0.75 : 0.25) (a) and the 0.75(Gd<sub>0.4</sub>Nd<sub>0.6</sub>)<sub>2</sub>O<sub>3</sub>-0.25Ba(NO<sub>3</sub>)<sub>2</sub> solid calcined at 550°C (b) or 800°C (c).

### 3. Electrical conductivity measurements

In order to make it clear that the high ion conductivity of the  $0.75(\text{Gd}_{0.4}\text{Nd}_{0.6})_2\text{O}_3\text{-}0.25\text{Ba}(\text{NO}_3)_2$  solid is caused by the formation of solid solution of  $(\text{Gd}_{0.4}\text{Nd}_{0.6})_2\text{O}_3$  and  $\text{Ba}(\text{NO}_3)_2$ , we measured the electrical conductivity of the sample in which  $\text{Ba}(\text{NO}_3)_2$  intentionally decomposed to  $\text{BaO}$  by the calcination at  $800^\circ\text{C}$ . The temperature dependence of the electrical conductivity for the sample which was obtained by calcining the mixture of  $(\text{Gd}_{0.4}\text{Nd}_{0.6})_2\text{O}_3$  and  $\text{Ba}(\text{NO}_3)_2$  at  $800^\circ\text{C}$  is shown in Figure S2 with the corresponding data for the  $0.75(\text{Gd}_{0.4}\text{Nd}_{0.6})_2\text{O}_3\text{-}0.25\text{Ba}(\text{NO}_3)_2$  solid at  $550^\circ\text{C}$ .

As we expected, the conductivity of the sample calcined at  $800^\circ\text{C}$  was considerably lower than that of the  $0.75(\text{Gd}_{0.4}\text{Nd}_{0.6})_2\text{O}_3\text{-}0.25\text{Ba}(\text{NO}_3)_2$  solid, and the value was still lower (1 – 2 orders of magnitude) than that of pure  $(\text{Gd}_{0.4}\text{Nd}_{0.6})_2\text{O}_3$  solid solution (The conducting species were not identified.). This result indicates the fact that the high conductivity can be obtained only for the sample containing  $\text{Ba}(\text{NO}_3)_2$ .

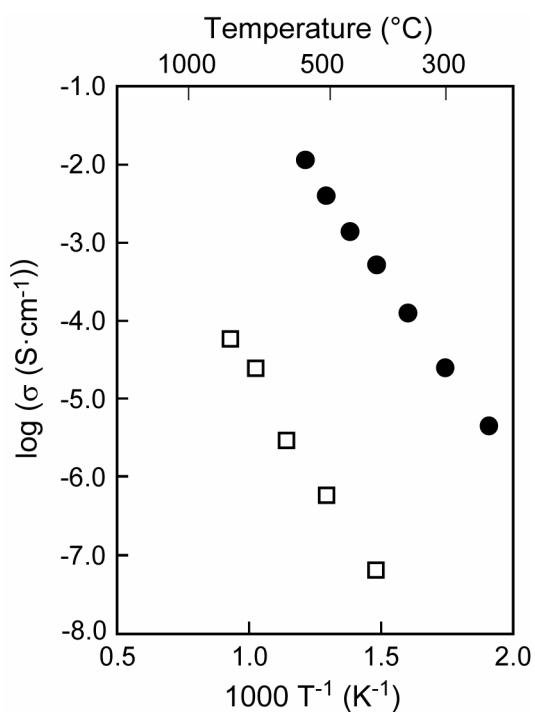


Figure S2. Temperature dependencies of the electrical conductivity for the  $0.75(\text{Gd}_{0.4}\text{Nd}_{0.6})_2\text{O}_3\text{-}0.25\text{Ba}(\text{NO}_3)_2$  solid calcined at  $550^\circ\text{C}$  (●) and the  $(\text{Gd}_{0.4}\text{Nd}_{0.6})_2\text{O}_3$  and  $\text{Ba}(\text{NO}_3)_2$  mixture calcined at  $800^\circ\text{C}$  (□).