Supporting information

Enhanced Oxide-Ion Conductivity of Solid-State Electrolyte Mesocrystal

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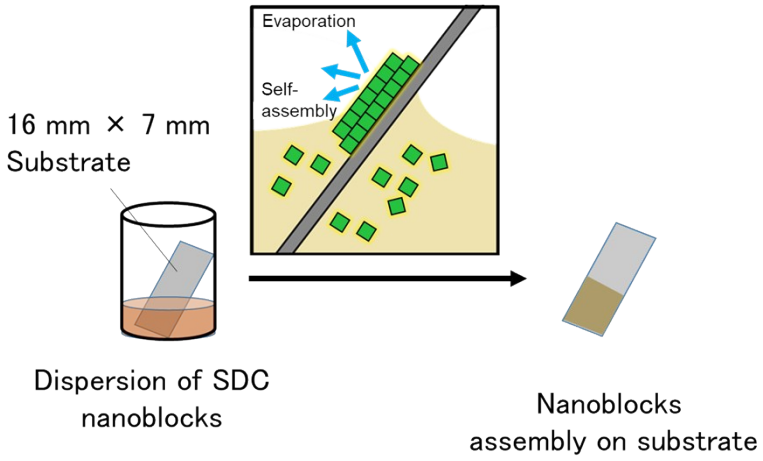

Fig. S1 A schematic illustration of the fabrication system for thin films consisting of nanoblocks thorough evaporation-driven self-assembly using dispersion

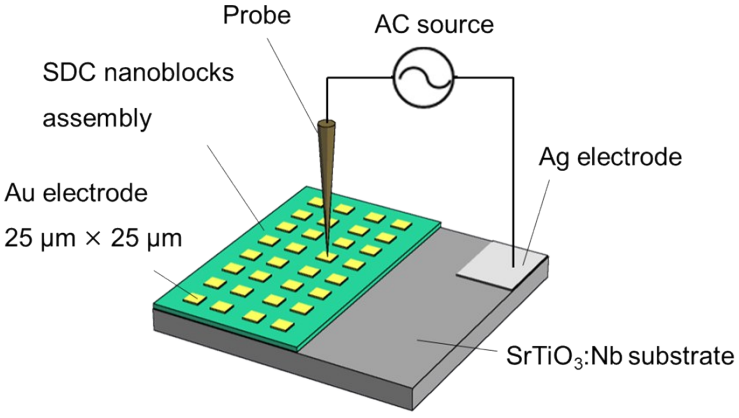

Fig. S2 A schematic illustration of the measurement system for alternating current impedance of thin films. Film thickness of SDC10, SDC20, and SDC30 was 175, 231, and 86 nm respectively.

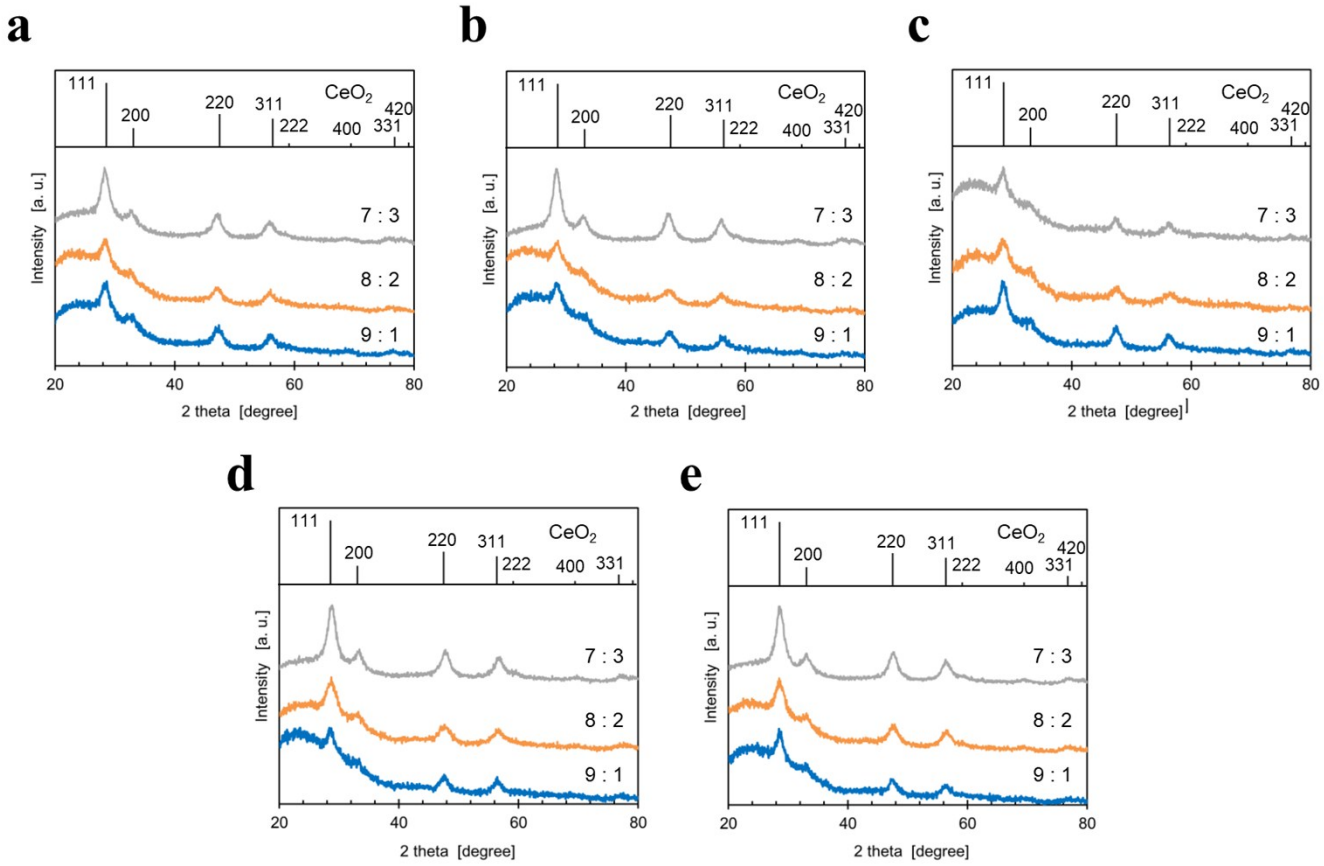

Fig. S3 XRD profiles of REDC nanoblocks. Dopant cations were (a) La^{3+}, (b) Nd^{3+}, (c) Gd^{3+}, (d) Y^{3+}, and (e) Yb^{3+}. The numbers represent the initial molar ratio of the dopant cations.

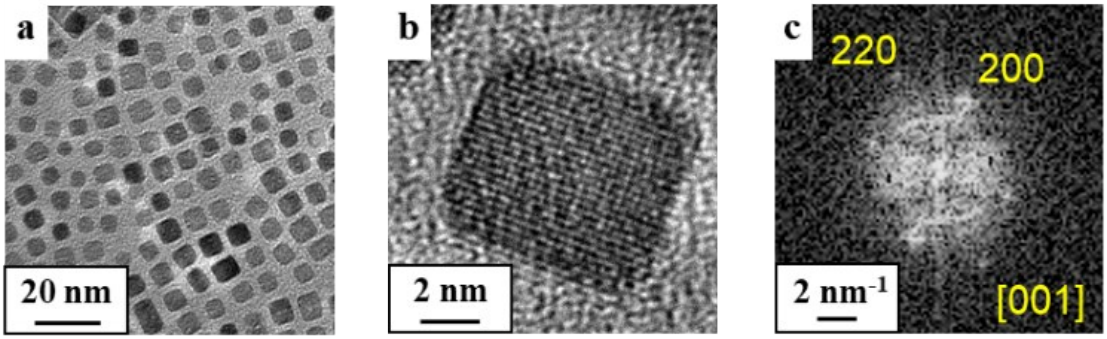

Fig. S4 HRTEM images of CeO_2 nanocubes (a, b) and its FFT image (c) synthesized by the two phase solvothermal method.

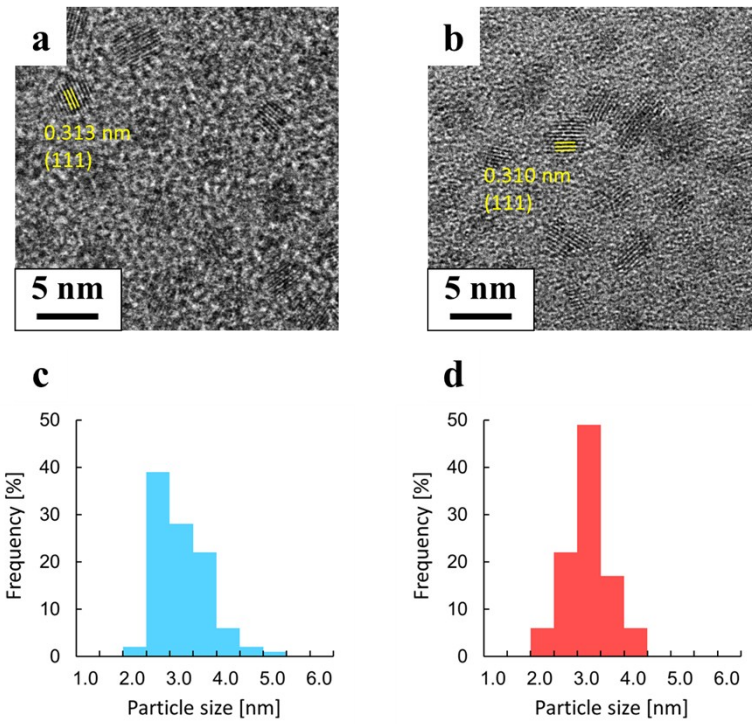

Fig. S5 TEM images (a, b) and size distribution (c, d) of SDC nanoparticles in initial stage of reaction. (a, c) SDC10 and (b, d) SDC30.

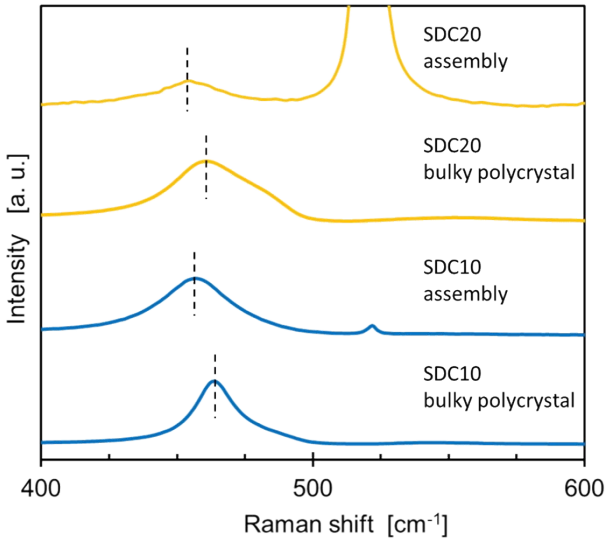

Fig. S6 Raman scattering spectra of SDC nanoblocks assembly. The 465 cm^{-1} peaks of the F_{2g} mode from the nanoblocks assembly were located at a wavenumber lower than those from bulky samples. The 520cm^{-1} peaks were attributed to the vibration of an Si substrate. Bulky polycrystals were obtained by calcination of pellets of the nanoblocks at 1400°C.

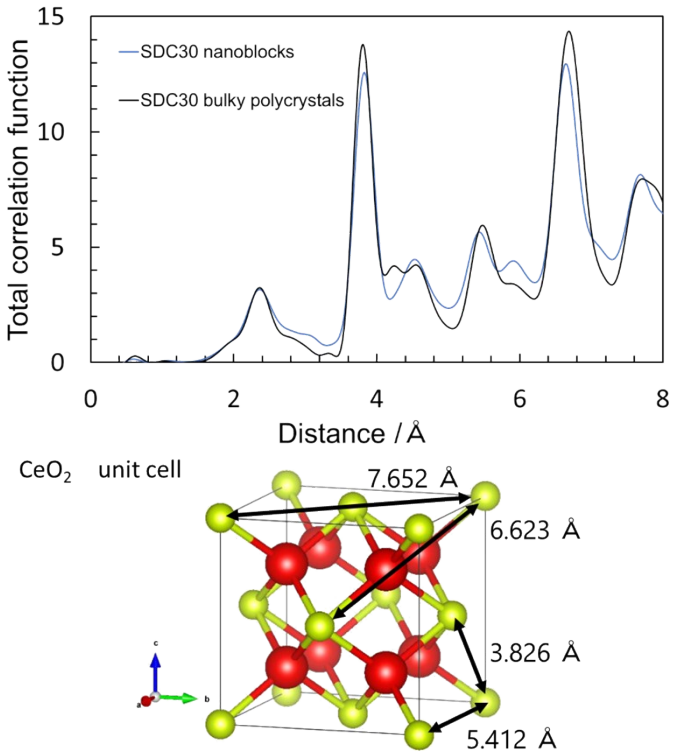

Fig. S7 Total correlation function of SDC30 nanoblocks and a bulky sample obtained from a high-energy X-ray diffraction experiment. The nearest Ce–Ce distance in SDC nanoblocks was larger than that of a bulky sample. We performed a high-energy X-ray diffraction experiment with an incident beam of 61.3 keV by BL04B2 at SPring-8, Japan.[1] The incident X-ray intensity was monitored by an ionization chamber filled with Ar gas, and the scattered beam was measured by Ge and CdTe detectors. The obtained data was corrected using a standard program.[1]

1) S. Kohara, M. Itou, K. Suzuya, Y. Inamura, Y. Sakurai, Y. Ohishi and M. Takata, *J. Phys. Condens. Matter*, 2007, **19**, 506101.

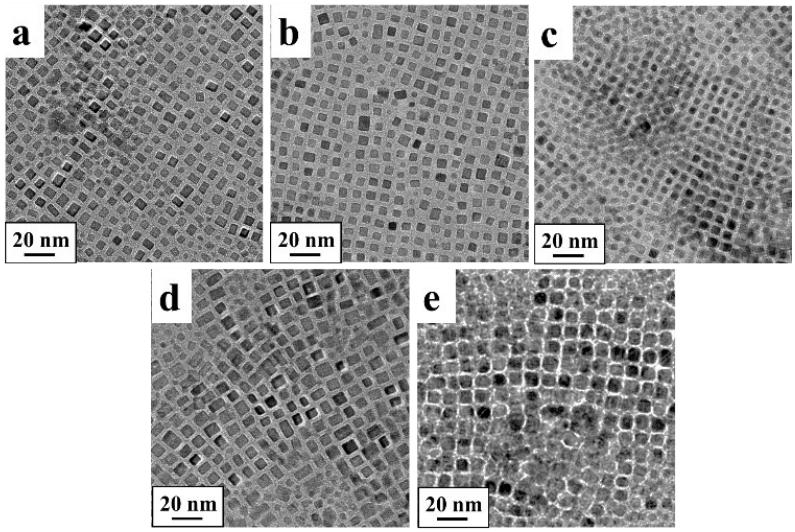

Fig. S8 TEM images of REDC nanocubes prepared in the mother solution at $[Ce^{3+}] : [RE^{3+}] = 9 : 1$. Dopant cations were (a) La^{3+}, (b) Nd^{3+}, (c) Gd^{3+}, (d) Y^{3+}, and (e) Yb^{3+}.

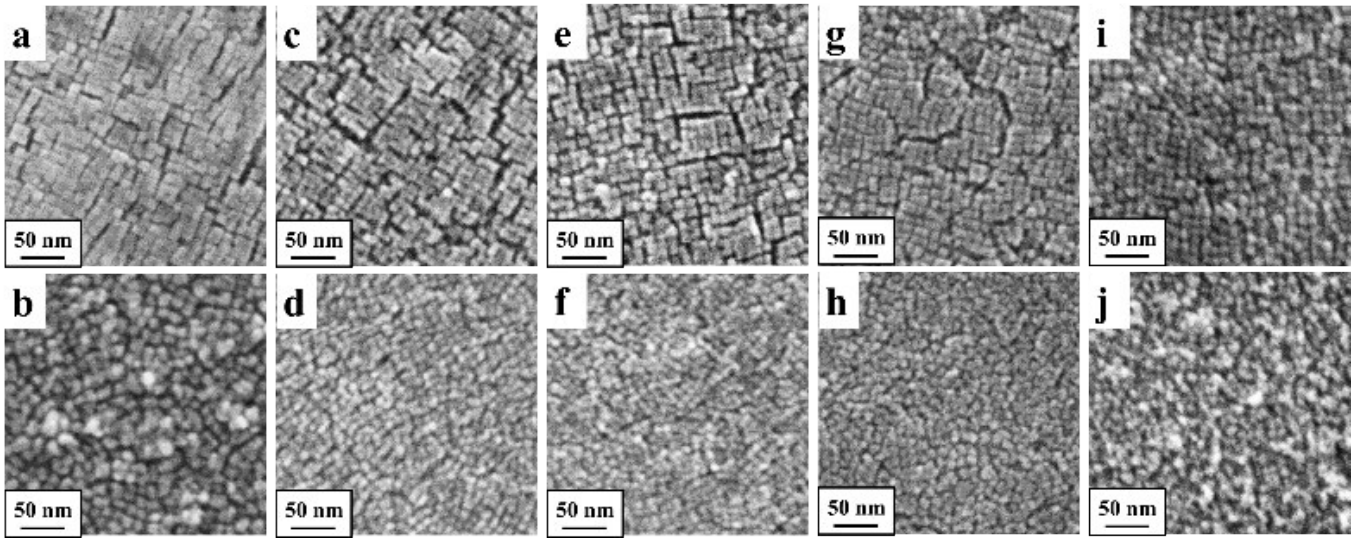

Fig. S9 SEM images of thin films consisting of REDC nanoblocks prepared in the mother solution at [Ce^{3+}] : [RE^{3+}] = 9 : 1 (upper row) and 8 : 2 (lower row). Dopant cations were (a, b) La^{3+}, (c, d) Nd^{3+}, (e, f) Gd^{3+}, (g, h) Y^{3+}, and (i, j) Yb^{3+}.

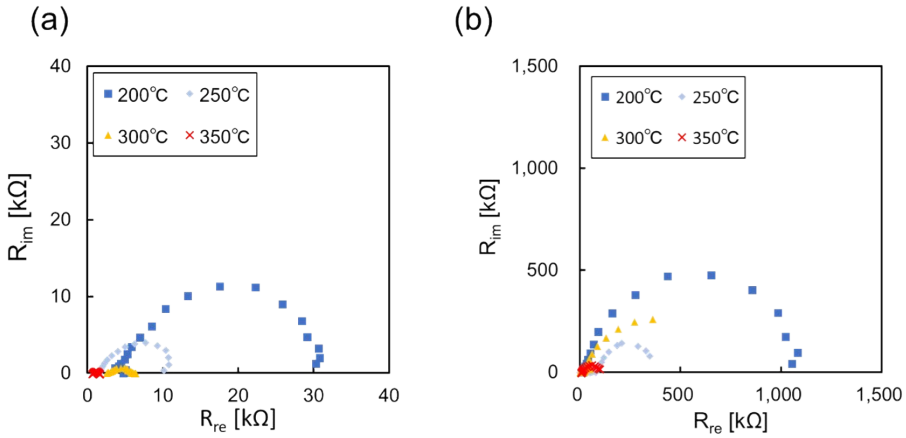

Fig. S10 Nyquist plots (the relationship between real part R_{re} and imaginary part R_{im} of the frequency response) for films consisting of SDC10 (a) and SDC20 (b).

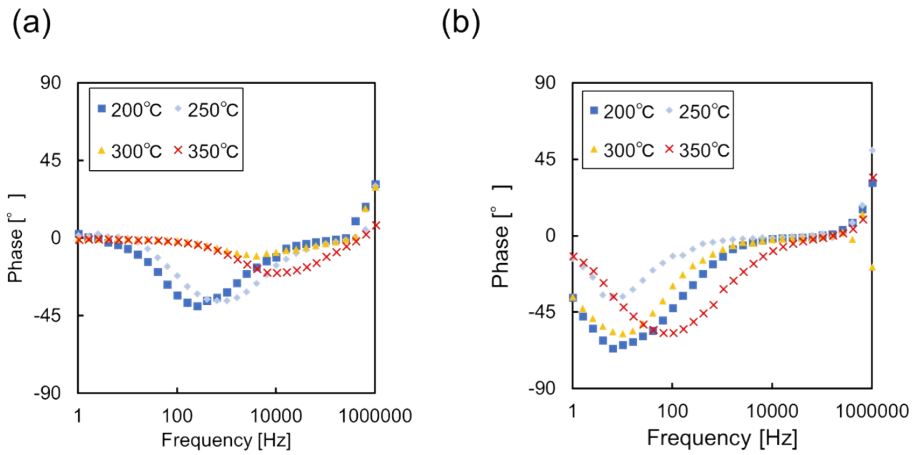

Fig. S11 Bode plots (the phase shift as a function of frequency) for films consisting of SDC10 (a) and SDC20 (b).

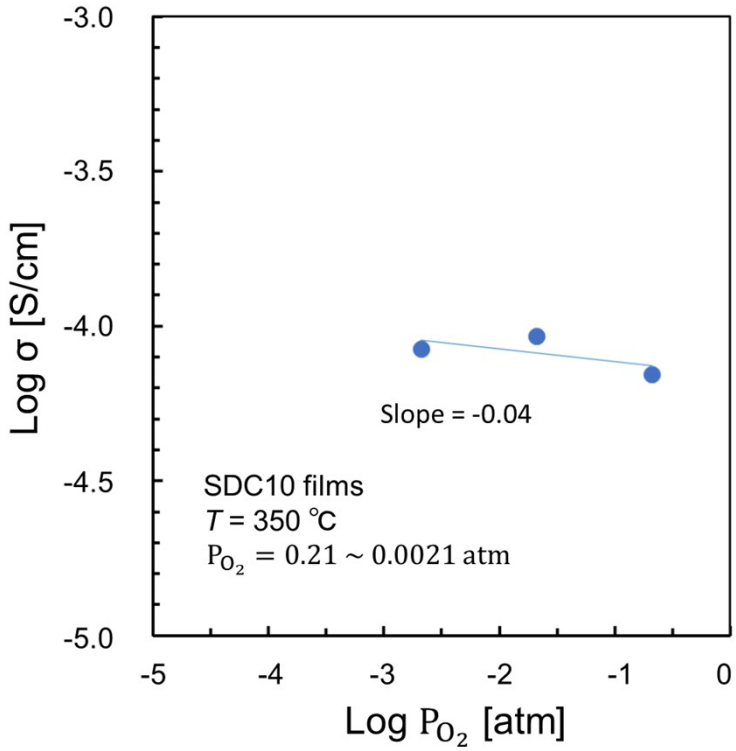

Fig. S12 Dependence of electrical conductivity of SDC10 film on oxygen partial pressure.

The electrical conductivity σ was determined from the obtained resistance R, film thickness l, and electrode area A using the following equation.

$$\sigma = \frac{l}{AR} \qquad (1)$$

From the relationship between temperature and ionic conductivity, activation energy E_a and pre-exponential factor σ_0 were calculated using the following Arrhenius equation.

$$\sigma T = \sigma_0 exp(-\frac{Ea}{k_B T}) \qquad (2)$$