The synthesis of high-entropy perovskite oxide epitaxial thin films using a pulsed laser deposition technique

Tokyo Tech [1], JST-PRESTO [2]
○ (D) Kaidong Wang[1], Kazunori Nishio[1], Kurei Edamura[1], Yuki Sasahara[1], Ryo Nakayama[1], Ryota Shimizu[1,2] and Taro Hitosugi[1]
E-mail: wang.k.ae@m.titech.ac.jp

[Introduction]: ABO_3 perovskite oxides exhibit excellent and diverse physical properties for applications in a variety of areas, such as solid oxide fuel cells, proton conductors, photocatalysts, dielectrics, ferroelectrics, and multiferroics. Recently, ABO_3 perovskite has been extended to high-entropy compositions to explore an unexpected functionality [1]. In general, a single structural phase of high-entropy perovskite oxides (HEPOs) is formed by enhancing the configurational entropy of a multiple component solid solution through mixing a large number of cations. However, the synthesis of HEPOs is at a very early stage, and thus, the stabilization of the perovskite structure has yet to be well studied [2,3]. Accordingly, we focused on the synthesis of HEPO epitaxial thin films, containing a variety of cations in A and B sites with more than 10 elements. As a result, we succeeded in the synthesis of a single phase of HEPO epitaxial thin films, which contains Ca, Sr, and Ba in A site and Si, Ti, Cr, Mn, Fe, Co, Ni, Ge, Zr, Sn, Ce, and Hf in B site in equiatomic ratios, respectively (Figure 1(a)).

[Experiment]: HEPO thin films were deposited on $SrTiO_3(001)$ single crystal substrates using pulsed laser deposition technique. A KrF excimer laser (wavelength: 248 nm, pulse duration: ~20 ns, repetition frequency: 5 Hz, spot size: 0.015 cm^2 and fluence: 1.33 J/cm^2) was used to ablate a polycrystalline ABO_x target (A = Sr, Ca, Ba, B = Si, Ti, Cr, Mn, Fe, Co, Ni, Ge, Zr, Sn, Ce, Hf). The substrate temperature (T_s) during the thin film growth was varied in the range of 600–800°C. An oxygen pressure was set at 100 mTorr for the thin film growths. X-ray diffraction (XRD) measurement was performed to investigate the crystal structure and orientation for synthesized thin films.

[Results]: Figure 1(b) shows the out-of-plane XRD patterns of the thin films grown at varied T_s. No diffraction peaks from a thin film were confirmed when grown at T_s = 600°C. In contrast, the thin films exhibited 002 and 004 diffractions when deposited at ≥ 700°C, indicating the successful synthesis of (001)-oriented HEPO epitaxial thin films.

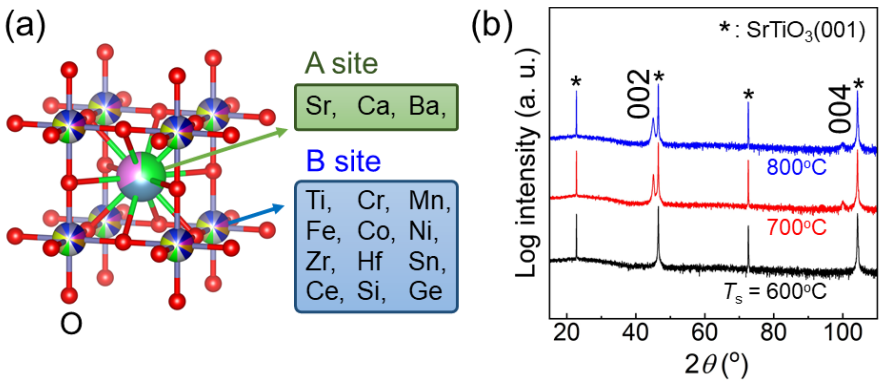

Figure 1. (a) High-entropy ABO_3 perovskite structure and selected elements in A and B sites. (b) Substrate temperature (T_s) dependence of out-of-plane X-ray diffraction patterns for the fabricated thin films.

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[3] R. K. Patel *et al.*, *Appl. Phys. Lett.* **116** (2020) 071601.