Ferromagnetic double perovskites ($A_2BB'O_6$, $A$=alkaline earth metals and $B/B'$=transition metals) have recently attracted great attention due to their presumed half-metallicity as well as high Curie temperature. To this end, Sr$_2$FeReO$_6$ (SFRO), one of typical double perovskites, is being actively studied with this purpose. The magnetic structure of $A_2BB'O_6$ is known to originate from the ordered arrangement of parallel Fe$^{3+}$ ($3d^5$, $S=5/2$) magnetic moments, antiferromagnetically coupled with Re$^{5+}$ ($5d^2$, $S=1$) spins, and therefore the properties of the material depend on their ordering in B-sites. In this study, the cationic ordering was controlled by excessive Re amount and their magnetic properties have been investigated as a function of cationic ordering. Furthermore, what the cationic disordered defects, so called antisite (AS) defects, look like was examined by aberration-corrected STEM.

0, 5, 10, and 15 wt%-Re-excess SFRO powders were prepared via a conventional solid state reaction for 24hr and annealed at 1000°C for 20min in Ar. The prepared powders were sintered by Spark Plasma Sintering (SPS) at 1150°C for 20min in Ar. XRD results confirm that cationic ordering of Fe and Re, which is one of major parameters to affect the magnetic properties in SFRO and the secondary phases are not observed in all samples (Fig. 1(a)). As shown in Fig. 1(b), the height of (011) and (110) peaks appear to be variable by amount of excess-Re and thus cationic ordering percentages are found to be ~ 75% (0%-Re), ~ 80% (5%-Re), ~ 90% (10%-Re) and ~ 95% (15%-Re) by calculating the ratio of (011) and (110) peak height and XRD simulation. Since the $M_s$ of SFRO are deteriorated by the cationic disordering, $M_s$ of 15wt%-Re-excess SFRO sample is the largest value amongst the prepared samples. These results directly demonstrate that excess of Re effectively enhance the cationic ordering in SFRO and accordingly the magnetic properties as well, as shown in Fig. 1 and 2. Lower $M_s$ and higher $H_{c}$ in the 0wt% Re sample reveal that the ferromagnetic alignment of Fe and Re atoms is interfered by the Fe/Re disordering. Aberration-corrected STEM results of Fig. 3 show that high degree of Fe/Re ordering maintains in 15wt%-Re-excess SFRO sample whilst cationic-disordered regions are observed in the 0wt% Re sample. This STEM observation implies that AS defects inferred by cationic disordering exhibit clustered rather than randomly scattered.

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Fig. 1: (a) XRD patterns of SFRO-xRe with 0wt%≤x≤15wt% (b) Zoom in from 19° to 21°

Fig. 2: (a) measured magnetic property via VSM by increasing amount of Re (b) The change of the magnetic saturation (Ms) and AS defect

Fig. 3: High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images from (a) SFRO-0wt% Re sample (b) SFRO-15wt% Re sample.