

## Supporting materials for

### **The magnetic and crystal structures of $\text{Sr}_{1-\delta}\text{FeO}_{2-x}\text{F}_x$ , a new oxyfluoride**

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Including

Figure S1

Figure S2

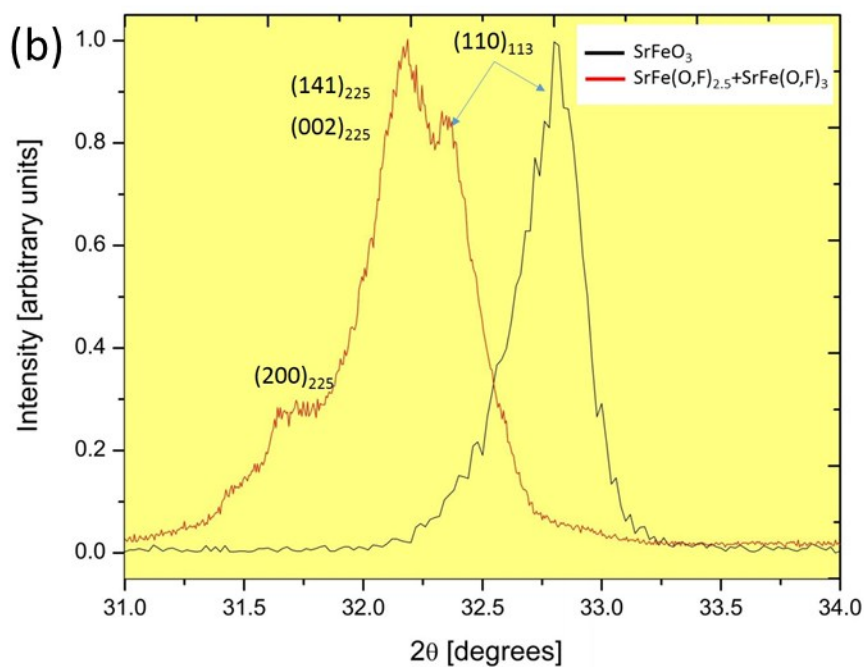
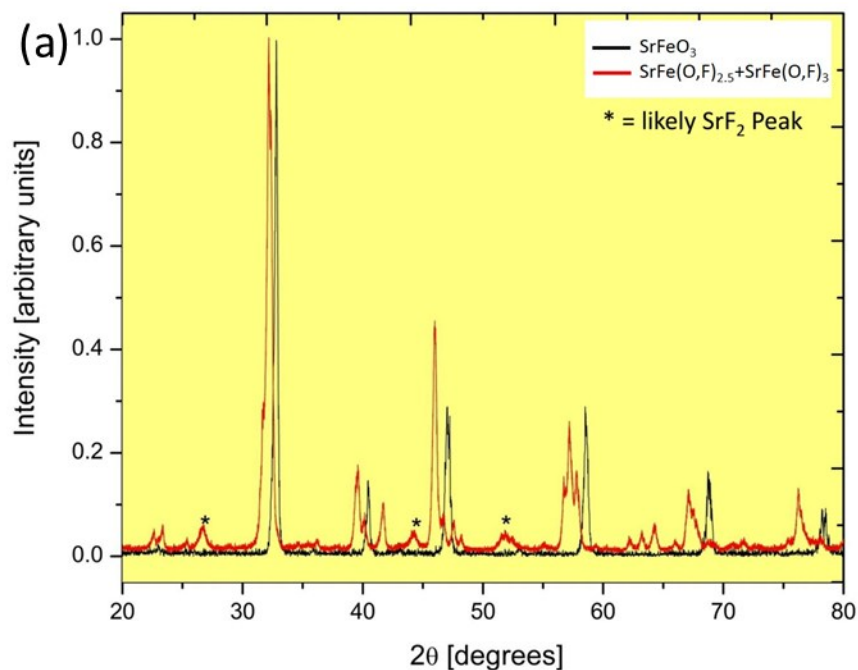
Figure S3

Figure S4

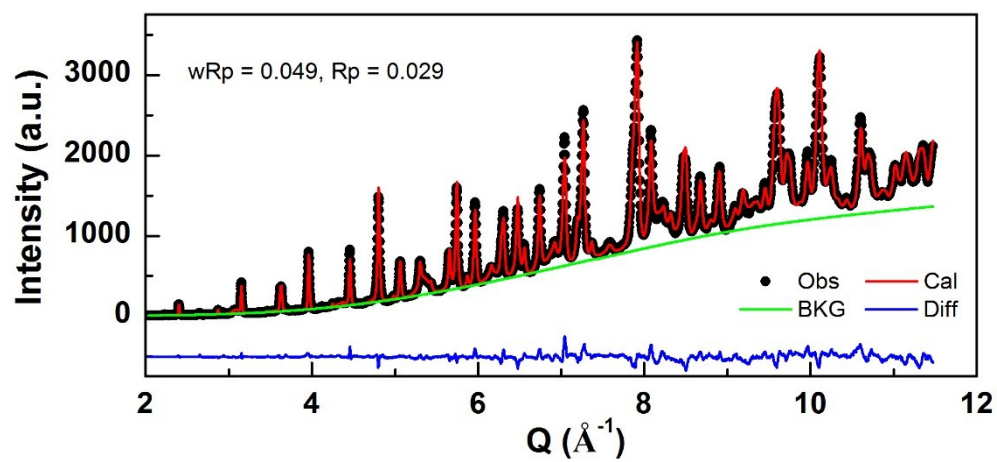
Table S1

Table S2

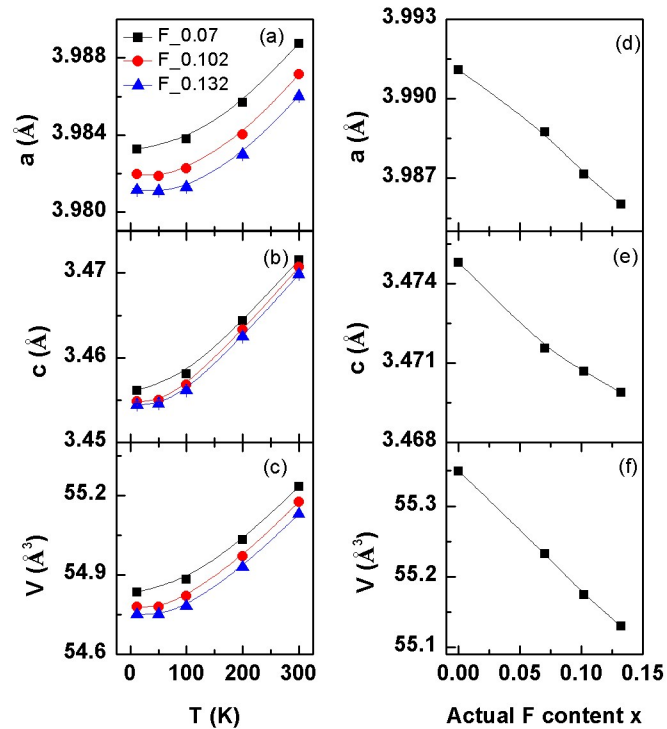
Table S3



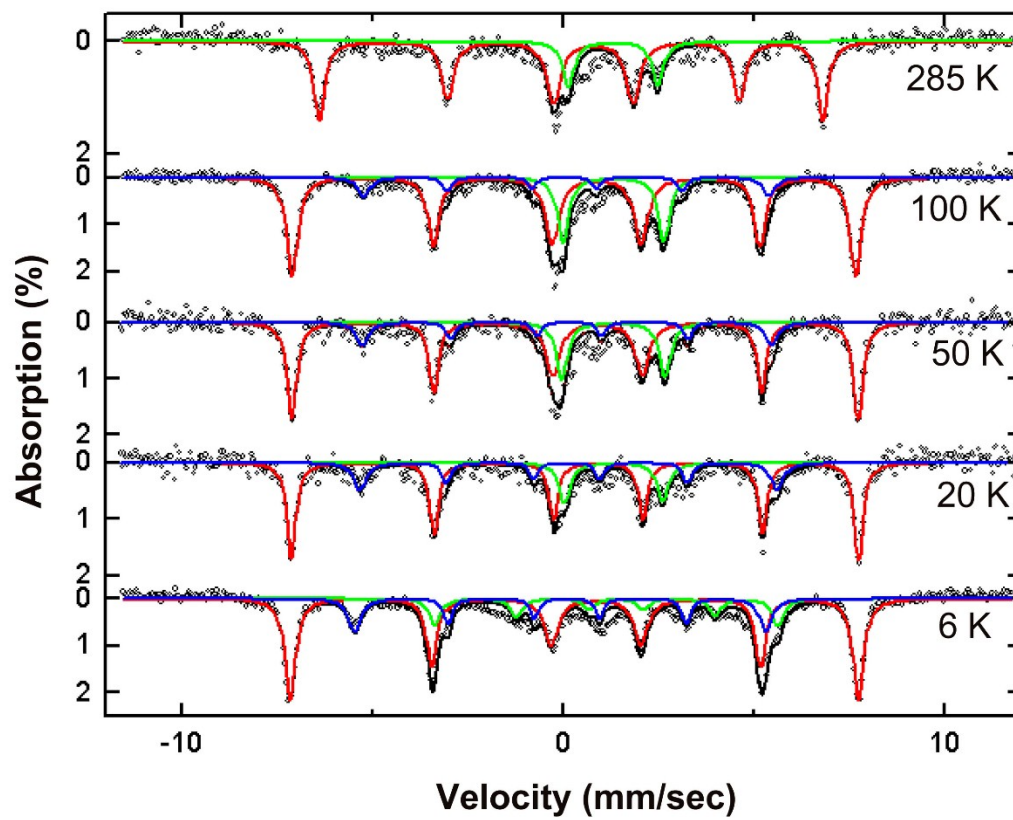
**Figure S1.** The X-ray diffraction of product after reaction of  $\text{SrFeO}_3$  with PVDF for nominal composition  $\text{SrFeO}_{2.5}\text{F}_{0.5}$ . It consists of three phases: cubic  $\text{SrFe}(\text{O},\text{F})_3$ , orthorhombic brownmillerite  $\text{SrFe}(\text{O},\text{F})_{2.5}$  and impurity  $\text{SrF}_2$ . The fluoridated cubic phase has expanded lattice compared with parent one. The subscripts 113 and 225 of the Miller indices represent cubic  $\text{SrFe}(\text{O},\text{F})_3$  and orthorhombic brownmillerite  $\text{SrFe}(\text{O},\text{F})_{2.5}$ .



**Figure S2.** Rietveld refinement of neutron diffraction of nominal  $\text{SrFeO}_{1.6}\text{F}_{0.4}$  at 10 K with  $P4/mmm$  model, in which Sr (0.5, 0.5, 0.5), Fe (0, 0, 0) and O/F (0.5, 0, 0).



**Figure S3.** The temperature and composition dependence of lattice parameters  $a$ ,  $c$ , and volume of unit cell,  $V$ , determined from the Rietveld refinements with  $P4/mmm$  model.



**Figure S4.** The temperature dependence of the Mossbauer spectra.

**Table S1** Local structure model for  $x = 0.132$  at 300 K. Space group: **P2/m**,  $a = 15.9614 \text{ \AA}$ ,  $b = 7.9745 \text{ \AA}$ , and  $c = 3.4720 \text{ \AA}$ ,  $\beta = 89.83^\circ$ .

Atoms	Sites	x	y	z	$U_{11}$ ( $\text{\AA}^2$ )	$U_{22}$ ( $\text{\AA}^2$ )	$U_{33}$ ( $\text{\AA}^2$ )	$U_{12}, U_{13}, U_{23}$ ( $\text{\AA}^2$ )
<b>Sr</b>	Sr1	0.1208	0.2516	0.5092	0.0008	0.0006	0.0082	0
	Sr2	0.6282	0.2520	0.5187	0.0008	0.0006	0.0082	
<b>Fe</b>	Fe1	0	0	0	0.0024	0.0082	0.0048	
	Fe2	0	0.5	0	0.0024	0.0082	0.0048	
	Fe3	0.25	0	0	0.0024	0.0082	0.0048	
	Fe4	0.25	0.5	0	0.0024	0.0082	0.0048	
	Fe5	0.5	0	0	0.0024	0.0082	0.0048	
	Fe6	0.5	0.5	0	0.0024	0.0082	0.0048	
<b>O/F</b>	O1	0.1261	0	0.9450	0.0006	0.0022	0.0122	
	O2	0.6301	0	0.9635	0.0006	0.0022	0.0122	
	O3	0.1218	0.5	0.0120	0.0006	0.0022	0.0122	
	O4	0.6236	0.5	0.0165	0.0006	0.0022	0.0122	
	O5	0	0.2503	0	0.0006	0.0022	0.0122	
	O6	0.5	0.2488	0	0.0006	0.0022	0.0122	
	O7	0.2452	0.2424	0.0200	0.0006	0.0022	0.0122	

**Table S2** The Mossbauer parameters deduced from a least-squares fit of the red spectra corresponding to Fe<sup>2+</sup>. The hf fields agree with the results from Ref. 12 (401 kG at 285 K) and Ref. 23 (463 kG at 4.2 K).

<b>Temperature (K)</b>	<b>B<sub>hf</sub></b>	<b>δ</b>	<b>½e<sup>2</sup>Qq</b>
285	410	0.48	-0.58
100	458	0.57	-0.6
6	457	0.57	-0.57

**Table S3** Typical values of the isomer shifts of different valence states of iron for sixfold and fourfold oxygen coordination are shown. These values are expected to be reduced for smaller coordination numbers  $z$  as shown in the Table. For  $\text{Fe}^{2+}$  in fourfold square-planar coordination (Gillespite) the shift has been measured (Ref. 24 in the text) to be 0.75 mm/s (when corrected for the  $\text{Pd}^{57}\text{Co}$  source) in good agreement with the value in the Table. Thus, a similar reduction for  $\text{Fe}^+$  would give a value  $\delta = 1.25$  mm/s which is consistent with the value  $\delta = 1.24$  mm/s found for the doublet.

<b>Coordination Number (z)</b>	<b><math>\text{Fe}^{3+}</math></b>	<b><math>\text{Fe}^{2+}</math></b>	<b><math>\text{Fe}^{1+}</math></b>
6	0.5	1.10	1.55 (Ref. 25)
4	0.20	0.80	1.25 (Refs. 26, 27)