# A Cluster Expansion Framework for the $Sr(Ti_{1-x}Fe_x)O_{3-x/2}$ ($0 < x < 1$) Mixed Ionic Electronic Conductor: Properties based on Realistic Configurations

## SUPPORTING INFORMATION

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### Section 1. Cluster information and ECIs:

The total number of atomic sites in our supercells is 160. The cluster expansion built here is restricted to stoichiometries given by the reference composition $Sr(Ti_{1-x}Fe_x)O_{3-x/2}$ for variable x. These considerations introduce constraints to the cluster sums present, resulting in a reduction in the independent set of clusters needed. For example, all dimer clusters are subject to the following constraints:

$$n_{TiFe} + n_{TiTi} = N_{Ti}^{C} n_{Ti} \quad (1)$$

$$n_{TiFe} + n_{FeFe} = N_{Fe}^{C} n_{Fe} \quad (2)$$

$$n_{Ti} + n_{Fe} = n_{B-sites} \quad (3)$$

where $N_{Ti}^{C}$ and $N_{Fe}^{C}$ are the number of sites that could form a dimer of a particular length with Ti and Fe sites respectively. With these three constraints applied, there remain only

two degrees of freedom amongst variables $n_{TiTi}$, $n_{TiFe}$, $n_{FeFe}$, $n_{Ti}$, and $n_{Fe}$. That is to say, upon establishing the number of Ti atoms, and number of Ti-Fe dimers, the number of the remainder clusters are set due to the fixed composition.

Upon adoption of such constraints, the final set of clusters used in the cluster expansion are listed below in Table S1. These include 1 monomer, 8 dimers, 13 trimers and 4 tetramers. Within Table S1, the column labeled "cluster shape" refers to the length of all edges for the given cluster.

| Cluster Composition | Cluster Shape | ECI (eV) |
|---|---|---|
| Constant part | N/A | -321.5457 |
| Ti | Single Atom | -27.6898 |
| Ti-Fe | (3.95) | -0.0286 |
| Ti-Fe | (5.58) | -0.0017 |
| Ti-Fe | (6.84) | -0.0175 |
| Ti-Fe | (7.90) | 0.0120 |
| O-Vacancy | (2.79) | -0.7514 |
| O-Vacancy | (3.95) | 0.4188 |
| Fe-Vacancy | (1.97) | -5.3952 |
| Fe-Vacancy | (4.41) | -0.1099 |
| Ti-Fe-O | (1.97,1.97,3.95) | -0.0134 |
| Ti-Fe-O | (1.97,3.95,5.92) | -0.0192 |
| Ti-Fe-O | (1.97,3.95,4.41) | 0.0102 |
| Ti-Ti-Ti | (3.95,3.95,5.58) | 0.0166 |
| Ti-Ti-Ti | (5.58,5.58,7.90) | -0.0021 |
| Ti-Ti-Ti | (3.98,5.58,6.84) | -0.0059 |
| Ti-Ti-Ti | (5.58,5.58,5.58) | -0.0085 |
| O-O-Vac | (2.79,2.79,2.79) | 0.4339 |
| O-O-Vac | (2.79,2.79,3.95) | -0.2770 |
| O-O-Vac | (2.79,2.79,5.58) | 0.0166 |
| Fe-O-O | (1.97,1.97,2.79) | -0.9386 |
| Fe-O-O | (1.97,1.97,3.95) | -1.0370 |
| Fe-O-O | (1.97,4.41,3.95) | -0.0773 |
| Ti-Ti-Fe-Fe | (3.95,5.58,3.95,3.95,5.58,3.95) | -0.0107 |
| Ti-Fe-Fe-Vac | (3.95,5.58,1.97,3.95,1.97,4.41) | 0.0091 |
| Fe-O-Vac-Vac | (1.97,1.97,1.97,2.79,2.79,2.79) | 0.5323 |
| O-O-Vac-Vac | (2.79,2.79,3.95,2.79,2.79,2.79) | -0.2263 |

## Section 2: Determination of equilibrium configurations using probability configuration

To obtain the high temperature T = 1000 K configurations, we first use Monte Carlo to generate a probability distribution of finding configurations of particular energies as shown in Fig. S1. Once the probability distribution has been established, a configuration from the peak of the spectrum is selected for further analysis.

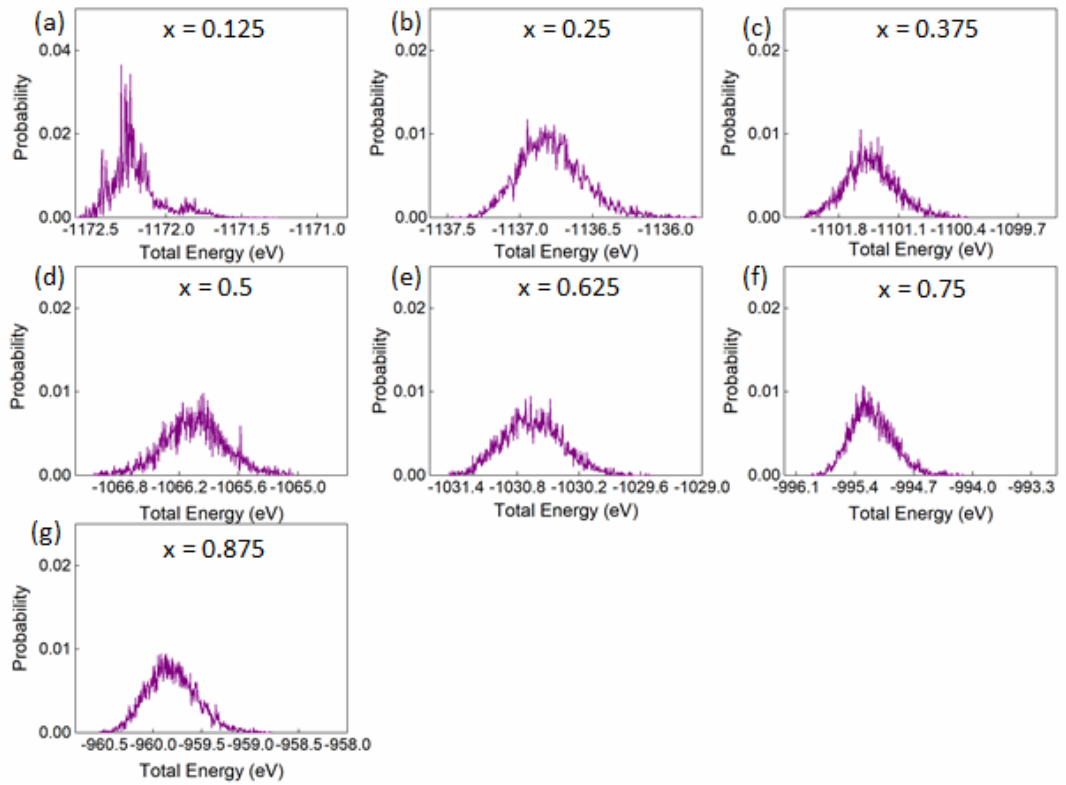


**Fig. S1:** Probability distribution from CEMC simulations for (a) x=0.125; (b) x=0.25; (c) x=0.375; (d) x=0.5; (e) x=0.625; (f) x=0.75; (g) x=0.875.

## Section 3: SQSs for different Fe composition with corresponding error correlation values

The values of the error correlation for different Fe compositions are tabulated below, and the corresponding generated SQS is also shown.

| Composition (x) | 0.125 | 0.25 | 0.375 | 0.5 | 0.625 | 0.75 | 0.875 |
|---|---|---|---|---|---|---|---|
| Error correlation | 0.0634 | 0.0443 | 0.0363 | 0.0396 | 0.0427 | 0.0458 | 0.0492 |

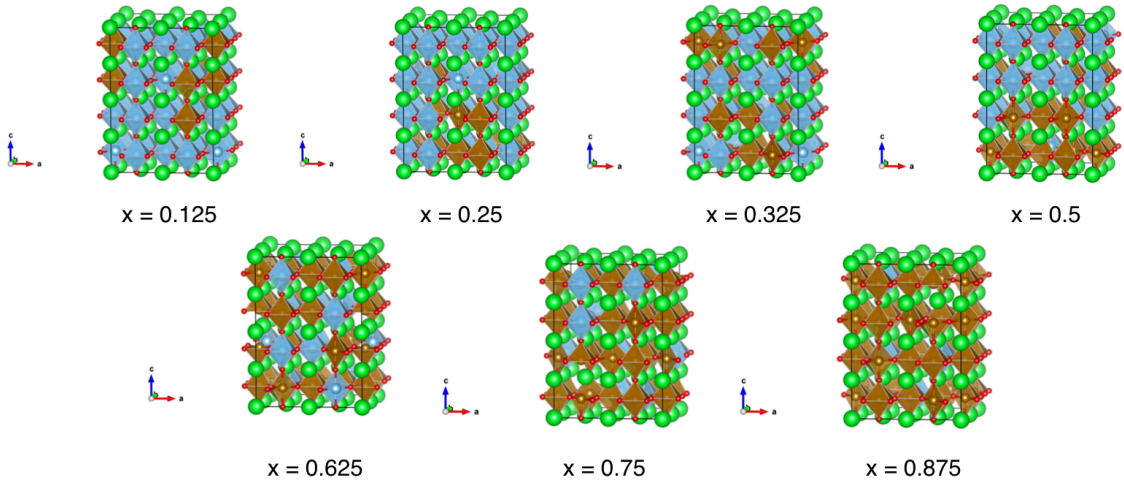


**Fig. S2:** SQSs of the $Sr(Ti_{1-x}Fe_x)O_{3-x/2}$ for different Fe composition. The labeling of the different atoms is same as the original manuscript.