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Bayesian optimization for efficient determination of metal oxide grain boundary structures

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ABSTRACT [1]

Recently, powerful methods for determining grain boundary structures with the aid of machine learning techniques have been proposed. However, the application of these methods to oxide materials has not been reported. Herein, we describe a Bayesian optimization method (Kriging) for effective and accurate determination of grain boundary structures of complex materials, namely metal oxides, including MgO, TiO_2, and CeO_2. The efficiency of this method is ~500 times higher than that of conventional all candidate calculations. We reveal that the grain boundary energy surface of metal oxides is very similar to that of metallic materials, enabling the use of the Kriging method to determine grain boundary structures.

1. Introduction

Material properties are strongly influenced by lattice imperfections. In particular, polycrystalline materials contain numerous lattice imperfections between grains, i.e., grain boundaries. The atomic structures of grain boundaries depend on the crystallographic orientation of two grains and are significantly different from those of bulk crystals. Therefore, the properties of polycrystalline materials are influenced by grain boundaries [1–6], with determination of grain boundary structure being one of the most significant tasks in materials research [7–11].

On the other hand, grain boundaries possess five macroscopic and four microscopic degrees of freedom [12], requiring extensive computations to determine a single grain boundary structure. Even for simplified coincidence site lattice (CSL) grain boundaries (Σ grain boundaries), the total number of candidate configurations to be considered is often above ten thousands.

To accurately and efficiently determine the grain boundary structures, we proposed two powerful methods based on a machine learning technique [13,14]. The first method is termed Kriging and is based on Bayesian optimization, being able to efficiently find the most plausible candidate structure corresponding to the most stable grain boundary structure [13,15]. The second method features virtual screening, where a prediction model is constructed from a training data set and is used to determine the most stable grain boundary structure [14]. Although these methods are more than several hundreds to tens of thousands times more effective than the previously reported strategies, they have only been applied to simple metallic systems, including face-centered cubic (fcc) Cu and body-centered cubic (bcc) Fe, i.e., to highly symmetric structures. The applicability of these methods to materials with lower symmetry, e.g., metal oxides, is still not disclosed.

In this study, we apply the Bayesian optimization approach, hereafter called Kriging, to grain boundaries of metal oxides including MgO, TiO_2, and CeO_2 (which commonly exhibit more complex structures than fcc and bcc metals) and determine their grain boundary structures with high efficiency and accuracy.

2. Methodology

In this study, four kinds of metal oxide grain boundaries, namely rock-salt-MgO Σ5[001]/(210) and Σ5[001]/(310), rutile-TiO_2 Σ5[001]/(210), and fluorite-CeO_2 Σ3[110]/(1-11) were selected to test the applicability of the present method. These grain boundaries have different complexities, e.g., the number of termination planes for MgO Σ5[001]/(210) and Σ5[001]/(310) is one (Fig. 1(a, b)), whereas that for TiO_2 Σ5[001]/(210) and CeO_2 Σ3[110]/(111) is two (Fig. 1(c, d)). In addition, the corresponding stable structures have been reported [16–19]. The supercell with grain boundaries was made by arranging each grain mirror-symmetrically in the y-axis direction and connected each other. The number of atoms in the supercell with each grain boundary of MgO Σ5[001]/(210), MgO Σ5[001]/(310), TiO_2 Σ5[001]/(210), and CeO_2 Σ3[110]/(1-11) was 80, 160, 60, and 72, respectively. The supercells and their lattice constants for the grain boundaries are shown in Fig. 1 and Table 1.

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[1] Coincidence site lattice, CSL; face-centered cubic, fcc; body-centered cubic, bcc; general utility lattice program, GULP.

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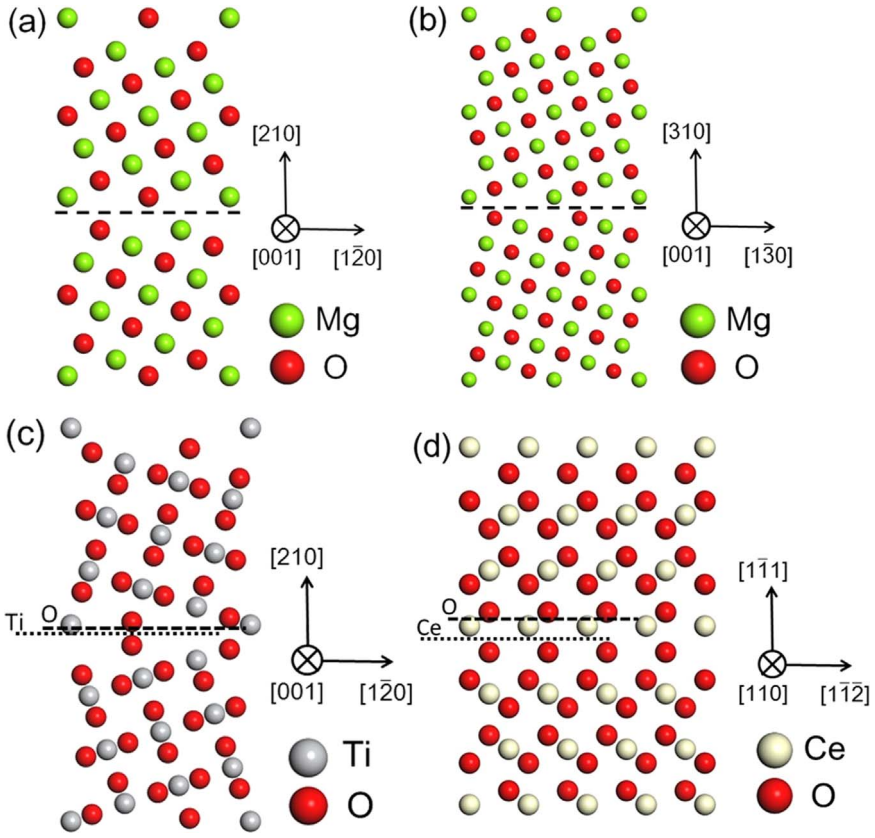

Fig. 1. Model grain boundary structures of (a) MgO Σ5[001]/(210), (b) MgO Σ5[001]/(310), (c) TiO_2 Σ5[001]/(210), and (d) CeO_2 Σ3[110]/(1-11). Red spheres represent oxygen atoms. Green, gray, and ivory spheres represent Mg, Ti, and Ce, respectively. Dotted lines indicate the position of the grain boundary.

Table 1
Lattice constants and number of configurations searched by a conventional all candidate calculation.

Grain boundary	Lattice constant			Number of configuration
	X (Å)	Y (Å)	Z (Å)	
MgO Σ5[001]/(210)	9.518	19.736 ~ 20.336	4.257	28,896
MgO Σ5[001]/(310)	13.460	27.620 ~ 28.320	4.257	40,635
TiO_2 Σ5[001]/(210)	10.275	21.250 ~ 21.850	2.959	21,630
CeO_2 Σ3[110]/(1-11)	13.254	19.444 ~ 20.444	3.826	36,309

In this study, we define the x and z directions to be parallel to the grain boundary plane, with the y direction being orthogonal to it. Three-dimensional rigid body translations were considered in increments of 0.1 Å. As a result, the number of possible atomic configurations, N, which is identical to the number of different rigid body translations, was estimated as:

$$N = \frac{L_x}{0.1} \times \frac{L_z}{0.1} \times 7$$

where L_x and L_z represent lattice constants to the x and z directions, respectively. The number of rigid body translations to the y-direction was limited to seven irrespective of the model used, because y values of 0.7–1.3 Å were considered.

The data space to be considered is shown in Fig. 2(a). Each axis corresponds to three directions of rigid body translation, and each grid point corresponds to one candidate configuration. 'Brute force calculations' afford grain boundary energies of all candidate configurations, with the obtained data space shown in Fig. 2(b). In this case, the structure optimization and energy calculations need to be performed for all candidate configurations to find the most stable (lowest energy) structure.

In the Kriging scheme, the most stable structure is determined using as few points as possible. Kriging is a geostatistical method based on Gaussian process that is utilized in resource exploration [20]. The exact Kriging procedure used in this study is same as the one reported previously [13]. Two hyper-parameters, pre-distribution and kernel parameter, were set to 0 and 3.0, respectively so that the kernel is not biased to 0 or 1. The random selection number for the initial calculation was set to five, with the actual size of three-dimensional rigid body translations in each xyz-direction acting as descriptors. The grain boundary energy, E_{GB}, is estimated as follows:

$$E_{GB} = (E_{total} - E_{bulk})/2A$$

where E_{total} is the total energy of the supercell with grain boundaries, E_{bulk} is the total energy of the supercell without grain boundaries, and A is the grain boundary area, whereas the factor of two arises from the periodic boundary condition. For structure optimization and energy calculation, static lattice calculations using an empirical potential were performed using a general utility lattice program (GULP) code [21]. Buckingham-type potentials were used for MgO (Catlow et al. [22]), TiO_2 (Bandura et al. [23]), and CeO_2 (Minervini et al. [24]). Subsequently, the joint probability distribution of the data space is extrapolated based on the calculated values using the Gaussian process, with the corresponding parameters determined in a previous study [13]. The Z-scores, calculated from the following equation, are estimated for all points in the search space: $Z - score_i = (GBE_{current\ min} - GBE(x_i))/\sqrt{\sigma(x_i)}$ where $GBE_{current\ min}$ represents the current minimum grain boundary energy, and