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High-throughput prediction of activation energy for impurity diffusion in fcc metals of Group I and VIII

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ABSTRACT

We develop a high-throughput calculation method that rigorously mines correlations embodied within experimental data and use it to direct quantum mechanical techniques efficiently toward the diffusion activation predictions and assessments. The new diffusion activation energies data of 298 binary systems, covering six faced-centered cubic (fcc) host metals of Group I (Cu, Ag, Au) and Group VIII (Fe, Co, Ni) and 76 elements of periods of 3, 4, 5 and 6, are predicted in this work. Furthermore, a new analytic expression, derived from data-mining, has been developed, which sheds light on the delicate relationship between diffusion activation energy and materials fundamental properties. The present approach can be extended to study the impurity diffusion in metal alloys of other complex structures where diffusion activation energies are difficult to obtain from first-principles calculation due to the complex diffusion mechanism. The present results could be used to provide useful data for other material simulation techniques such as CALPHAD or facilitate the choice of materials for technological applications.

1. Introduction

Mass diffusion is a fundamental process in the processing, operations, and aging of many materials. Diffusion in solids plays an important role in many technological applications such as doping for fabrication of microelectronic devices, developing solid electrolytes for batteries and fuel cells, surface hardening of steel, and developing advanced materials like superalloys with superior properties at elevated temperatures, etc. [1–3]. Systematic investigation of self-diffusion and impurity diffusion in metals has been conducted for decades [4]. In spite of extensive experiments, diffusion data of many alloy systems remain unattainable, mainly due to difficulty in measurement using conventional techniques. For instance, data of impurity elements of technetium (Tc) and osmium (Os) in Nickel (Ni) are absent, as Tc is rare and radioactive element and Os is very toxic. Determination of diffusion parameters through experimentation is not only time and resource consuming but also often clouded by various effects such as the presence of defects.

An earlier theoretical model for the calculation activation energy was proposed, derived from the elastic, thermodynamic and electrostatic properties of the systems [5]. As the authors pointed out, however, the atomic radii were not taken into account in this model although a possible influence of the radius of the impurity atom on diffusion was present. The activation energy of impurity diffusion and self-diffusion have also been empirically correlated with various factors such as the charge difference or the melting temperatures of the impurity and host [4,6], the deviating ion radius of the impurity and difference in the elastic constants [7], the binding energy of a solute (impurity) in the host [8], the metallic radius of the impurity [9], as well as the differences in charge, compressibility and atomic radius [10]. While these empirical relations correlate activation energy with a single or a few bulk properties of the constituents, it remains unclear which factors, e.g., atomic size or electro configuration, govern the impurity diffusion activation energy [11]. In fact, the diffusion activation energy may be determined by multiple coupling factors.

Owing to significant advances in both computational power and basic materials theory, it is possible to predict diffusion coefficients with an accuracy and reliability comparable with experiments. Recently, a few publications reported results of the first-principles calculations of impurity diffusion in faced-centered cubic (fcc) structured metals of Ni [12,13], Al [14] and Fe [15]. The results from all of the above methods display the similar trends, namely, impurities with radii larger than that of the host lead to low diffusion activation energy which indicates a tendency of faster diffusion. In spite of the advances in first-principles calculations, these methods are computationally intensive and time-consuming. More

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importantly, the success of the methods is highly dependent on the chosen diffusion mechanism such as the 5-frequency mechanism [16] in which the following assumptions are made: (i) (single) vacancies are the dominate defect, (ii) the impurity (or solute)-vacancy is short ranged, and the vacancy jump frequency is perturbed only in the immediate vicinity of the solute [17].

Artificial intelligence method offers an alternative solution to deal with multi-parameter relation and predict materials properties for a large amount of systems with less computation effort [18,19]. In this study, pattern recognition (PR) and artificial neural network (ANN) are employed. The success of any artificial intelligent model depends on accuracy of the input data, selection of appropriate descriptors and statistical tools. All the input data, namely, the diffusion activation energies of 152 couples are extracted from the handbook [6] in which a large body of experimental data are complied. The descriptors are chosen as the fundamental properties of the solute and host elements, and the correlation technique used in the present work is an artificial intelligence expert systems APEX (Advanced Process Expert) which was developed with MATLAB in the Institute of High Performance Computing, Singapore.

Once the ANN model is successfully trained, a linear approximation is developed to estimate the activation energy by using the significant parameters derived from ANN model. In this work, we restrict our study to the activation energy of the binary solute (impurity) diffusion in six hosts of Fe, Co, Ni, Cu, Ag and Au, i.e., fcc crystals in which diffusion can be described by substitutional mechanism. A total number of 76 elements are covered. By examining the predicted activation energies against the published experimental data, we find two impurity–host couples, i.e., Hf and U in fcc-Fe, displaying large abnormity. Thus we further investigate the two systems by first-principles calculation. In addition, among all the new results by the ANN model, a small number of systems are examined by an empirical relation [20] and compared with the data acquainted from other publications of first-principles calculation. Furthermore, the trends in the variations of the predicted and experimental activation energies with the atomic numbers are discussed for the diffusion of transition metals of 3d, 4d and 5d series in the six host metals. The present work enables a comprehensive prediction of impurity diffusion in fcc systems with much lower cost and provide critical data for further materials simulation such as CALPHAD technique. These results may facilitate the choice of materials for technological applications.

2. Computation details

2.1. Artificial neural network (ANN) method

Materials properties are quantitatively correlated with the elemental property parameters of its constituent chemical elements [21]. Diffusion activation energy has been found correlated with charge difference, melting temperature, metallic radii of impurity and host [4,6,7,10,22,23]. In order to develop a reliable ANN model to predict the diffusion activation energy, fundamental element properties of the impurity and host metals are used as the input. Chemical element properties are generally classified into six groups which can represent a chemical elements most significantly as being distinct different [21,24], namely electrochemical factor, atomic number factor, cohesive energy factor, size factor, valence electron factor, and Mendeleev number factor. Furthermore, elemental property parameters belonging to the same group (factor) can be interchanged and the results do not principally change. In the present work, five elemental property parameters, i.e., electronegativity (Pauling scale) [25], atomic number, melting temperature, atomic radii [26], and valence electron, are selected from each of the first five groups.

As mentioned previously, the focus of the present work is placed on the prediction of diffusion activation energies of impurities diffusion in six fcc host metals of Group I and Group VIII, namely, Fe, Co, Ni, Cu, Ag and Au. The impurity (solute) elements covered in the present work are elements in Period 3, 4, 5 and 6 of the periodic table, excluding halogen and noble gas elements, as shown in Table 1. The input diffusion activation energies of 152 couples of impurity–host are extracted from the handbook [6] and summarized in Appendix A. It is noted that experimental anomalies are observed in these reported activation energies with marked discrepancy (20–30%) between different measuring methods, and the data also vary with different temperature ranges. As such, we select the data measured near or above 1000 K. If data of different crystal structures are given, only those with fcc structure are selected. In addition, if multiple measurements are available, the average value is taken for each impurity–host couple. The standard deviation of these experimental data ranges from 0.2 to 37.5 kJ mol^{-1} (Appendix A).

The pattern recognition is first performed to determine significant parameters (descriptors) that govern the diffusion activation energy in fcc metals. To begin with, we select the following 17 parameters as listed in Table 2, namely, 5 elemental properties of the impurity and host, respectively, 5 elemental property differences of the impurity and host, and 2 simple non-linear combinations of atomic radii and melting temperatures of the impurity and host. The 17 parameters are reduced to significant parameters that are subsequently used to train the artificial neural network (ANN) model for the prediction of activation energy.

2.2. First-principles calculation

In order to investigate the large discrepancies in the reported and predicted results for the activation energy of diffusion of Hf

Table 1
The chemical elements studied in this work.

	Be														
Na	Mg											Al	Si	P	S
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po

Ac	Ce	Pr	Nd	Pm	Sa	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
	Th	Pa	U	Np	Pu	Am								