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Ab Initio Calculations for Search Optimization of Multicomponent Alloy Configurations

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Abstract. The paper presents an algorithm for optimization of searching configurations of multicomponent alloys that have a predetermined value of physical and mechanical properties. Values obtained by Exact MT Orbitals (EMTO) were used for calculations. The algorithm efficiency is demonstrated on an example of estimating the bulk modulus of a three-component alloy based on Ti, Nb and Zr. It is shown that the use of the algorithm can in some cases reduce the amount of calculations by 10 times or more.

INTRODUCTION

Rapid development of modern technology significantly raises requirements to properties of existing materials and encourages development and creation of new materials and alloys with predetermined characteristics. This makes the problem of determining physical-mechanical properties of crystalline materials a fundamental problem in physics of strength and plasticity. Recently new classes of materials were developed qualitatively, which is of interest both in terms of investigating their fundamental properties and applying such materials in practice. In particular, alloys with unique stress-strain properties, such as high-strength steel [1, 2] or nanostructured high strength molybdenum (Mo) alloys, [3] have been developed. The obtained alloys of titanium-niobium (Ti-Nb) are interesting not only in terms of enhanced biocompatibility, but also because they demonstrate mechanical properties that are similar to that of bones [4], which makes them an indispensable material in production of medical implants.

An experiment was and remains the dominant approach in the search for new materials. At the same time, theoretical studies play an increasingly important role in material science. Computer simulations in parallel with experimental data allow obtaining a much better understanding of the physical mechanisms that determine the conditions of synthesis and properties of materials. This increases the predictive power of theory and motivates rapid progress in the field of quantum simulations at the fundamental level. The emergence of a new field of computer modeling of material properties "from the first principles" (*ab initio*) based on Density Functional Theory (DFT) has become one of the most spectacular events in quantum theory of solids within the last three decades. A number of studies carried out in the framework of DFT allowed predicting some properties of new materials that have fundamental importance for a number of practical problems [5–7].

Despite these significant results, theoretical prediction of properties of new materials has several limitations that significantly reduce their potential effective use. In particular, inaccurate accounting of interstitial contribution to the potential and/or neglect of overlapping Maffin-Tin (MT) spheres centered on different atoms was a problem of early DFT [8]. The method of Exact MT Orbitals (EMTO) that appeared later [8, 9] allows solving this problem; however, similar to other *ab initio* methods of calculating material properties, each EMTO calculation is time consuming and resource-intensive, especially if properties are determined in nonzero conditions. This raises the problem of optimizing the volume of required computations when desired properties of a multicomponent alloy are predefined.
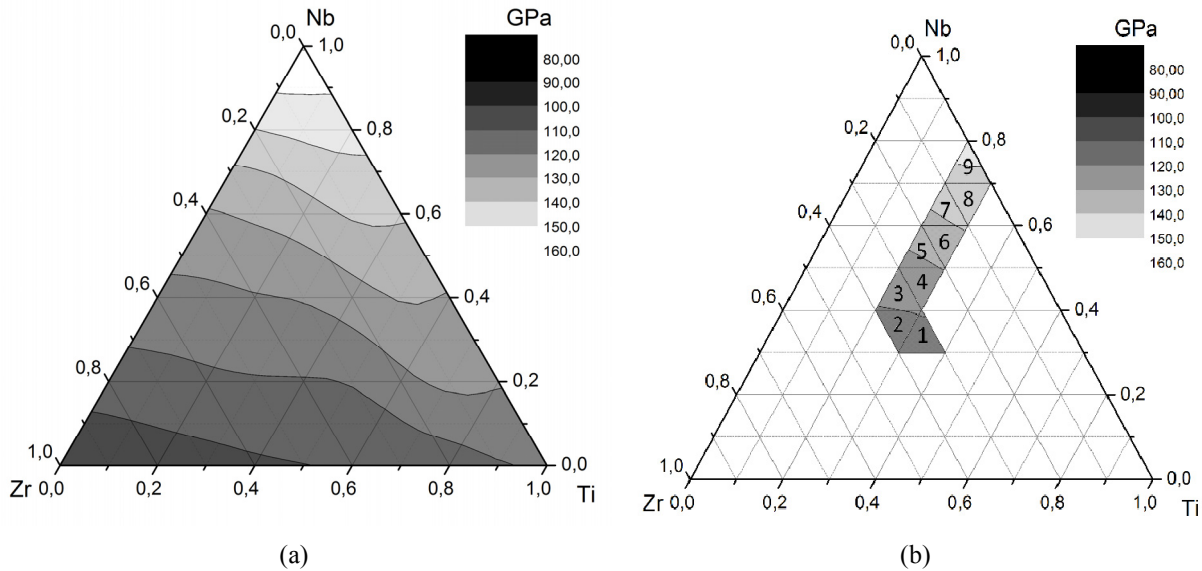
FIGURE 1. (a) distribution of the bulk modulus for a three-component alloy Ti$_x$Nb$_y$Zr$_z$ obtained using ab initio calculations in the framework of the EMTO method; (b) the scheme of the calculation optimization algorithm

The main aim of this work was to develop an algorithm that allows optimizing the amount of calculations required to determine chemical composition of a multi-component alloy with predetermined properties by using *ab initio* calculations within the EMTO method. Efficiency of the algorithm was demonstrated by calculating elastic properties of ternary alloys based on Ti, Nb and Zr.

THE METHOD OF CALCULATING MECHANICAL CHARACTERISTICS OF MULTICOMPONENT ALLOYS

The bulk modulus of a three-component Ti$_x$Nb$_y$Zr$_z$ alloy was calculated by the EMTO method combined with the Coherent Potential Approximation (CPA) [10]. The EMTO method allows accurate calculation of the one-electron Kohn-Sham state and the full one-electron energy for the optimized overlapping MT potentials using the Green's function formalism. In the EMTO theory, it is possible to achieve the accuracy of the full potential methods and simultaneously maintain the effectiveness of MT potential-based methods with a full charge density technique [9]. Combination of the EMTO method with CPA makes it possible to carry out studies of disordered alloys.

The basic set of EMTO wave functions includes *s*-, *p*-, *d*- and *f*-orbitals. In this study we have used the Generalized Gradient Approximation (GGA) [11] to describe the exchange and correlation effects in an electron gas. Integration over the irreducible part of the Brillouin zone has been performed using a 29×29×29 grid of *k*-points in the reciprocal space for a bcc lattice. Energy integration has been carried out in a complex plane using a semi-elliptic contour comprising 24 energy points. Full charge density has been represented by single-center expansion of electron wave functions in terms of spherical harmonics with orbital moments $l_{FCD}^{max} = 8$. Convergence of the energy with respect to calculation parameters has been set to 10^{-8} Ry.

Figure 1a shows the results of the EMTO calculations of the bulk modulus for a three-component alloy Ti$_x$Nb$_y$Zr$_z$ with different percentage of each element. Indices *X*, *Y* and *Z* are subject to the rule $X + Y + Z = 1$ (100%), where each index is varied in increments of 0.1. In Fig. 1a, calculated configurations correspond to the nodes of intersection of the triangular mesh, and their total number is 66.

DESCRIPTION OF THE CALCULATION OPTIMIZATION ALGORITHM

Let us demonstrate the basic idea of the calculation optimization algorithm on the example of defining the specific configuration of a three-component alloy with the bulk modulus near 140 GPa. We calculated the values of

the required variable in three neighboring points located in the vertices of a triangle with the side length corresponding to the minimum concentration step. In this example, it is 0.1 (10%). In the future, we will investigate the difference between calculated parameters for a specific alloy configuration and the required values (in this case $B(Ti_xNb_yZr_z)$-140). The ultimate goal of the algorithm is to determine the vertex of one of triangles with the minimum absolute value of obtained difference.

Originally the required variable is calculated for all vertices of a randomly selected triangle. The best choice is a triangle that is located in the central part of all considered configurations of a three-component alloy. We find the largest/smallest values by comparing the bulk modulus values that correspond to the vertices of the triangle under consideration. The highest bulk modulus is set where all three concentration values are greater than desired; in a similar fashion, the lowest bulk modulus is set where all three concentration values are less than desired. The triangle with a common face to the initial triangle is considered in the next step of the algorithm. This face is located opposite the vertex with the maximum/minimum value of the calculated parameter. Thus, it is necessary to calculate the desired parameter only in one vertex in order to determine the following step, because the other two values are known from the previous triangle. Then the sequence of operations is repeated. Thus it is possible to find a triangle in which the value of the desired parameter is as close to the target value as possible. Figure 1b shows the search path to identify the required value of the bulk modulus.

Implementation of the algorithm is represented in Table 1, where sequential numbers of the triangles was marked by whole numbers 1, 2, 3, etc. The three lines in each triangle contain information about the current concentration (in parts) of each component and the calculated value of the desired parameter obtained in vertices of the triangle. The alloy configuration excluded from further consideration (in our example, it is the smallest value of the three) is denoted in bold. The final triangle defined by the optimization algorithm has parameter values that are close to the desired ones. Further search for the desired configuration of the alloy can be implemented on one side of the triangle. At that, the concentration of only two of the three components will change. Linear interpolation of values within the final triangle can also be used to search for the exact concentration. For the studied alloy with the fixed concentration of 20% titanium, and niobium and zirconium concentration in the range from 70 to 80% and from 0 to 10% respectively, the bulk modulus can be expected near 140 GPa, which is indicative of the ternary alloy $Ti_{20}Nb_{74}Zr_{06}$.

TABLE 1. Element concentrations in $Ti_xNb_yZr_z$ and their calculated values of the bulk modulus corresponding to the vertices of the triangles, depicted in Fig. 1b

Ti	Nb	Zr	B, GPa	Ti	Nb	Zr	B, GPa
		1				6	
0.3	0.3	0.4	104.8429252	**0.3**	**0.5**	**0.2**	**120.0507819**
0.3	0.4	0.3	110.137002	0.2	0.6	0.2	124.9371527
0.4	**0.3**	**0.3**	**104.8006284**	0.3	0.6	0.1	132.4072777
		2				7	
0.3	**0.3**	**0.4**	**104.8429252**	**0.2**	**0.6**	**0.2**	**124.9371527**
0.3	0.4	0.3	110.137002	0.3	0.6	0.1	132.4072777
0.2	0.4	0.4	110.059274	0.2	0.7	0.1	137.5901745
		3				8	
0.3	0.4	0.3	110.137002	**0.3**	**0.6**	**0.1**	**132.4072777**
0.2	**0.4**	**0.4**	**110.059274**	0.2	0.7	0.1	137.5901745
0.2	0.5	0.3	115.7309698	0.3	0.7	0	137.0314744
		4				9	
0.3	**0.4**	**0.3**	**110.137002**	0.2	0.7	0.1	137.5901745
0.2	0.5	0.3	115.7309698	0.3	0.7	0	137.0314744
0.3	0.5	0.2	120.0507819	0.2	0.8	0	143.6319255
		5					
0.2	**0.5**	**0.3**	**115.7309698**				
0.3	0.5	0.2	120.0507819				
0.2	0.6	0.2	124.9371527				

CONCLUSION

The proposed algorithm can significantly reduce the required amount of computations. Estimates show that in certain cases the number of calculations can be reduced by an order of magnitude. The search for a more exact value may be implemented either as shown in the introduced example (by interpolation of values in the vertices of a triangle) or by using a finer mesh in the desired area.

It should also be noted that accuracy of the unknown parameter depends primarily on accuracy of the calculations by the EMTO method. The results presented in [12], which shows a comparison of calculated theoretical values of elastic moduli and lattice parameters with available experimental data, as well as many other similar papers give a reason to assert that the *ab initio* calculations allow evaluating materials' physical-mechanical properties with high proximity to their actual values. Significant reduction in required calculations, for example by the use of the developed algorithm, allows the use of more complex methods to calculate physical-mechanical properties of multicomponent alloys; thus, even more accurate qualitative result can be obtained.

It should be noted that the proposed theoretical method to determine parameters of multicomponent alloys cannot be completely independent. Experimental verification was and remains an essential step in implementing any theoretical approach. Another, no less important limitation of the proposed algorithm is possible existence of the so-called local minima and plateaus. In both cases, standard methods of exiting from local minima or plateaus can be applied if obtained configurations do not correspond to the required characteristics. One method, for example, is to set a random value in order to exit a plateau or local minimum. In any case, the use of the optimization algorithm seems to be reasonable when calculating properties of complex multicomponent alloys.

ACKNOWLEDGMENTS

REFERENCES

1. J. Wang, S. Afshan, M. Gkantou, et al., J. Constr. Steel Res. **121**, 97 (2016).
2. R. V. Goldstein, V. A. Gorodtsov, and D. S. Lisovenko, Phys. Mesomech. **18**(3), 213–222 (2015).
3. G. P. Grabovetskaya, I. P. Mishin, I. V. Ratohka, et al., Appl. Phys. Lett. **34**(11), 1 (2008).
4. A. Yu. Nikonov, A. M. Zharmukhambetova, N. V. Skripnyak, et al., AIP Conf. Proc. **1683**, 020165 (2015).
5. D. A. Andersson, S. I. Simak, N. V. Skorodumova, et al., PNAS **103**, 3518 (2006).
6. H. Lind, R. Forsén, B. Alling, et al., Appl. Phys. Lett. **99**, 091903 (2011).
7. A. S. Ingason, A. Mockute, M. Dahlqvist, et al., Phys. Rev. Lett. **110**, 195502 (2013).
8. O. K. Andersen, O. Jepsen, and G. Krier, in *Lectures on Methods of Electronic Structure Calculations* (World Sci. Publ. Co., Singapore, 1994), pp. 63–124.
9. L. Vitos, *Computational Quantum Mechanics for Materials Engineers: The EMTO Method and Applications* (Springer-Verlag, London, 2007).
10. L. Vitos, I. A. Abrikosov, and B. Johansson, Phys. Rev. Lett. **87**, 156401 (2001).
11. J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).
12. I. A. Abrikosov, A. Yu. Nikonov, A. V. Ponomareva, et al., Rus. Phys. J. **56**, 1030 (2014).