Effect of Metalloid Impurities on Grain Boundary Stability in Tantalum

Genrich L. Krasko

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Effect of Metalloid Impurities on Grain Boundary Stability in Tantalum

Genrich L. Krasko

U.S. Army Research Laboratory
ATTN: AMSRL-MA-CC
Aberdeen Proving Ground, MD 21005-5069

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Metalloid impurities have a very low solubility in tantalum and, therefore, prefer to segregate at the grain boundaries (GBs). In order to analyze the energetics of the impurities on the tantalum GB, the first-principles calculations were performed on a simple eight-atom supercell emulating a typical (capped trigonal prism) GB environment. The "environment-sensitive" embedding energies were calculated for hydrogen, boron, carbon, nitrogen, oxygen, phosphorus, and sulphur, as a function of the electron charge density due to the host atoms at the impurity site. The calculations showed that at the electron density typical of a GB, carbon has the lowest energy (followed by nitrogen and boron) and, thus, would compete with the other impurities for the site on the GB, tending to displace them from the GB. These energies were then used in a modified Finnis-Sinclair embedded atom approach for calculating the cohesive energies and the equilibrium interplanar distances in the vicinity of an (111) Σ3 tilt GB plane, both for the clean (CL) GB and that with an impurity. These distances were found to oscillate, returning to the value corresponding to the equilibrium spacing between (111) planes in bulk BCC tantalum by the 10th-12th plane off the GB. Carbon, nitrogen, and boron somewhat dampen the deformation wave (making the oscillations less than in the CL GB), while oxygen, phosphorus, and sulphur result in an increase of the oscillations. The cohesive energies follow the same trend, the GB with carbon being the most stable. Thus, carbon, nitrogen, and boron may be thought of as being cohesion enhancers, while oxygen, phosphorus, and sulphur result in decohesion effects.
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ACKNOWLEDGMENTS

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1. INTRODUCTION

The reduced cohesion of grain boundaries (GBs) is known to be often the controlling factor limiting ductility, and, actually the performance and reliability of high-strength metallic alloys (Briant and Banerji 1983; Guttman and McLean 1979). Intergranular embrittlement in metals is usually caused by impurities segregating towards the GBs (Lee et al. 1984; Meyers et al. 1965; Seah 1980; Seah and Hondros 1983). Impurities present in bulk concentrations of $10^{-3}$–$10^{-4}$ atomic percent can result in a drastic decrease of plasticity, drastically degrading mechanical properties of metallic alloys and, thus, posing significant technological problems. This detrimental effect of impurities on the order of parts per million may be readily understood: a simple estimate shows that a ppm amount of impurity is sufficient for saturating all the GBs in a typical grain-size polycrystal (see Table 1).

Table 1. Amounts of Impurity Atoms Sufficient for Forming a Monolayer in a Grain Boundary

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Recent progress in developing efficient methods of first-principles calculations and computational algorithms made possible systematic studies of the role of impurities in intergranular cohesion on the electron-ion level. Calculations on the supercell models of GBs with impurities in iron (Wu, Freeman, and Olson 1992, 1993, 1994a, 1994b; Krasko and Olson 1990, 1991; Krasko 1992) and tungsten (Krasko 1993, 1994) have provided an in-depth insight into mechanisms of GB cohesion/decohesion processes.

Since the first-principles electronic calculations on low-symmetry systems (such as lattice defects or GBs) are still extremely complicated and costly, semi-empirical methods based on solid first-principles foundations have been developed. Among them, the most popular is the embedded atom method (EAM) (Daw 1989; Daw and Baskes 1984; Finnis and Sinclair 1984, 1986; Daw and Baskes 1983, 1987). This method has been successfully used in a wide variety of calculations.

The purpose of this paper is to elucidate energetics of impurities on a tantalum GB and analyze the effect of impurities on the intergranular cohesion in tantalum.
Rather than doing sophisticated first-principles calculations on multiautom models of a GB, we have chosen to calculate the quantity that may be called environment-sensitive embedding energy (EE), the energy of an impurity atom in an atomic environment typical for a GB (capped-trigonal prism). Knowledge of these energies for different impurities enables one to compare the relative stability of a particular impurity in the tantalum GB. Our approach is obviously an extension of the "Effective Medium" (EM) theory (Nørskov and Lang 1980; Puska, Nieminen, and Manninen 1981; Jacobsen, Nørskov and Puska 1987; Stott and Zaremba 1980, 1982; Nørskov 1982). Earlier, the EEs of impurities in iron (Krasko and Olson 1990, 1991; Krasko 1992) and tungsten (Krasko 1993, 1994) were calculated and used in analysis of GB stability in those metals.

Having calculated the EEs for a number of impurity atoms, one can use this information on the impurity energetics in a modified EAM approach for calculating the GB relaxation. The latter calculation enables one to draw important conclusions regarding the intergranular cohesion in tantalum in the presence of a definite impurity in the GB.

2. RESULTS AND DISCUSSION

2.1 Environment-Sensitive EEs. The basic idea of the EM approach was to replace the low-symmetry system consisting of an atom plus a host matrix by a high symmetry effective system of the atom and the homogeneous electron gas of a density equal to that seen by the atom. The energy of interest was called the EE and was equal to the energy difference between the atom embedded in the electron gas and separately the isolated atom and the electron gas.

The EM theory, as a first and very crude approximation, completely neglected any covalent effects, though one could expect that a metalloid impurity in a transition metal would develop strong sp-d hybridization, resulting in covalent bonds. The introduction of covalent effects via perturbation theory (Nørskov 1982), resulted in significant corrections in the embedding function for hydrogen. Further attempts to improve upon the EM method were undertaken in recent years (Raeker and DePristo 1990; Chetty et al. 1992).

Improvements in the EM theory actually made calculations more sophisticated, spoiling the elegant simplicity of the original method. Rather than introducing corrections to the EM concept, we have chosen to perform first-principles calculations on a simplified model of a GB environment, hydrostatically varying
all the characteristic volumes, thus generating a series of "environment-sensitive" EEs as a function of the electron charge density due to the host (tantalum) atom at the impurity site. Thus, an impurity is actually "embedded" into a crystal lattice environment, rather than into an electron jellium, as in the original EM theory.

The model chosen for the GB environment is an eight-atom hexagonal supercell (Ta₂₅X, where X is an impurity atom). The supercell, together with the capped-trigonal prism coordination of the surrounding tantalum atoms, is shown in Figure 1.

![Figure 1](image)

Figure 1. The Ta₂₅X hexagonal supercell emulating a typical trigonal prism environment of tantalum atoms in the (111)Σ3 GB: a) the supercell; b) the trigonal-prism coordination (© Ta, •impurity).

A trigonal prism GB configuration follows from the theory of hard-sphere packing. Atomistic relaxation studies in iron have also shown (Hashimoto et al. 1984a, 1984b) that an impurity atom, such as phosphorus or boron, is likely to occupy an interstitial position in the center of the trigonal prism formed by iron atoms in the GB core (even if, as in case of phosphorus and boron in iron, the impurity forms a substitutional solid solution with the host). The hexagonal supercell has been chosen both because of its relatively high symmetry and its emulation of a (111) Σ3 GB environment.
We performed the spin-polarized, scalar-relativistic, Linear Muffin Tin Orbitals (LMTO) (Skriver 1984) calculations; the Von Barth and Hedin (1972) exchange-correlation and the frozen-core approximations were also used. First, a series of calculations (for six different volumes) was performed with an impurity absent from the supercell (i.e., an empty sphere of the same radius as that of the radius of the impurity's Wigner-Seitz sphere was substituted for the latter). Similar calculations were then performed for each of the impurities: hydrogen, boron, carbon, nitrogen, oxygen, phosphorus, and sulphur. The EEs were defined as follows:

\[ EE = E(Ta_6\bullet) - E(Ta_6O) - E(\bullet), \]  

where \( E(Ta_6\bullet) \) and \( E(Ta_6O) \) are respectively the energies of the supercell with and without the impurity (\( O \) stands for an empty sphere substituted for the impurity atom), and \( E(\bullet) \) is the energy of the free impurity atom. In order to make the calculations more consistent, we have chosen to use, as \( E(\bullet) \)s, the values of \( E(Ta_6\bullet) - E(Ta_6O) \) extrapolated to the zero charge density (\( n = 0 \)), which would correspond to the energies of impurities in the GB environment with the host crystal lattice infinitely expanded. The EE energies (equation [1]) as a function of \( n \), the electron charge density due to tantalum atoms at the impurity site, are presented in Figure 2.

![Graph showing EE vs. n for different impurities](image)

**Figure 2.** The environment-sensitive EEs vs. electron charge density (in atomic units [a.u.]).
Plots in Figure 2 explain the phenomenon experimentally observed in many metals, the "site-competition" effect. As one can see, in the range of electron charge density typical of a GB (0.015–0.025 a.u.), carbon has the lowest energy and, thus, would push the other impurities off the GB. To the best of our knowledge, any experimental information on the site-competition effect of impurities in tantalum is absent.

The plots in Figure 2 also reveal an important aspect of GB impurity behavior. All the plots (except for that for hydrogen) have well-pronounced minima. The positions of the minima correspond to electron density at the impurity site due to the surrounding tantalum atoms that would occur if the GB were allowed to relax in such a way as to minimize the impurity’s energy. The minima positions systematically shift toward lower densities with the impurity loosing its competitive power. Lower charge density means a more "loose" GB, less strong and more prone to decohesion. The minimization of the total GB energy (not only the energy of the impurity atom) gives the characteristic charge densities which are somewhat higher than those in the minima. However, from this point of view, only sulphur and phosphorus are the obvious candidates for being "decohesive," boron and oxygen are marginal, while carbon and nitrogen may be called "cohesion enhancers." Calculations of GB relaxation shed more light on intergranular cohesion/decohesion due to impurities.

2.2 Grain Boundary Relaxation. First-principles calculations are still too time-consuming and costly to be used for the investigation of GB relaxation. A semiempirical EAM is obviously more appropriate. In order to find both the embedding functions and the pair potentials, the EAM uses experimental data, such as cohesive energies and elastic moduli, for the system of interest. This approach, easily applicable to pure metals and compounds, may become, in fact, inappropriate if the effect of an isolated impurity atom or an impurity atom on a GB is to be studied. For example, one can, in principle, in a metal-hydrogen system, use the embedding function and the pair potentials found from experimental information on that metal’s hydrides. This information, however, can be misleading since the effect of isolated hydrogen atoms on electronic structure and cohesive properties of the metal may be completely different from that of periodic arrays of hydrogen atoms typical of hydrides. In Krasko and Olson (1991), we found that hydrogen in the iron GB does not at all contribute its electron to the iron d-valence band, contrary to a general belief that in transition metals the hydrogen’s electron will inevitably go to a d-band.

In order to resolve this difficulty, we have chosen to calculate the energy contributions due to impurity atoms in the GB by using the EEs discussed previously. Since the EAM functions are also fundamentally
dependent on the electron charge density at an atom site, the EEs may simply be added to the EAM energy of the host atoms:

$$E = \Sigma_R E_{emb}(n(R)) + 1/2\Sigma_{R,R'} V(R,R') + EE[n(R_{imp})],$$

(2)

where $E_{emb}(n)$ and $V(R,R')$ are the EAM EE and the pair potential as found for the bulk BCC Ta (we used the Finnis-Sinclair functions and parameters for tantalum [Finnis and Sinclair 1984, 1986]). The third term is the energy of the impurity atom. $R$ and $R'$ are the positions of the host atoms, $R_{imp}$ is that of the impurity, and $n(R)$ and $n(R_{imp})$ are the electron charge densities at the cite of a host atom and the impurity, respectively. The electron charge density at a given site can be taken to be a superposition of the free atom charge densities or found from more sophisticated procedures.

As mentioned previously, the GB environment we were dealing with was that of the (111) $\Sigma 3$ tilt GB. The GB structure can be represented as a succession of (111) hexagonal planes:

$$\text{...CBACBACBACBACBABCABCABCABC...}$$

(the GB plane is marked by $A$). The CBABC atomic structure of the core of the GB—clean (CL) or with an impurity—was just emulated by the supercell and is shown in Figure 1. In order to find the GB structure corresponding to a minimum of energy, equation (2), the interplanar distances were varied, while the interatomic spacings and the structure within the (111) planes were unchanged.

The counterpart of the nearest neighbor distance in the bulk BCC lattice (2.859 Å) is the distance between two tantalum atoms in the (111) direction Ta1-Ta4 (see Figure 1). As a result of relaxation, the CL GB this distance becomes larger than in the bulk (2.951 Å), while the shortest distance is the one between two Ta2 atoms (Ta2-Ta2) across the GB: 2.650 Å. In the CL GB there is a significant void (occupied by a $\star$) (see Figure 3); the distance between Ta3 atoms (Ta3-Ta3) across the void (and the GB plane) is quite large: 3.504 Å. Thus, in the CL GB the strongest interaction is Ta2-Ta2, followed by Ta1-Ta1 and Ta1-Ta2. With an impurity atom in place of $\star$, the interatomic distances and interactions change significantly. Now the shortest distance is that between an impurity atom and Ta3 (●-Ta3), the distance between Ta2 atoms across the GB being larger. The interaction between the impurity atom and
atom Ta3 becomes of utmost importance. It is actually responsible for the intergranular cohesion; this is the interaction between two Ta3 atoms across the GB via the impurity atom (see Figure 3) that is the strongest. This qualitative picture explains why even the tiniest amounts of "bad" impurities at the GB may cause detrimental worsening in metal's mechanical properties. On the other hand, an impurity with strong interaction with the host atoms will result in cohesion enhancement.

The GB relaxation calculations also show that, as in the case of the iron and tungsten GBs (Krasko and Olson 1990, 1991; Krasko 1992, 1993, 1994), the interplanar separations oscillate as a function of distance from the GB, the deformation waves decaying by the 10th–12th plane away from the GB (Figure 3). An interesting feature of the CL GB relaxation is that the distance between the second and third planes is less than half of the (111) interplanar distance in bulk BCC tantalum (0.430 Å vs. 0.954 Å). This "ω-phase" effect (collapsing of two (111) planes) has been also found in iron (Krasko and Olson 1990, 1991; Krasko 1992) and tungsten (Krasko 1993, 1994). The simple physical explanation of it is that (as one can see from Figures 1 and 3) the Ta3 atoms across the GB move toward each other simply to decrease the volume of the void between them. When an impurity atom sits in the void, the ω-phase effect disappears.
Figure 4. GB relaxation. Plots show distances $d(i-1,i)$, in angstroms, between planes #i-1 and i (the GB plane is $i = 0$). (Open circle plots in all cases correspond to relaxation of CL GB and are identical.)
The impurity atoms, carbon and hydrogen, cause some "damping" of the relaxation deformation waves (i.e., decreasing the oscillation amplitudes with respect to those in a CL GB).* The amplitude increases with oxygen, phosphorus, and sulphur. Damping the deformation wave may be interpreted as "cohesion enhancement," while increasing the deformation wave oscillations may be thought of as resulting in "decohesion."

Carbon and nitrogen are known to be GB cohesion enhancers in steels, while oxygen, phosphorus, and sulphur (strong embritlers) are believed to weaken the GB cohesion. There is evidence that oxygen in parts-per-million concentrations results in intergranular embrittlement of tantalum (Diaz and Reed-Hill 1979). At the same time, the addition of carbon seems to somewhat neutralize the adverse effect of oxygen (Kumar, Mosheim, and Michaluk 1994). The latter may be a result of the site-competition effect (a site-competition cleansing). Unfortunately, reliable experimental information on effect of impurities on tantalum mechanical properties is still lacking.

Hydrogen is one of the worst embritlers in many metals. The EAM calculations (Raeker and DePristo 1990) have shown that a hydrogen atom on a nickel GB does weaken the metallic bond across the GB, lowering the fracture stress by some 15%. The first-principles calculations in iron (Daw and Baskes 1983, 1987) suggest that one of the decohesive factors is a weak Fe-H-Fe bond across the GB. Experimental information on the effect of hydrogen on tantalum mechanical properties is also lacking.

2.3 Grain Boundary Stability. From a thermodynamic point of view (Rice and Wang 1989; Anderson, Wang, and Rice 1990), the impurity’s embrittling potency depends on the difference between the free energies of the impurity’s segregation on the initial GB and on the two free surfaces emerging upon fracture. The higher the difference, the stronger the embrittling potency of the impurity. As a less rigorous but simpler criterion, in Seah (1980) and Seah and Hondros (1983) the sublimation energy differences between the host and impurity were compared in an ideal solution model for over 60 elements. According to that criterion, in tantalum, only nitrogen, boron, and carbon are cohesion enhancers (with carbon being the strongest) (Figure 5).

* At the present time, we cannot explain such a behavior of hydrogen; its EE curve, in Figure 2, is quite different from that of carbon, though the relaxed interplanar distances are almost identical.
Figure 5. The diagram: sublimation energies vs. atom size calculated from an ideal solution model in Seah (1980) and Seah and Hondros (1983). (Elements above and below the broken line, respectively, increase and reduce the fracture energy in tantalum.)

The effects of impurities on GB stability can be also analyzed by simply comparing the GB energy differences between the GB with impurities and the clean GB. The corresponding values for the impurities discussed are plotted in Figure 6. One can see that the $\Delta E$ values are more negative again for boron, carbon, and nitrogen than can be thought of as "cohesion enhancers," and less negative for the potential embrittlers; hydrogen, oxygen, phosphorus, and sulphur. Hydrogen should be the worst embrittlter; its $\Delta E$ is only slightly negative (a positive $\Delta E$ would mean instability of the GB with impurity at absolute zero temperature).
Figure 6. $\Delta E = E_{GB}(X) - E_{GB}(\text{cln})$, the energy difference between the GB with impurity $X$ and clean GB, vs. the periodic chart group number.

3. CONCLUSIONS

The environment-sensitive embedding energies (EEs)—the energies of an impurity atom in an environment typical of a GB in tantalum as a function of electron charge density at the impurity site—were calculated for a number of impurities. The knowledge of these energies enabled us to predict the site competition effect in tantalum, and draw conclusions about the relative stability of GBs with different impurities. The calculation of GB relaxation also revealed some interesting features of the GB. Since the EEs can be easily calculated for various host environments, a modified EAM can be used to analyze the impurity's energetics in a variety of situations.
4. REFERENCES


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