Computational Materials Science 139 (2017) 39–47

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Supercell size convergence testing in uniaxial tensile test studies of an Al grain boundary: A proposed path to a robust analysis

F.J.H. Ehlers [a,b,*], M. Seydou [a], D. Tingaud [b], F. Maurel [a], Y. Charles [b], S. Queyreau [b]

[a] University Paris Diderot, Sorbonne Paris Cité, ITODYS, UMR 7086 CNRS, 15 rue J.-A. de Baïf, 75205 Paris cedex 13, France
[b] Université Paris 13, Sorbonne Paris Cité, Laboratoire des Sciences des Procédés et des Matériaux, LSPM, CNRS, UPR 3407, 99 avenue Jean-Baptiste Clément, F-93430 Villetaneuse, France

ARTICLE INFO

Article history:
Received 19 May 2017
Received in revised form 20 July 2017
Accepted 21 July 2017

Keywords:
Aluminium
Grain boundary
Traction-separation curve
Density functional theory
Cell size convergence
Linking length scales

ABSTRACT

The issue of supercell size convergence for a metal grain boundary (GB) traction-separation curve is addressed through an atomistic first principles based uniaxial tensile test. Focussing on the face-centred cubic Al Σ5 [1 0 0] 36.87° twist GB, the cell size convergence problem is reformulated to involve a local region hosting a fixed number of atoms, fully contained in each supercell under investigation. In contrast with the original task, the modified problem is shown to be easily tractable: convergence is achieved when the local region covers the part of the GB environment affected significantly structurally and electronically by the GB. For the chosen test system, this criterion is met at a very modest region (and hence supercell) size. It is proposed that a robust integration of the results in a larger scale model scheme may be accomplished by linking the atomistic traction-separation curve to a relation that connects structural distortions on either side of the local region boundary. Application to other CSL-based metal symmetric GBs should be straightforward.

1. Introduction

Over the last two decades, the atomistic first principles based study of grain boundary (GB) supercells has established itself as an invaluable tool for examining fracture energies, and especially impurity promoted intergranular embrittlement (see, e.g. [1–4]). The success of this modelling framework rests on a favourable trade-off between model system size and computational precision. At the heart of the scheme, density functional theory (DFT) [5,6] is able to provide a high accuracy in the description of the chemical bond. At the same time, DFT based calculations remain feasible for systems containing a few hundred atoms, allowing for a reliable description of a series of structurally simple GBs and their vicinity. When integrated in a multiscale modelling scheme, these atomistic simulations may provide robust information on the GB energetic response to loading. Such knowledge would otherwise have to be estimated on the basis of a more conjectural approach.

As a direct consequence of the diminutive size (typically a few nm^3) of the model system, DFT based GB studies as presented in the above are strictly concerned with *local* properties. From a mesoscopic perspective, this should not be a cause of concern as long as the GB under investigation is considered free of local defects. In this scenario, the stress field spatial variation in the GB plane should be small compared to the supercell dimensions, with DFT based studies hence being perfectly suitable for predicting the (local) GB response. From an atomistic perspective, by contrast, the supercell size generally requires that the details of the structural coupling to the surrounding grains be carefully addressed, even for the case of a perfect GB with no added impurities. Fig. 1 shows a schematic illustration of a standard GB supercell (see, e.g. [7]), hosting a symmetric GB and resting on periodic boundary conditions (PBCs). The chosen cell is entirely filled with atoms and containing two (equivalent) GBs for the sake of compatibility with the PBC along the GB normal n_{GB}. Evidently, the reliability in the description of the GB properties will be linked to the distance between these two GBs, i.e. to the size of the cell along n_{GB}. By comparison, the presence of PBCs in the GB plane is not connected with significant assumption, as long as the GB is defect-free and the part of the real system covered by the supercell is well away from GB edges and corners.

Despite a series of theoretical studies probing the optimal cell size choice in Fig. 1 [8–13], this question has remained elusive. Consequently, prior to any considerations on the integration of the atomistic simulations in a multi-scale scheme, key quantities connected with the GB response to loading are observed to exhibit

* Corresponding author at: University Paris Diderot, Sorbonne Paris Cité, ITODYS, UMR 7086 CNRS, 15 rue J.-A. de Baïf, 75205 Paris cedex 13, France.
E-mail addresses: flemming.ehlers@univ-paris-diderot.fr, waningaspect@gmail.com (F.J.H. Ehlers).

http://dx.doi.org/10.1016/j.commatsci.2017.07.028
0927-0256/

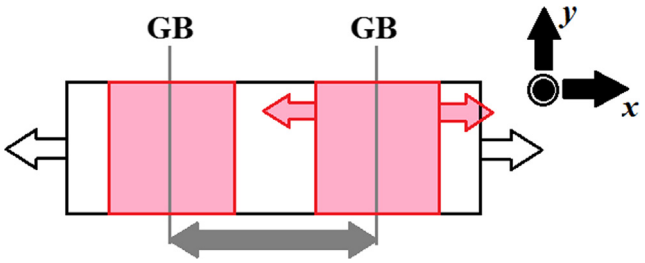

Fig. 1. Schematic representation of a simulation cell of the present work, describing a symmetric grain boundary. The two atom grains (atoms not represented) occupy the entire cell volume. The x axis denotes the grain boundary normal n_{GB} and the direction of the tensile strain (white arrows) introduced to generate fracture in the simulations. The light red boxes surrounding each grain boundary represent the regions where the loading response deviates strongly from the bulk grain. For these regions, the behaviour under local load (light red arrows) should be cell size independent, whereas the full system response depends on grain boundary distance (dark grey arrow). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

model system size dependence. For the origin of this phenomenon to be understood, the atomistic GB fracture model scheme must be detailed.

Usually, the supercell is subjected to a uniaxial tensile test, conventionally simplified as uniaxial extension [7], in the direction of n_{GB}. An energy-separation curve $E(\delta)$ is computed, describing the response of the system energy to the loading [8,14]. The atomistic *traction-separation* (T-S) curve, showing the evolution in the GB cohesive stress σ during the same process, may be obtained from $E(\delta)$ as

$$\sigma(\delta) = \left(\frac{1}{A}\right) \times \left(\frac{\partial E}{\partial \delta}\right). \quad (1)$$

Here, A denotes the GB area, while δ, the 'GB separation', defines the cell extension. For the cell of Fig. 1, both E and δ should be divided by two, reflecting the presence of two identical GBs.

The origin of the cell size dependence in $E(\delta)$, and hence $\sigma(\delta)$, is most easily explained for the case of cleavage, i.e. when no GBs are present in Fig. 1. Early investigations into this process [15,9,8] stressed that the model systems describing intact and broken configurations respond differently to a cell size increase along the normal to the fracture plane. The surface energy contribution upon fracture displays close to no variation to this modification. Hence, in the limit of large δ, the change in E compared to the case of zero load is practically fixed. For the intact configuration under load, by contrast, this quantity decreases for a fixed value of δ as the cell size grows. This follows as E displays a near parabolic variation with strain ε, which in turn is distributed homogeneously over the cell. While the picture is complicated by the presence of a GB in the system, the fundamental outcome is unchanged [12,13]. Hence, there is effectively no reason to believe that well-defined values for certain key quantities describing the GB response to the loading can be obtained other than as asymptotic limits. This involves the critical stress σ_c – the maximum tensile stress that the GB can withstand – as well as the GB breakage point [8,14]. As discussed in the literature (see, e.g. [15,9]), the DFT values obtained for both of these quantities are hugely at variance with results of continuum mechanics modelling as well as experiment.

Evidently, the reason for the cell size convergence problem is connected to the presence of both local (GB/surface energies) and global (strain energy) properties within the supercell. Different schemes have proposed that a spatial confinement of the strain to the immediate vicinity of the fracture plane be introduced as a method to produce robust fracture quantities [10,11,13]. It is possible that these schemes may capture some of the physics lost in the absence of knowledge on the optimal cell size. At the same time, however, they also involve the introduction of structural constraints in the atomistic simulations. Such constraints may compromise the accuracy in the description of the system energetic response, and hence counteract the key motivation for the DFT based studies.

The cell size convergence problem may also be viewed in a different light, however. When large enough, the cell hosts two subsystems – (i) a 'GB vicinity' that includes the region significantly affected structurally and electronically by the GB and (ii) a part farther away from this defect (see Fig. 1). When subjected to loading, these two subsystems respond differently, with the latter not truly connected with the GB response, but merely following a bulk grain behaviour. It would appear from this simple consideration that one may achieve a robust and well-defined atomistic T-S curve if the supercell is terminated at the point where the GB vicinity is fully described, but with as little as possible of the additional parts of the grain included. In practice, such a supercell based model description may be achieved if the thickness of the region defining the GB vicinity should change only weakly between zero load and GB breakage. Importantly, the computations involved would invoke no structural constraints limiting the expected accuracy.

The target of the present paper is to set up the framework required for the identification of the proposed optimal GB supercell in a uniaxial tensile test, in a manner that is easily generalisable to the set of GBs conventionally subjected to DFT studies. The selected test system for this purpose is the face-centred cubic (fcc) Al Σ5 [1 0 0] 36.87° twist GB (TGB). The paper is arranged as follows: in Section 2, the model system describing the chosen GB is introduced and computational details outlined. Section 3 describes the convergence testing framework and results obtained. In Section 4, formalism justifications and implications are discussed, with special attention paid to the coupling of atomistic and larger scale modelling. Section 5 provides summary and conclusions.

2. Model systems and computational details

The GB structure subjected to examination in Section 3 is the fcc Al Σ5 [1 0 0] 36.87° TGB (see Fig. 2(a)). Here, the notation '[1 0 0] 36.87° TGB' refers to the fact that one grain has been rotated relative to the other around the specified axis, normal to the GB plane (twist). Often, the specification of the rotation angle 36.87° is suppressed, with the nomenclature 'Σ5' providing indirect information on this issue. The Σ labelling implies a rotation compatible with the creation of a substructure – the coincidence site lattice (CSL) – that is not reoriented when crossing the GB. The number 5 specifies that the atoms of the CSL represent 1/5 of the bulk grain atoms.

The supercells used for the computational studies are constructed with close reference to the underlying CSL for this GB structure. It may be shown that the CSL unit cell basis vectors are related to the conventional fcc unit cell basis vectors of one grain as {(1,0,0); (0,3/2,1/2); (0,−1/2,3/2)}, with the first of these vectors pointing along n_{GB} (see Fig. 2(b)). It follows that each (10 atom) CSL unit cell describing a bulk grain environment has the dimension a_{Al} ($\sqrt{5}a_{Al}/\sqrt{2}$) along a basis vector out of (in) the GB plane. Here, a_{Al} denotes the fcc Al lattice parameter. For all supercells of this work, the two grains present in the cell were volume-filling. Due to PBCs, these cells hence include two GBs. As various cell sizes along n_{GB} were considered in the studies, the notation N CSL supercell was used throughout to describe the cell hosting $N/2$ CSL unit cells in each grain. Computations in Section 3 have involved the 2, 4, 6, 8, and 10 CSL supercells. In Fig. 2, the 4 CSL supercell at zero load is shown. Due to the GB presence in the