Acta Materialia 146 (2018) 42–54

Contents lists available at ScienceDirect

Acta Materialia

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

Full length article

Spectral graph theory for characterization and homogenization of grain boundary networks

Oliver K. Johnson*, Jarrod M. Lund, Tyler R. Critchfield

Department of Mechanical Engineering, Brigham Young University, Provo, UT 84602, USA

ARTICLE INFO

Article history:
Received 14 June 2017
Received in revised form
20 October 2017
Accepted 27 November 2017
Available online 21 December 2017

Keywords:
Grain boundary
Networks
Homogenization
Diffusion
Spectral graph theory

ABSTRACT

Grain boundary networks (GBNs) have a profound influence on the properties of both structural and functional materials. However, existing methods to characterize their complex structure have almost universally relied upon a binary classification of GBs as either “special” or “general”, which ignores the rich and continuous spectrum of GB types and properties. Furthermore, characterizing the aggregate network structure of GBs has proven complicated, with traditional methods focusing on local structure and also relying on a binary GB taxonomy, e.g. by evaluating how many “special” or “general” boundaries meet at triple junctions or quadruple nodes. Here we develop new structural metrics for GBNs, based on spectral graph theory, that encode both global network topology and the full spectrum of constituent GB properties, enabling high-fidelity characterization of arbitrary GBNs. Using these metrics, we derive an new structure-property relation for GBN diffusivity. The dominant term in this expression provides an efficient and accurate approximation, whose corresponding spectral metrics reveal the dominant microstructural features influencing the property of interest. The spectral index of this term serves as a type of global order parameter that reveals a fundamental structural transition in GBNs. This work provides a new framework to characterize the structure of GBNs in greater generality than previously possible and facilitates the development of new defect-sensitive structure-property models.

1. Introduction

Grain boundaries (GBs) in polycrystalline materials form a complex network of inter-crystalline interfaces, whose collective structure strongly influences many materials phenomena including strength [1,2], corrosion [1,3,4], stress-corrosion cracking [5], spallation [6], creep [7], hydrogen embrittlement [2], solar cell efficiency [8,9], superconductivity [10], and nuclear fuel performance [11–13]. Because of the complexity of their combined crystallographic and topological degrees of freedom, grain boundary networks (GBNs) have historically been characterized using reduced order metrics. Computational and experimental studies (including our own) both routinely employ a binary classification of the constituent grain boundaries as either “special” or “general” and assign all boundaries of a given type identical properties (a few notable exceptions include [14–16]). It is also common to model grain boundary networks using simple idealized geometries as illustrated in Fig. 1a. In contrast, real grain boundary networks (Fig. 1b) are composed of a continuous spectrum of grain boundary types, whose properties span many orders of magnitude and whose topology is far more complex than what is captured by such idealizations. If the structure of grain boundary networks were better understood, it would be possible to devise novel processing routes to control it and, thereby, achieve substantial performance enhancements across all classes of polycrystalline materials.

A complete description of the structure of grain boundary networks requires specification of the character (e.g. crystallography or properties) of individual grain boundaries and how they are assembled. The network structure of grain boundaries may, at first, appear random, but is, in fact, highly correlated due to crystallographic constraints, processing history, and both kinetic and thermodynamic influences. Several approaches have been devised to quantitatively characterize various aspects of GBN structure; these include tools from percolation theory (e.g. correlation lengths, cluster sizes, etc.) [18], Betti numbers [19,20], triple junction fractions [21–25], quadruple node fractions [26], swatches [27], and twin-related domain (TRD) sizes [28,29]. These descriptions focus primarily on topological structure. Those that incorporate information about GB types employ the binary “special vs. general”

* Corresponding author.
E-mail address: ojohnson@byu.edu (O.K. Johnson).

https://doi.org/10.1016/j.actamat.2017.11.054
1359-6454/

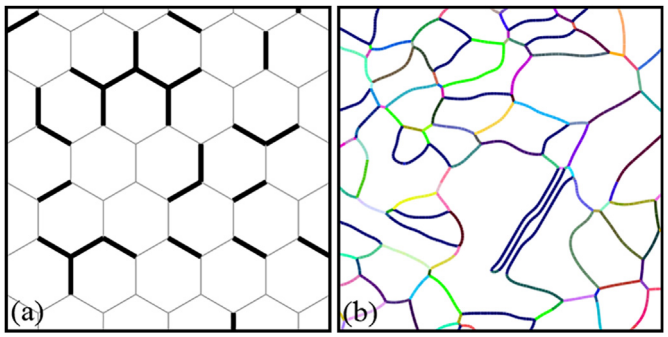

Fig. 1. (a) Idealized model of a grain boundary network with general (high-angle) boundaries in light grey and special (low-angle) boundaries in bold black lines. (b) A real grain boundary network from a sample of Inconel 690 (data courtesy C.A. Schuh) with boundaries colored according to their misorientation [17].

taxonomy of boundaries. Building on this previous work, we develop a new technique to simultaneously characterize the complex topological structure of GBNs and the full spectrum of GB types/properties in arbitrary polycrystalline materials.

2. A new approach

In the diverse fields of computer science, ecology, and biology, complex networks exist (e.g. plant-pollinator networks, or protein interaction networks) and have been analyzed using the mathematical tools of spectral graph theory (SGT) [30–34]. Perhaps the best known example is the PageRank algorithm that forms the core of Google's search engine [30,34,35]. PageRank models the complex network of internet websites as a graph (corresponding to a Markov Chain), which is represented as a matrix (the transition matrix). The PageRank—an importance metric used to determine the relevance of a website to a search query—is given by the principal eigenvector of this matrix. In other words, PageRank uses spectral decomposition of the network structure of the Web to determine its dominant features and correlate these with query relevance. By analogy, we hypothesize that spectral decomposition can be applied to the network structure of materials to characterize their dominant microstructural features and correlate these with material properties.

Studying an analogous problem for membranes, Marc Kac famously asked the question “Can one hear the shape of a drum?” [36], which was the title of a study in which the author investigated what information about the shape of a drum (or tambourine) could be inferred from its timbral spectrum. Here we ask a related question for grain boundary networks: “What can we learn about the complex structure of grain boundary networks and their effective properties by analyzing their eigenvalues and eigenvectors?”

3. Methods

3.1. Eigendecomposition of a GBN and its properties

We will consider two-dimensional GBNs, though the methods presented should be applicable to fully three-dimensional GBNs as well. A GBN is represented as an undirected weighted graph, defined by the ordered triplet $G = (V, E, W)$, via discretization, where V is the set of vertices (also called nodes), E is the set of edges, and W is the set of edge weights. Nodes (vertices) are located at triple junctions (TJs) and along GBs, and are connected by GB segments (edges) as illustrated in Fig. 2. The details of node placement and density for our particular examples are discussed in Section 3.2. In addition to the standard nodes, we create a

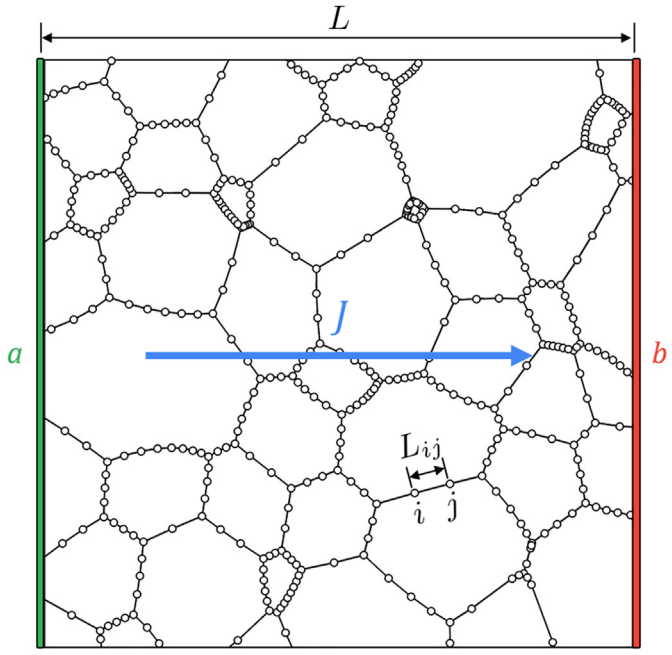

Fig. 2. Discretization of a GBN showing nodes and edges. The source and sink supernodes are shown in green and red, respectively. The direction of the macroscopic flux, J, is indicated along with the sample length, L. Representative nodes, i and j are labeled along with the length of the edge connecting them, L_{ij}. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

supernode on one side of the microstructure that represents a diffusion source (node a in Fig. 2) and another supernode on the opposite side representing a diffusion sink (node b in Fig. 2). These supernodes span the entirety of their respective side of the sample and have many edges, whereas normal nodes in these GBNs possess only 2 or 3 edges. We use periodic boundary conditions in the direction orthogonal to the macroscopic flux (i.e. GBs that exit the top of the microstructure re-enter at the bottom with identical diffusivity so that flux across the periodic boundary is continuous).

By exploiting an analogy with electrical resistor networks [37,38], the effective diffusivity of an arbitrary GBN can be expressed as:

$$D_{\text{eff}} = \frac{L}{A}\left[(\mathbf{e}_a - \mathbf{e}_b)^T \mathscr{L}^+ (\mathbf{e}_a - \mathbf{e}_b)\right]^{-1} \tag{1}$$

In Eq. (1), $\mathbf{e}_a$ is a vector who's a-th element (corresponding to the index of the source supernode) is equal to 1 and all others are 0, $\mathbf{e}_b$ is defined similarly. L and A are the sample length and cross-sectional area, respectively (without loss of generality, we assume the sample to be square with some out-of-plane thickness H so that $A = LH$). For the present work we considered microstructures with $L = 1$ mm, $H = 0.1$ mm, and $A = 0.1$ mm^2. The superscript ^{T} denotes the usual transpose operation. The superscript $^+$ indicates the Moore-Penrose pseudoinverse of the matrix $\mathscr{L}$, where $\mathscr{L}$ is the weighted Laplacian of the GBN graph. $\mathscr{L}$ encodes the topology of the GBN and the properties of each GB. With edge weights defined as $W_{ij} = D_{ij}A_{ij}/L_{ij}$, the elements of $\mathscr{L}$ are given by

$$\mathscr{L}_{ij} = \begin{cases} \sum_{i\sim m} D_{im}A_{im}/L_{im} & \text{if } i = j \\ -D_{ij}A_{ij}/L_{ij} & \text{if } i \sim j \\ 0 & \text{otherwise} \end{cases} \tag{2}$$

where D_{ij}, A_{ij}, and L_{ij} are, respectively, the diffusivity, cross-