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Molecular mechanics of polycrystalline graphene with enhanced fracture toughness

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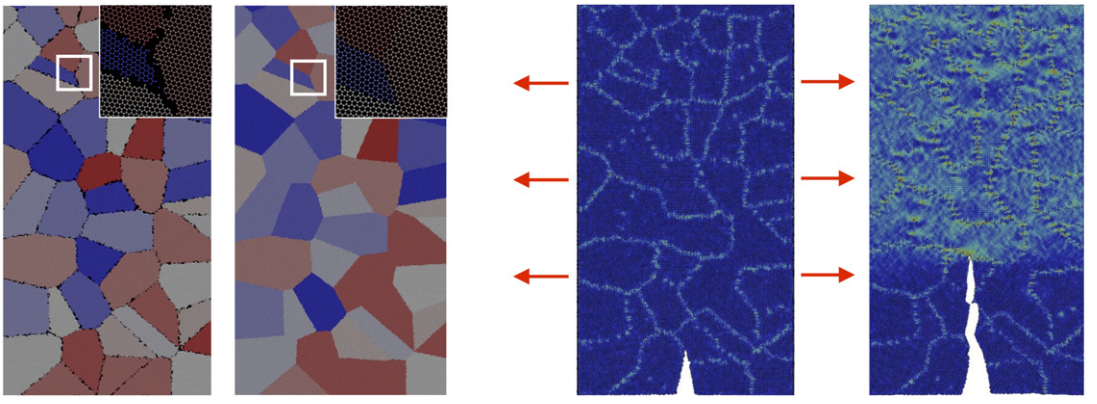
GRAPHICAL ABSTRACT



HIGHLIGHTS

- A novel algorithm is proposed to generate well-stitched polycrystalline graphenes.
- The defects of our model are well matched with the experimental observation.
- The enhanced mechanism of fracture toughness of polycrystalline graphene is studied.
- The measured fracture toughness shows good agreement with previous experiments.

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ABSTRACT

Although polycrystalline graphene generated by chemical vapor deposition features defects at grain boundaries, experimental results show that the strength of polycrystalline graphene is comparable to that of the pristine graphene. This is in contrast to the widespread knowledge that defects typically weaken a material's strength. Here, we examine why polycrystalline graphene has high strength and high fracture toughness,

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by combining an innovative algorithm with classical molecular dynamics simulation to systematically build well-stitched (99.8% heptagon and pentagon defects without void) polycrystalline graphene models with regular and irregular grain boundaries, and use these models to systematically examine the fracture toughness of polycrystalline graphene composed of grains of different characteristic length. Our study reveals that polycrystalline graphene under fracture releases up to 50% more energy than the pristine graphene. Per mechanism, we find that grain boundaries increase the critical energy release rate to fracture by reducing stress concentration and making branches near the crack tip. We conclude that these effects are likely governed by the out-of-plane deformation of polycrystalline graphene.

1. Introduction

Graphene is an ultrathin material composed of only a single layer of carbon atoms. Pristine graphene represents the strongest tensile strength of 130 GPa [1], making it ideal for applications under extreme mechanical, thermal and electronically conditions with exceptional performance [1–3]. For example, it can be imbedded in matrix materials for enhanced mechanical strength and thermal and electrical conductivity [4]. It is critical to precisely measure its strength before using it in applications. Comparing to mechanical exfoliation of graphene, which provides small flakes during peeling, chemical vapor deposition (CVD) is a much more promising technique for large-scale production [5]. However, graphene produced by this means is polycrystalline, having pentagon–heptagon defects distributed along the grain boundary as well as different chirality for neighboring grains [6], as illustrated in Fig. 1(a) and (b). Those defects may cause stress concentration, initiate crack propagation and weaken the material strength [7,8]. Yet it is also known that the existence of a grain boundary may influence the movement of crack and enhance the fracture toughness. It is intriguing how the fracture properties of graphene can be affected by the different geometries of grain boundaries.

Former studies mainly have been focused on the strength of a single graphene grain boundary [7]. Theoretical and computational studies have found that the strength is affected by the arrangement of defects, and it can have values as low as 33 GPa. The results of indenting tests show that the strength of a well stitched polycrystalline graphene consistently exhibits high values above 90 GPa, which is slightly smaller than that of the pristine graphene but much larger than the prediction of the strength of polycrystalline graphenes [9]. These results suggest that the weak grain boundary does not have a significant effect on the fracture strength of a large piece of graphene. This finding is aligned with what is observed in several biological materials, such as nacre and bone, which are composed of mineral plates and weak interfacial protein materials but have high fracture toughness [10,11].

Modeling of polycrystalline graphene and systematic analysis of how its strength and toughness alter with its geometry are crucial to understand its failure mechanism under extreme loading conditions. Based on the high-resolution transmission electron microscopy data, the shapes of grains are generally irregular, and the defects are mainly 5–7 ring pairs to reduce the total energy and the tensions of defects [6,12]. However, previously suggested models are limited to mimic the geometry revealed by experiments because the model has limited shape of regular grains or the grain boundaries have numerous voids with irregular grains [8,13]. In this work, we report an innovative algorithm that is applied to generate well-stitched polycrystalline graphene with any grain shape and any grain orientation, showing mainly 5–7 ring pairs without any voids.

In polycrystalline metals, the grain size plays a key role in their strength [14]. It is also reported that the strength of polycrystalline graphene depends on its grain size in MD simulation [8,13]. However, since defects generally exist in polycrystalline graphene, its fracture toughness is more critical for the applications and the mechanical strength measurement in experiments [15]. It is not understood how grain size and geometry affect the fracture toughness of polycrystalline graphene. Although some experiments have measured the fracture toughness of polycrystalline graphene, their experimental samples are not well characterized for grain size. Thus, the results provide a range of fracture toughness values, making the dependence of fracture toughness on grain size still elusive [16]. The knowledge of the fracture toughness is vital for the application of CVD to generate graphene with reliable mechanical performance as the grain sizes are sensitive to the growing conditions [5].

2. Material and methods

To understand the mechanical effect on grain boundary in graphene, it is crucial to obtain a fully equilibrated structure that mimics the grain boundaries obtained in CVD experiments. Here we develop a homology method to generate an entire piece of graphene sample composed of multiple grains. This method allows us to control the shape, size and crystal orientation of each grain and it automatically reconstructs the boundary by removing and adding atoms. Since the models of polycrystalline graphene in previous studies have voids and considerable non-pentagon–heptagon defects [13,17], our algorithm is especially unique and innovative. (See supplemental information for modeling details, as well as a supplemental video illustrating the process of generating the grain