

Inverse Hall-Petch Nature of Fe-Ni-Cr-Co-Cu High Entropy Alloy at Atomic Scale

M Zaenudin^{1*}, Adhes Gamayel¹, YKP Saleh¹, MN Mohammed²

¹Department of Mechanical Engineering,

Jakarta Global University, Boulevard Raya St. No. 2, Tirtajaya, Sukmajaya, Depok 16412, INDONESIA

²Mechanical Engineering Department, College of Engineering,

Gulf University, Sanad 26489, BAHRAIN

*Corresponding Author: mzaenudin@jgu.ac.id

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Abstract

High entropy alloy is a class material composed of several principal elements with equiatomic composition. The entropic of this material with near equiatomic configuration composition can theoretically decrease the strength-ductility trade-off, which differs their application from their classical alloy counterpart. The strength of nanocrystalline materials, known as the Hall-Petch phenomenon, tends to increase. Still, when it hits the critical grain size, the softening behavior of decreasing strength is observed and known as the inverse Hall-Petch phenomenon. Although there are numerous reports on the inverse Hall-Petch effect, both numerically and experimentally, the deformation mechanism of these materials is not clearly described, especially at the atomic scale. This study aims to tackle the issue by reporting the molecular dynamics simulation of nanocrystalline Fe-Ni-Cr-Co-Cu high entropy alloy with an average grain size of 7.3 nm³, 6.8 nm³, and 6.4 nm³. The study shows that the tensile deformation is mainly attributed to the grain boundary sliding due to the high grain boundary volume within the system. However, in the case of compressive deformation, at the earlier stage, the deformation is mainly focused on the grain boundary deformation, although the new cluster of grain has infrequently happened due to the compression. Both tensile and compressive strength exhibit the inverse Hall-Petch nature of Fe-Ni-Cr-Co-Cu high entropy alloy.

1. Introduction

The development of high entropy alloy (HEA) was first introduced by two different research groups in 2004 [1,2], which were concerned with exploring the parts of alloy design where it is yet to be extensively explored. Instead of focusing on conventional alloy with one primary element, extensive research on HEA explores the multicomponent phase diagram, where all alloy elements are equally concentrated; thus, there are no such base elements [3]. This approach opens the vast compositional space of the HEA system to be explored. However, the challenge of exploring the enormous compositional space of the HEA system is significant, and in the case of experimental studies, the trial-and-error approach is arguably impractical. For example, five elements of HEA mathematically have hundreds of possible compositions; hence, they are too large to be explored experimentally. Some elements affect a specific property more than others; thus, a focused study of a particular property is sometimes necessary [4,5].

Moreover, some aspects within the alloy may have a more pronounced effect on specific properties. In contrast, the others may play a secondary role, adding another layer of complexity to the investigation process.

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Thus, a more focused study on a specific property or a subset of compositions becomes necessary to gain deeper insights into the underlying mechanisms governing the alloy's behavior. The unique characteristics of HEA, which consists of several equiatomic elements theoretically, offer the possibility of decreasing the strength-ductility trade-off from its classical alloy counterparts [6–8]. The theoretical superior property is due to its high configurational entropy (many ways to place atoms in a crystal lattice), which allows for stabilizing a solid solution over a competing intermetallic phase [9]. Several HEAs have been introduced with different processing routes with advantageous properties in recent years [10,11].

The property of an alloy is not necessarily dependent only on the compositional configurations but is also somewhat influenced by its processing method and microstructure [12]. In the case of the strength and hardness of a material, it is known that as the size of the microstructure—and thus grain size—is decreased, the strength and hardness of a material is increased [13]. This behavior is known as the Hall-Petch effect. The known behavior thus stimulates the development of a processing route to produce a smaller microstructure until a nanoscale regime [14]. However, as the grain size reaches a specific limit (which could be different in every alloy), the study shows that the strength and hardness of an alloy decreased [15]. Although, in the early reported research, there is skepticism about the finding [16], the recent study has confirmed the finding experimentally and numerically [17,18].

This paper uses molecular dynamics simulation to shed light on the inverse Hall-Petch nature of the Fe-Ni-Cr-Co-Cu high-entropy alloy at the atomic scale. Molecular dynamics is a robust method for studying the materials' behavior and properties at the atomic scale [19-21], which is suitable for studying the inverse Hall-Petch phenomenon. This study aims to determine the factors contributing to the observed decrease in strength with decreasing grain size and to elucidate the interplay factors between various microstructural-deformation features, such as dislocations, grain boundaries, and segregation of alloying elements. Understanding the role of each constituent component of the inverse Hall-Petch behavior offers the possibility of tailoring the alloy's microstructure and composition to optimize its properties for specific engineering applications.

2. Simulation Procedure

Firstly, the AtomSK [22] software is utilized for the pre-processing step to generate the structural model of the HEA system. AtomSK is capable of creating molecular systems with grain generation for crystalline alloys. In this study, the HEA model consists of Fe-Ni-Cu-Co-Cr elements and is constructed with face-centered cubic (FCC) crystal structures. Also, the boundary is set to periodic. This approach helps approximate a system's behavior as if it were in an infinite bulk environment, thus reducing the artificial effects of the system's finite size. The size of each model is approximately $20 \times 20 \times 20$ nm. The number of grains is set to 20, 25, and 30, respectively. Each grain has a different crystallographic orientation, such as [100], [010], and [001] in the x, y, and z directions, respectively. The distribution of the constituent elements within the HEA model is constructed at an approximately equal ratio. The model of polycrystalline HEA is depicted in Figure 1. The model consists of a nanocrystalline structure with an average grain size of 7.3 nm^3 , 6.8 nm^3 , and 6.4 nm^3 , for Models A, B, and C, respectively. The average grain size in this study is below the average value of the grain size of HEA, which is experimentally reported to experience softening behavior as theoretically described in the inverse Hall-Petch effect, which is around 10 nm [23].

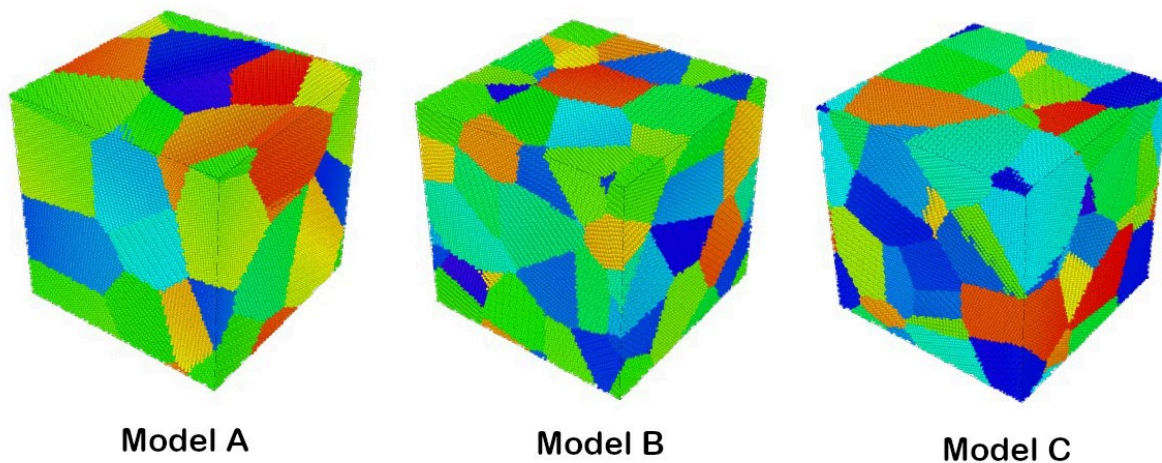


Fig. 1 High entropy alloy atomic model with a different number of grains: (A) 20 grains, (B) 25 grains, and (C) 30 grains

Secondly, the LAMMPS software [24] simulates the HEA system by integrating the Newtonian equation of motion into an ensemble of atoms. The objective is to investigate the properties and behavior of the HEA alloy with varying numbers of grains. The simulation is carried out under equilibrium conditions with a constant temperature. The timestep used in the simulation is typically set to 1 femtosecond, allowing for the capture of molecular dynamics phenomena with high resolution. For the post-simulation, we utilized OVITO [25] to help analyze the simulation results from the dump file produced by the simulation. Several methods, such as atomic strain for calculating the shear strain, allow us to identify whether the strain-induced deformation exists and whether the polyhedral template matching the identified structural features of the materials. OVITO is widely used and known for its flexibility, whether with a graphical user interface or through coding via Python programming.

The simulation process is divided into two main steps: initialization and production. During the initialization step, the HEA system is equilibrated at room temperature and atmospheric pressure for a certain period, typically around 30 picoseconds (ps). This equilibration process minimizes the system's energy and reduces undesirable effects, such as fluctuations, before the primary simulation. It ensures a stable starting point for the subsequent production step. The simulation is conducted at the desired temperature (kept constant) with zero pressure in the production step. The simulation time is generally set to capture the system's behavior over a sufficient timescale. In this study, the focus is revealing three phenomena related to the Hall-Petch theory: the tensile and compressive behavior of the HEA alloy with different numbers of grains. The tensile and compressive test is subjected in the z direction with both strain rates set to $1.0e^{10}/s$. For all the simulations, the details of the settings are as follows:

1. Tensile test: the tensile test is performed with a strain rate of $-1.0e^{10}/s$. The temperature during the tensile test is set to 300 K. This simulation is run for 150 ps, thus giving a precise stress-strain curve or the mechanical behavior related to its tensile strength.
2. Compressive test: the compressive test is performed at 300 K with a strain rate of $-1.0e^{10}/s$. The simulation is run for 75 ps. This simulation can only be run for a certain period, for which the length of the sample is reduced with time; thus, the sample length cannot be less than 0, or the simulation will show an error.

These molecular dynamics simulations focus on the Fe-Ni-Cr-Co-Cu High Entropy Alloy (HEA); thus, choosing an appropriate interatomic potential is essential for obtaining reliable and accurate results. Since the system comprises multiple elements (Fe, Ni, Cr, Co, and Cu), a specialized interatomic potential is required to accurately describe this high entropy alloy's unique behavior. For metallic systems like HEAs, the Embedded Atom Method (EAM) has proven to be a highly effective potential due to its ability to capture metallic materials' complex interactions and properties; thus, this EAM method is widely used in atomistic simulation.

In this study, the EAM interatomic potential developed by Farkas *et al.* in 2018 is adopted [26]. The following mathematical function describes this potential:

$$E_{tot} = \frac{1}{2} \sum_{ij} \Phi_{a_i-a_j}(r_{ij}) + \sum_i F_{a_i}(\bar{\rho}_i) \quad (1)$$

Breaking down the equation, it consists of:

1. $\sum_i \Phi_i(\rho_i)$ represents the embedding energy contribution, where Φ_i is the embedding energy function for atoms i , and ρ_i is the electron density at the site of atom i . This term captures the interaction of each atom with the surrounding electron cloud, accounting for the cohesion and stability of the HEA.
2. $\frac{1}{2} \sum_{ij} \Phi_{ij}(r_{ij})$ represents the pair interaction contributions. Here, Φ_{ij} is the pair potential function between atoms i and j , and r_{ij} is the distance between them. This term accounts for the interactions between neighboring atoms, describing bond strengths and repulsions within the atoms in the system.

3. Results and Discussions

The Hall-Petch phenomenon is indicated by the increase of the materials' strength as the decrease of materials grain size, especially metal. However, to some degree, the phenomenon comes in reverse, where the reduction in grain size no longer increases the materials' strength but decreases it. This so-called inverse Hall-Petch effect happens when the grain size is too small for most grain boundary deformation to occur and transforms the primary deformation mechanism into a slip between the grain boundaries, also known as grain boundary sliding. In this study, during the tensile test, the deformation of the system is dominated by the slip between the grain boundaries, as depicted in Figure 2. However, the grain boundary deformation still occurred in a small number. With this grain size, the grain boundary volume has a higher fraction; as a result, grain boundaries are thought to have a significant impact on the deformation process of this material and only a small strain is observed within the grain [23]. The corresponding stress-strain curve of this uniaxial tensile test is depicted in Figure 3. As reported in refs [27,28], the tensile elongation of nanocrystalline Fe-Ni-Cr-Co-Cu HEA is relatively low compared

to the several other HEAs with bigger grain sizes. This behavior is attributed to plastic instability from the lack of an effective hardening mechanism, which manifests as shear bands. This phenomenon also means the chance for an intermetallic diffusion of atoms between the neighboring grains is infrequent.

The deformation mechanism discussed then leads to the uncommon behavior of metallic alloy when the grain size is so tiny, called the inverse Hall-Petch effect, where the yield strength of the materials is decreased from Model A, Model B, and Model C in the order from the bigger grain size to the smaller grain size. Although the size of every grain may significantly vary between each other since it is constructed randomly following the Voronoi tessellation from the AtomSK software package [22], differences and decreases in grain size in Models A, B, and C can be calculated in average, with the information from the different amount of grain from the systems from Model A, B, and C and with the same total dimension of the system. The tensile strength values related to Figure 3 are 6.23 GPa, 5.57 GPa, and 5.38 GPa for Models A, B, and C, respectively. The decrease in the tensile strength confirms the inverse Hall-Petch effect from the Fe-Ni-Cr-Co-Cu HEA.

In the case of uniaxial compressive stress, a more substantial difference from the uniaxial tensile stress is observed, as depicted in Figure 4. The deformation during compressive stress is not only focused on the grain boundary but somewhat deformed evenly over the system. The amount of crystal with a face-centered cubic structure (denoted with green-colored atoms) is decreased with the simulation time, and a random structure is formed. This unexpected structure often creates its cluster, that is, when it is separated or divided from its original grain into several clusters. However, this phenomenon is not necessarily the primary deformation mechanism since the grain size is so tiny; hence, the grain boundary sliding more often happens in the system.

Furthermore, compressing materials to produce a finer grain is already widely known for severe plastic deformation. However, producing a smaller grain size is hard until a specific limit is reached, which may differ in every alloy [29]. This simulation proves that severe plastic deformation effectively creates a better grain structure, that is, a smaller grain size (microstructure to nanostructure) until a certain limit from the as-received material.

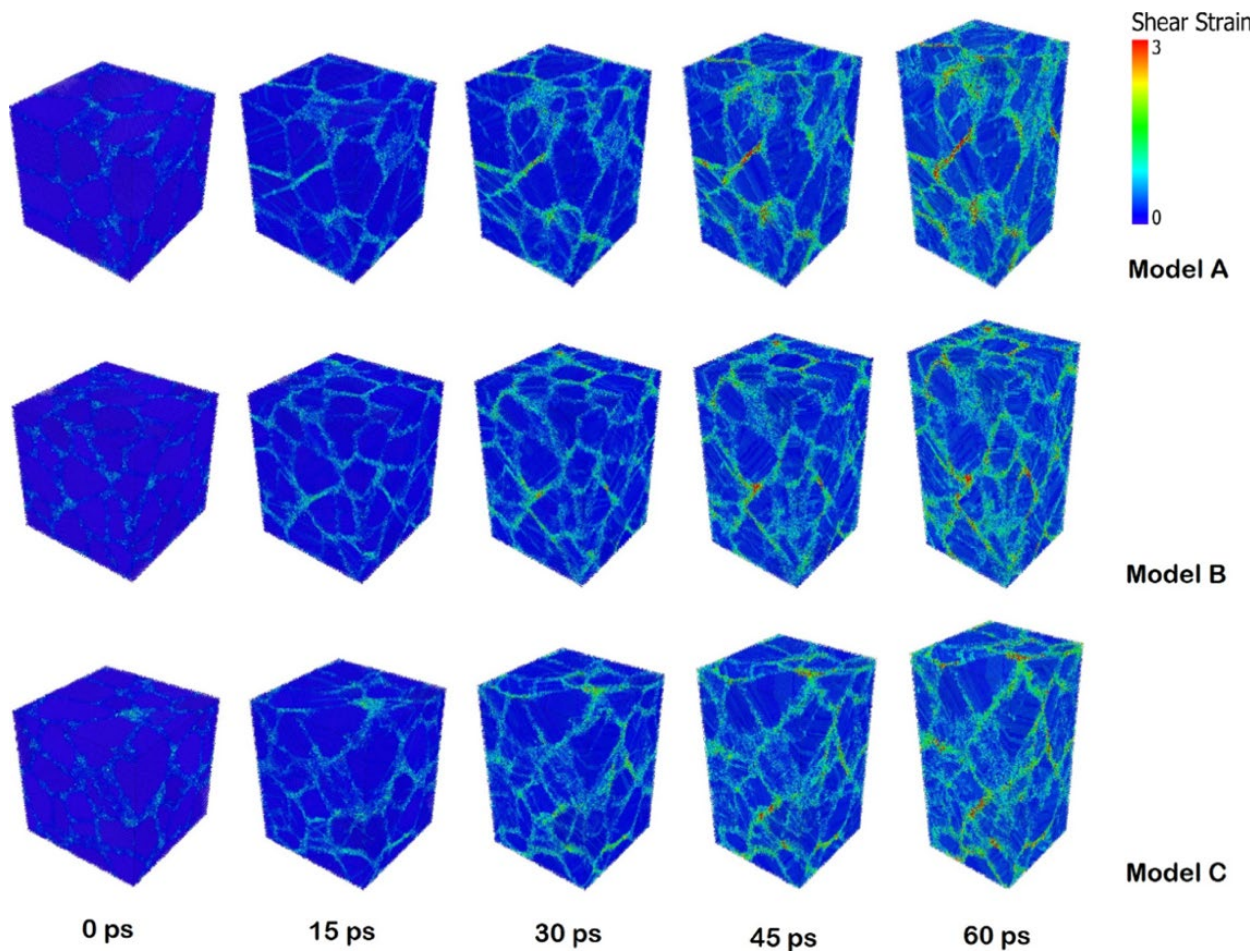


Fig. 2 Microstructure evolution of Fe-Ni-Cr-Co-Cu high entropy alloy during tensile test with different models for different simulation time colormap by atomic strain

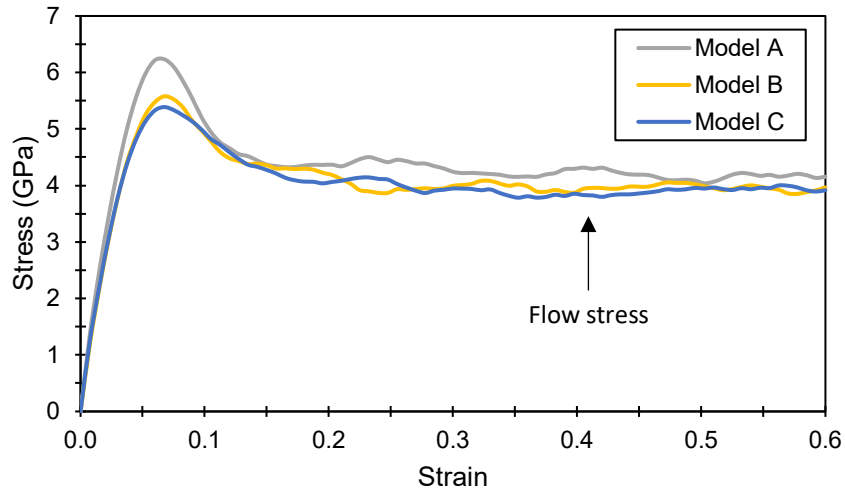


Fig. 3 Stress-strain curve of tensile behavior of Fe-Ni-Cr-Co-Cu high entropy alloy with a different model

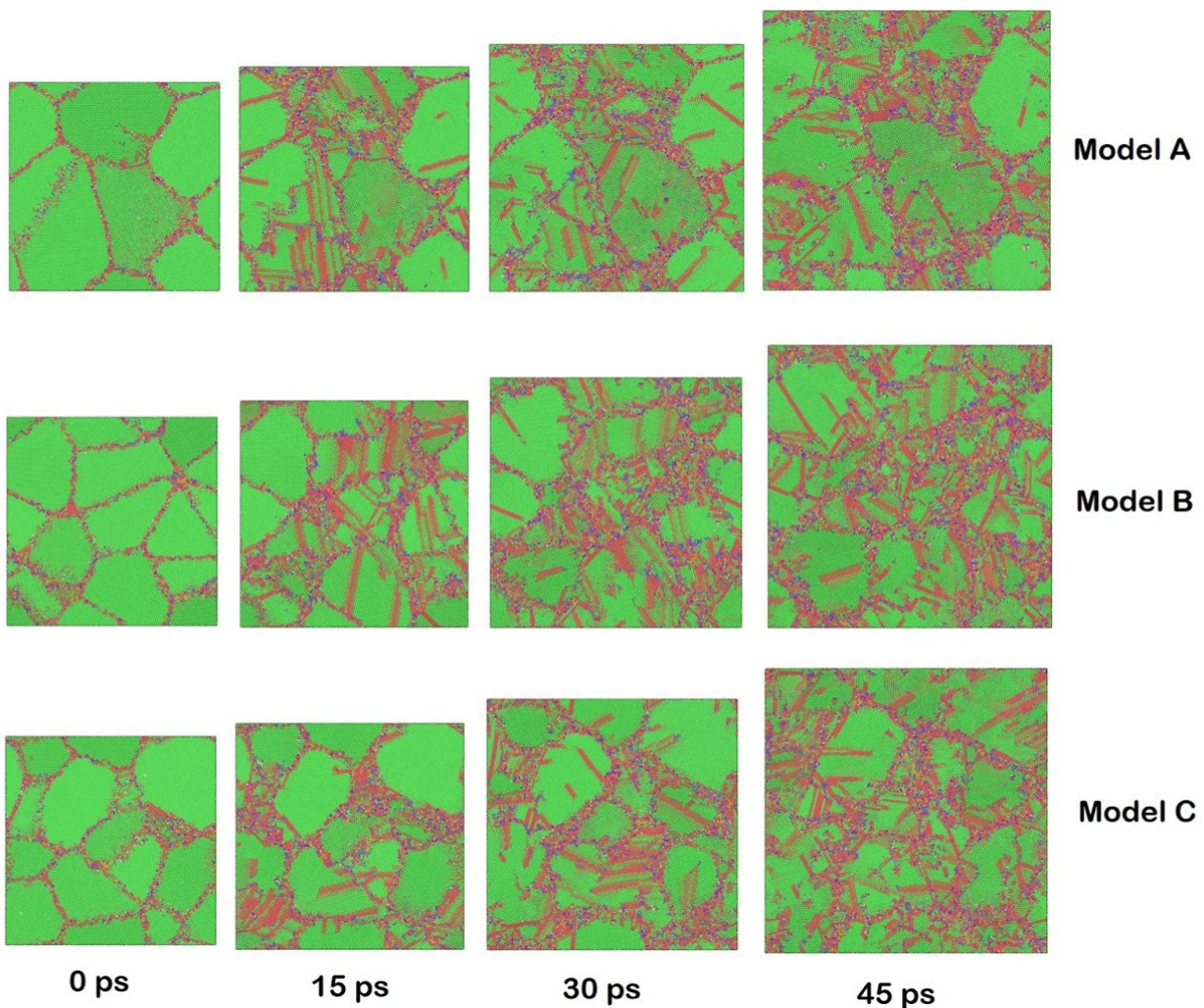


Fig. 4 Microstructure evolution compressive behavior of Fe-Ni-Cr-Co-Cu high entropy alloy with different models and simulation times analyzed with polyhedral template matching, where the green-colored atoms show the atoms with FCC structure and the red-brown atoms show the atoms with arbitrary structure

In Fig. 4, where the polyhedral template matching is utilized, the significant value of strain and displacement is hardly characterized since it only analyzes whether a group of atoms is matched to a specific crystal structure. However, in Fig. 5, when the atomic strain analysis method is utilized to analyze the system, the structural evolution of the system is slowly deformed arbitrarily. For all the models, from 0 ps to 30 ps of a compressive test

with a strain rate of $-1.0 \times 10^{10}/s$ at 300 K, the deformation is characterized mainly in the grain boundary—and thus the primary deformation mechanism occurs in the grain boundary, which is grain boundary sliding. Moreover, the deformation is evenly distributed when compressed within the grain boundary. However, finding a completely new cluster creation of a grain promoted by a larger compressed grain is hard. As mentioned before, this phenomenon could then be used to produce a new smaller grain size on average and promote a finer grain as in the severe plastic deformation method. The stress-strain curve of the compressive test, as depicted in Fig. 6, shows that the compressive strength is decreased as in the tensile test, with a similar pattern; that is, the strength different from Model A and B is significantly higher compared to the strength different from Model B and C. The compressive strength of the three models is 7.75 GPa, 6.75 GPa, and 6.28 GPa, respectively, for Models A, B, and C.

Furthermore, both tensile and compressive test results show that the flow stress between Model B and C shows a similar pattern, where it tends to have an average similar value, while Model A shows a higher value. This behavior indicates that the model with a bigger grain size on average, where the fraction of grain boundary volume is lower, has a higher strength than those with a smaller grain size. It should be noted that this phenomenon is only applied when the grain size is so tiny below the Hall-Petch threshold, which in all materials has a different limit. One may assume that if the grain size is smaller than that in Model C, the decrease in both tensile and compressive strength may not be significant. However, the study confirmed the inverse Hall-Petch nature of the Fe-Ni-Cr-Co-Cu HEA from both tensile and compressive tests, with a considerable decrease in the tensile and compressive strength values.

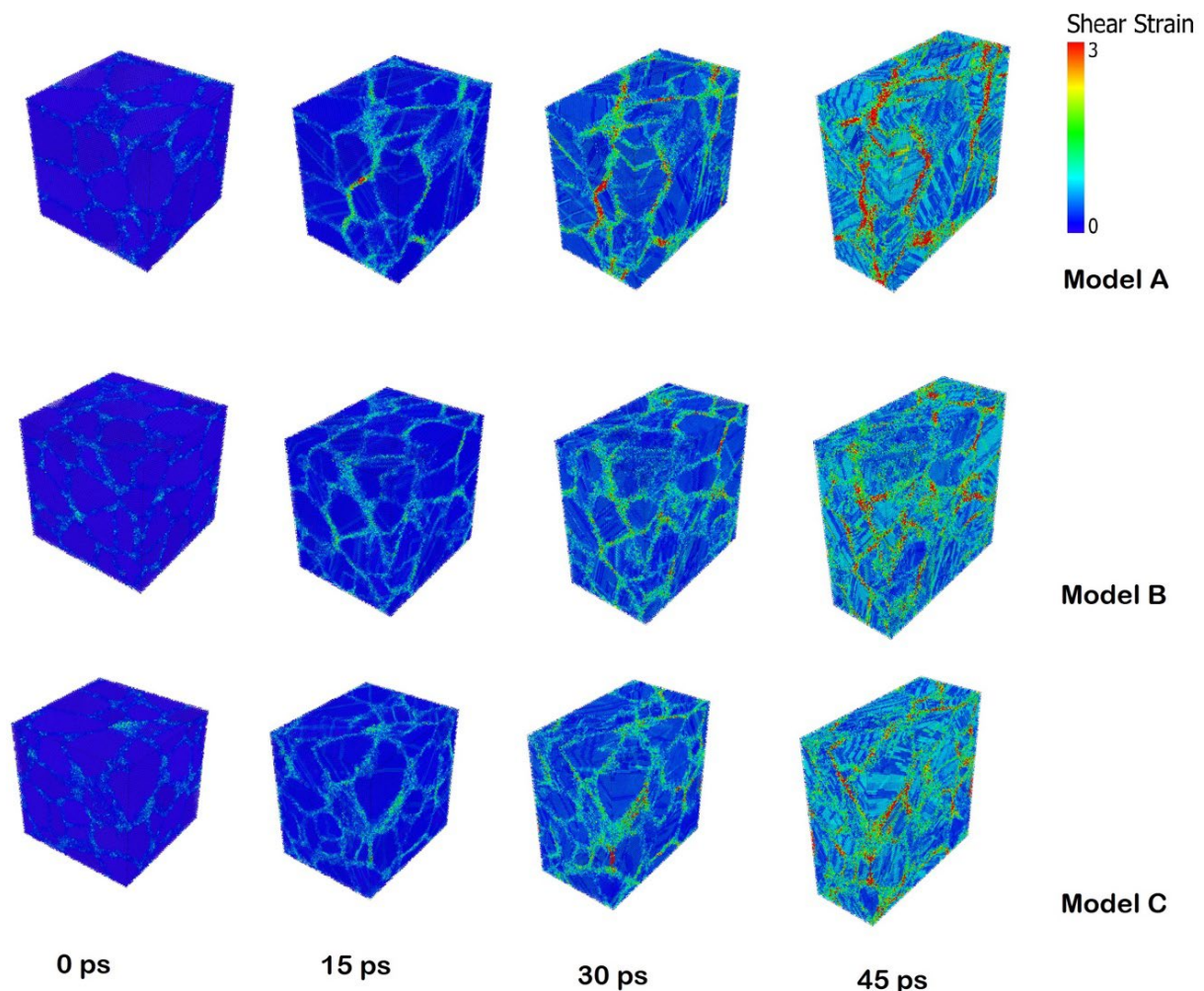


Fig. 5 Microstructure evolution compressive behavior of Fe-Ni-Cr-Co-Cu high entropy alloy with different models and simulation times analyzed with atomic strain

4. Conclusions

The tensile and compressive behavior of nanocrystalline Fe-Ni-Cr-Co-Cu high entropy alloy has been studied, and the results have been discussed in detail. It is concluded that the nanocrystalline Fe-Ni-Cr-Co-Cu HEA exhibits an inverse Hall-Petch effect, where the decrease in strength is affected by the grain size reduction. The deformation mechanism is dominantly characterized by the grain boundary sliding, manifested by the higher fraction of grain

boundary volume within the system. The deformation within the grain is only identified when it is subjected to a compressive test after 45 ps, where this also exhibits the effectiveness of using the severe plastic deformation method in producing a finer grain structure, i.e., smaller grain size. Both tensile and compressive behavior of nanocrystalline with the grain size of 7.3 nm^3 , 6.8 nm^3 , and 6.4 nm^3 for Model A, B, and C, respectively, confirming the inverse Hall-Petch nature of Fe-Ni-Cr-Co-Cu high entropy alloy.

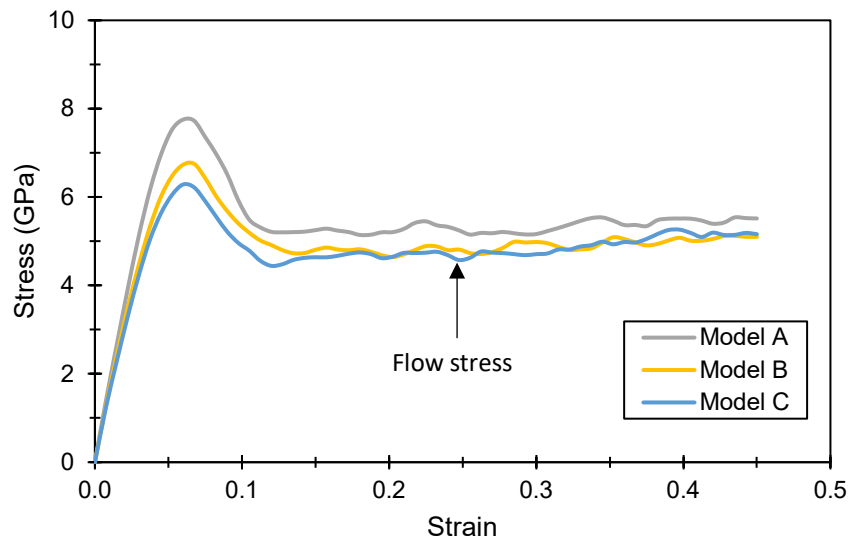


Fig. 6 Stress-strain curve of compressive behavior of Fe-Ni-Cr-Co-Cu high entropy alloy with different models

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Conflict of Interest

The authors state that there are no conflicts of interest related to the publication of this paper.

Author Contribution

The authors assumes full responsibility for the study conception, research design, data collection, data analysis, result interpretation and manuscript drafting.

Data Availability

The data supporting this study's findings are available from the corresponding author upon request.

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