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Molecular Dynamics Simulation on Creep Mechanism of Nanocrystalline Cu-Ni Alloy

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ABSTRACT– Creep mechanism is an essential mechanism for material when subjected to a high temperature and high pressure. It shows material ability during an extreme application to maintain its structure and properties, especially high pressure and temperature. This test is already done experimentally in many materials such as metallic alloys, various stainless steel, and composites. However, understanding the creep mechanism at the atomic level is challenging due to the limitation of the instruments. Still, the improvement of mechanical properties is expected can be done in such a group. In this work, the creep mechanism of the nanocrystalline Cu-Ni alloy is demonstrated in terms of molecular dynamics simulation. The result shows a significant impact on both temperature and pressure. The deformation supports the mechanisms as a result of the grain boundary diffusion. Quantitative analysis shows a more substantial difference in creep-rate at a higher temperature and pressure parameters. This study has successfully demonstrated the mechanism of creep at the atomic scale and may be used for improving the mechanical properties of the material.

KEYWORDS: *Cu-Ni alloy; creep behavior, molecular dynamics simulation.*

INTRODUCTION

A single system of most metallic materials such as Au, Ag, Cu, Al, and Ni is suffering from their ability to accommodate the complex demand of industry ranging from manufacturing, electronics, and transportation. Such a system cannot be used to satisfy every aspect of the orders that come from the mentioned industries. It is due to the economic efficiency issue in general. It could be because it does not have all the electronic, magnetic, catalytic, biological, and mechanical properties necessary for a specific application. Furthermore, binary systems often provide a unique solution due to their unusual properties and phase transition compared to a single system (Ahmed et al., 2008; Baskaran, Sankara Narayanan, & Stephen, 2006; Brillo & Egry, 2005; Gubicza et al., 2018; Mizushima, Chikazawa, & Watanabe, 1996; Sopousek et al., 2014; Studt et al., 2012; Teeriniemi, Taskinen, & Laasonen, 2015).

Cu-Ni is one of the binary systems that nowadays is widely used in various ranges of application, since it exhibits extraordinary properties compare to singular system. One of its applications is metal foams since it has excellent bending strength and superior anti-corrosion (Gubicza et al., 2018; Ji, Zhao, Zhang, Chen, & Dai, 2014; Mizushima et al., 1996). Another application is for a catalyst of CO's hydrogenation to methanol (Studt et al., 2012) and other nanoparticles (Ahmed et al., 2008). Cu-Ni could also be used for thin films (Cao, Short, & Yip, 2017; Mizushima et al., 1996), and even more recently has been used for inducing the transformation of chemical vapor

deposition (CVD) grown bilayer graphene into a fluorinated single-layer diamond (Bakharev et al., 2019). Such a wide range of applications needs a well-understanding of their physical, mechanical, and chemical properties; thus, the utilization of this material could be derived appropriately.

Creep is a plastic deformation of materials due to a couple of high temperatures and high pressure. High creeping phenomena restrict the application of nanocrystalline (NC) metals at medium and high temperatures. Understanding creeps in NC metals is essential for enhancing a thermal stability of the material and service life. Therefore, understanding creep in NC metals is crucial to improve thermal stability and the service lifetime of a material. It has been reported that several fundamental processes significantly influence creep deformation of NC metals: grain boundary (GB) diffusion, GB sliding and migration, and dislocation nucleation from GB with increasing applied stress (Wang, Ishii, & Ogata, 2011, 2012; Wolf, Yamakov, Phillpot, Mukherjee, & Gleiter, 2005). These thermally activated deformation mechanisms lead to a temperature dependence of strength in NC metals.

Several studies have been reported in terms of creep mechanism by employing molecular dynamics (MD) simulation. One of the first studies that have used MD simulation to study creep is a study conducted by Keblinski et al. (Keblinski, Wolf, & Gleiter, 1998). Several phenomenological behaviors correspond to the Arrhenius's diffusion equation and Coble formula related to grain boundary activated diffusion energy is well-identified by this study. However, several idealizations correspond to the grain boundary along with polycrystalline silicon has to be adopted.

A deeper understanding of grain growth on the grain boundary of Palladium diffusion-creep is contributed to Haslam et al. (Haslam et al., 2004). Grain boundary (GB) diffusion creep, indicated by the Coble-creep mechanism, is accommodated by geometrically necessary GB sliding, leading to several deformation types that they called Region 1 and Region 2. The other two studies have revealed the dependence of grain size (Wang et al., 2012) and time, stress, and temperature (Yang et al., 2016) during creep deformation. To understand the entropic effect during steady-state creep deformation, Wang et al. (Wang, Ishii, & Ogata, 2013) have performed MD simulation of nanocrystalline Cu. They have demonstrated that the activation entropy could contribute multiplicative factors as high as 10^3 and 10^9 for creep governed by GB dislocation nucleation and for GB diffusion. Investigation on the influence of adding a specific element to a binary alloy has been conducted by (Pal, Meraj, & Deng, 2017). They conclude that the addition of Zr on nanocrystalline Ni accelerates the amorphization during creep deformation and that nanocrystalline Ni with segregated Zr presents a longer time during creep process compared to nanocrystalline Ni with randomly distributed Zr atoms. Recently, Sun et al. (Sun, Liu, He, Xie, & Peng, 2019) has demonstrated the shift mechanism of creeping behavior in nanocrystalline Ni-Al alloy which results the effect of temperature, stress, and grain size on the creep rate, especially in tertiary creep stage. However, some phenomena of creep diffusion for particular material like Cu-Ni alloy have not clearly identified. Therefore, this study aims to tackle this issue.

SIMULATION PROCEDURE

Three software are used in this study for running the two steps of the whole study. For the simulation itself, the LAMMPS (Plimpton, 1995) software package is used. This software calculates and solves classical molecular dynamics simulation, Newtonian equation of motion, with an efficient algorithm in which a statistical ensemble of atoms is embedded by this software, thus making it such a complete solution for a classical molecular dynamic simulation, especially for metallic materials. LAMMPS is claimed as the most used classical MD software to study materials behavior at the atomic level, due to its capability to make use of large-scale simulation efficiently. Another

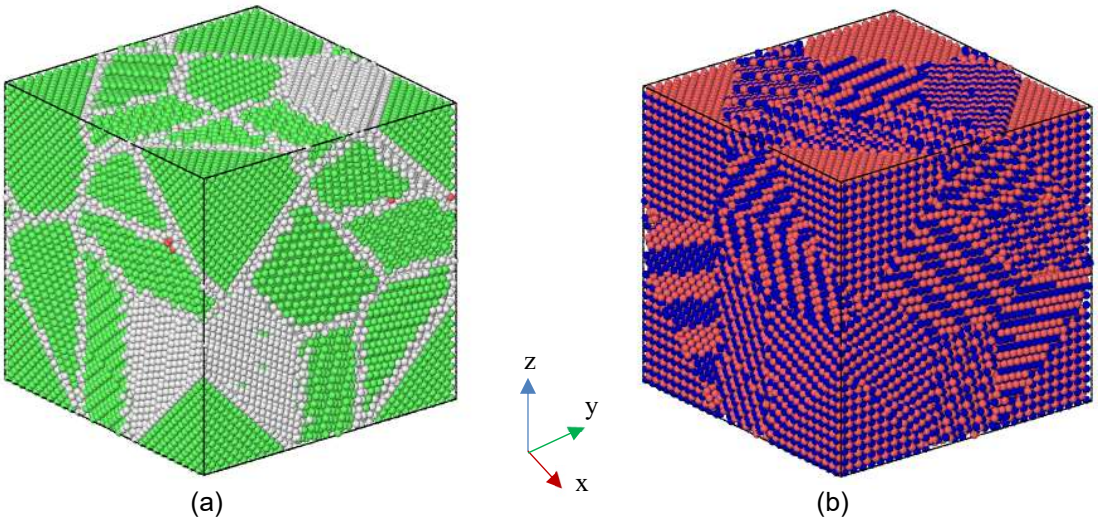

Figure 1 Simulation model of Cu-Ni alloy with 10 nm × 10 nm × 10 nm: (a) a view assisted by Common Neighbor Analysis (CNA) for separating view between each grain and (b) atom-type view with Cu (red) and Ni (blue) atoms.

step is the post-processing of the obtained data after running the simulation. In this study, the simulation data is processed using OVITO (Stukowski, 2010) software to view the atomic movement, thus showing the bulk materials transformation due to constant stress and temperature during a creep test. In this software, several structural analysis techniques are also included and improve the ease of identifying structural defects and deformation of metallic crystalline materials.

The simulation model as depicted in Figure 1 consists of Cu-Ni alloy with roughly the same atom number for each element (50% of Cu and 50 % of Ni). The simulation model's size is 10 nm × 10 nm × 10 nm in the *x*, *y*, and *z*-direction, respectively. The grain distribution and modeling are generated following the Voronoi tessellation using AtomSK software (Hirel, 2015). All of the bulk model volumes are divided into ten similar size grains with random grain orientation. The model is not a perfect cube at the first model, but an arbitrary shape corresponds to its grain size and orientation. However, AtomSK software facilitates wrapping the model in such a way to meet the perfect cube form. The software facilitates wrapping the model so that it is then covered into a perfect cube. A periodic boundary condition is applied in all directions to allow atoms to move freely due to induced force. If the atoms exceed the induced force limit during the creep test, it will occur in the opposite direction with the same properties. Thus, it will maintain the number of atoms in the system. It will also reduce the effect limit which is not normally involved in experimental work because the limit has to be defined for such simulations. Two parameters were used in this study: temperature and pressure, with three levels each. The details of the simulation parameters follow the sample number given in Table 1.

Table 1 Simulation parameters

Sample No.	Temp (K)	Press (GPa)
S1	1000	1
S2	1100	1
S3	1200	1
S4	1000	1.25
S5	1000	1.5
S6	1100	1.25
S7	1100	1.5
S8	1200	1.25
S9	1200	1.5

Typically, classical MD simulation consists of three steps. They are initialization, production, and relaxation. These three steps, though, could use the same statistical ensemble,

running different instances. In this study, first, the system is initialized by the NPT ensemble with room temperature and atmospheric pressure. This step is performed only for a short time to obtain an equilibrium condition; thus, the system is ready for the next production step. After the system reaches an equilibrium state, constant pressure and temperature, as listed in Table 1, are applied. This stage is performed for 300 ps under the NPT ensemble. Data on the relationship between strain and time corresponding to the creep test are generated along with the simulation. Subsequently, the system is cooled until ended with the relaxed final shape of the bulk material after a creep test. During the initialization stage, it should be noted how long the simulation will take for the system to be balanced against the complexity of the system. The more complex and bigger system will affect how long the system will be equilibrated and ready for the creep test. It could be monitored in LAMMPS by the energy associated with the system, typically potential and kinetic energy. If the system is running for creep test while it is not equilibrated correctly, it may result in the inaccuracy of the output.

Although the reliability of MD simulation is now recognized in many fields of study, in practice, several different factors may determine the quality of the simulation result and eventually the reliability of the simulation. For metallic materials, an interatomic potential plays a crucial role in determining the quality of the simulation, since it defines the properties of every atom inside the system. Embedded Atom Method (EAM) (Daw & Baskes, 1984; Foiles, Baskes, & Daw, 1986) is one of the interatomic potentials that is widely used for metallic materials. In this study, the EAM interatomic potential developed by Onat *et al.* (2014) for Cu-Ni alloy is adopted. Following the EAM formalism, the total energy is defined by:

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

Here $\Phi_{a_i-a_j}$ is a pair of interaction potential as a function of the distance r_{ij} between atoms i and j that have chemical sorts a_i and a_j (= A or B) and F_{a_i} is the embedding energy of an atom of a chemical sort a_i as a function of the host electron density $\bar{\rho}_i$ induced at site i by all other atoms in the system.

RESULTS AND DISCUSSIONS

Several aspects correspond to the creep test performance and its detailed features are described in this section, including time evolution of the sample during creep test, creep test results defined by the time-strain curve, and its creep rate. Also, for the sake of completeness, the relationship between the applied pressure and creep rate is finally discussed. It is worth mentioning that, although classical MD simulation have a scaling problem compared to the real experiment, in some cases, the evolution of a metallic material at atomic level due to applied parameter could be revealed in this study.

a. Time evolution during creep-test

Due to constant high temperature and pressure, the bulk materials experience compressive stress and then deformation may occur in which the magnitude of deformation depends on the applied parameters and the properties of the material itself. As depicted in Figure 2, a higher level of temperature and pressure that correspond to sample S3 experiences the highest deformation magnitude compared to S1 and S3. A perfect crystalline structure and grain boundary at $0\,ps$ have changed over time, showing evolution of the system due to applied parameter. The green-colored atom which indicates an FCC crystalline structure, has deformed randomly and made the structure have more white-colored atoms, which have a random structure. Diffusion at around the grain boundary, which is initiated immediately after the parameters are applied, is one of the responsible effects, besides the bulk material's deformation, that changed every grain's crystalline structure and subsequently creep phenomenon.

As listed in Table 1, several simulations have been performed. The relationship between time and strain, representing the ability of the materials to remain their crystalline structure under certain extreme temperature and pressure, has been obtained.

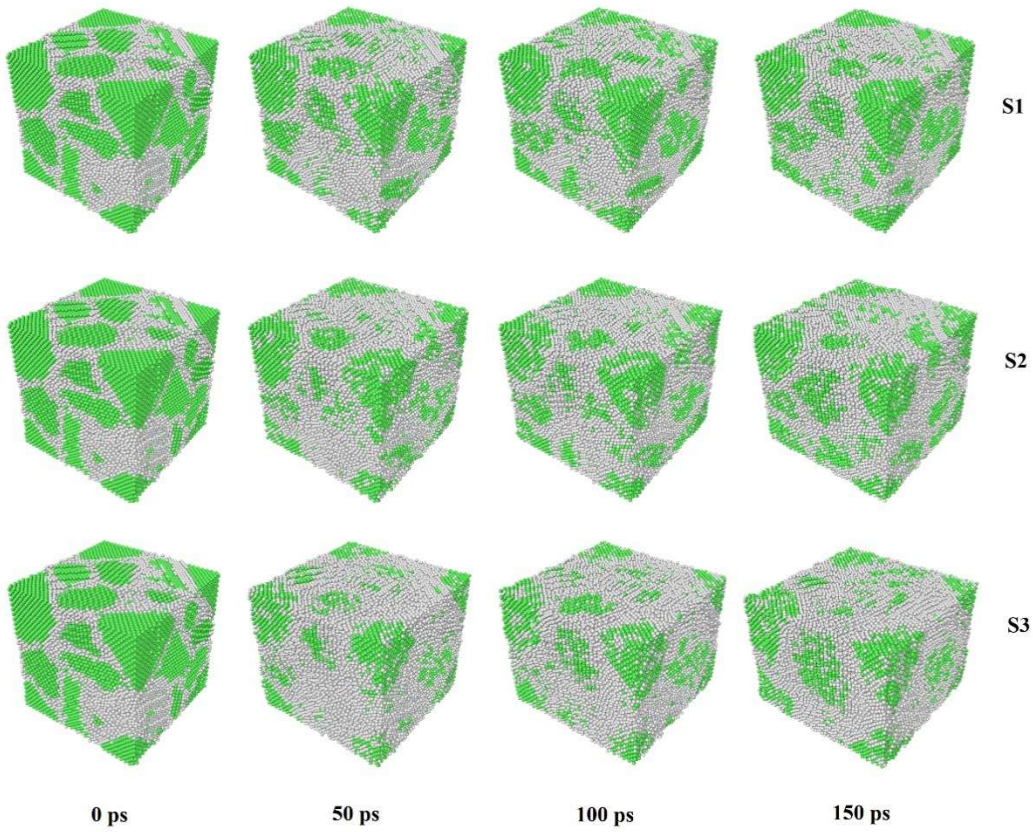

Figure 2 Time evolution of creep test for the samples with a pressure of 1 GPa at various temperature of 1000 K, 1100 K, and 1200 K, referred to as S1, S2, and S3, respectively.

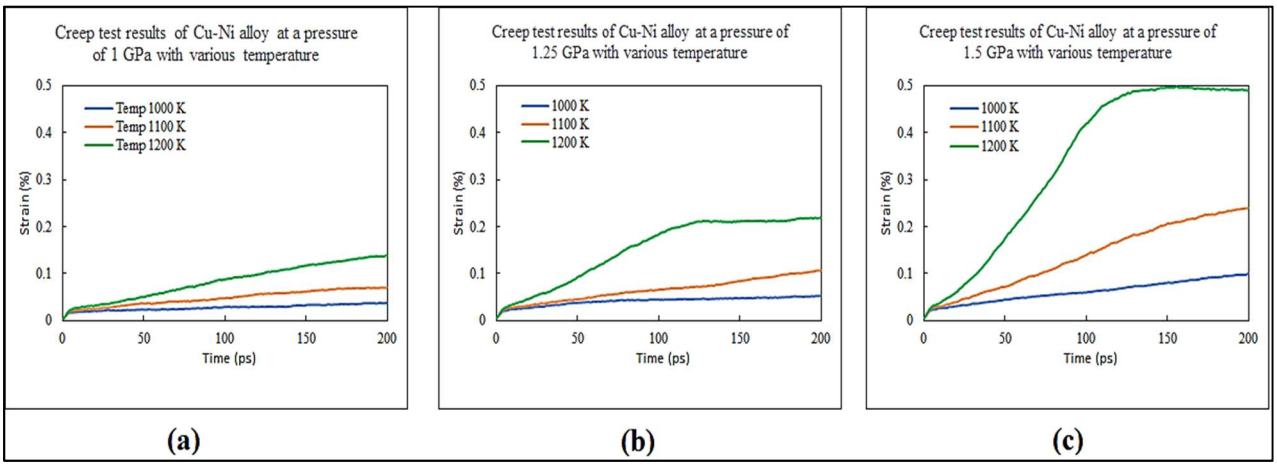

Figure 3 Creep test results of Cu-Ni alloy at the various temperature with the pressure of (a) 1 GPa, (b) 1.25 GPa, and (c) 1.5 GPa.

This time evolution which connected to the strain value of the materials over time, time-strain curve, is depicted in Figure 2 and Figure 3. Specifically, the comparison of temperature on the time-strain curve is illustrated in Figure 2, while Figure 4 shows the pressure effect.

Both Figure 3 and Figure 4 show an increasing effect, indicated by the significant difference between Figure 3(a), 3(b), and 3(c), and Figure 4(a), 4(b), and 4(c). Figures 3(b) and 3(c) indicate a plateau stage where no further strain changes. However, these plateau stages do not affect the diffusion around the boundary, as depicted in Figure 2. The diffusion around the boundary could still be seen, i.e., the primary creep stage due to constant force.

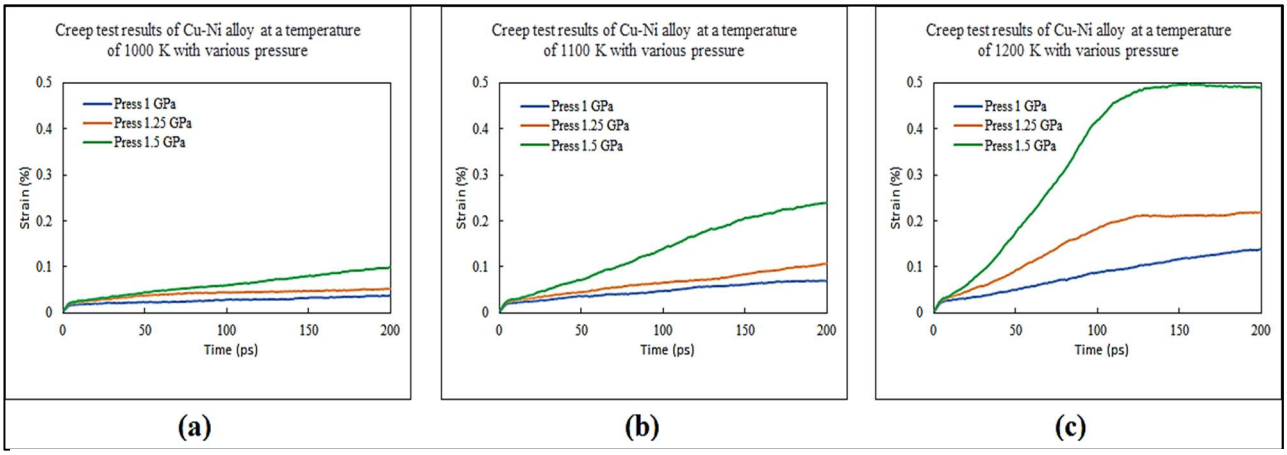

Figure 4 Creep test results of Cu-Ni alloy at the various pressure with the temperature of (a) 1000 K, (b) 1100 K, and (c) 1200 K.

b. Quantitative creep test analysis

There is also a more accurate way to determine the dominant mechanisms of creep deformation between the applied temperature quantitatively, comparing the creep-rate between the sample. It is done by calculating the slope of the tangent in the creep test curve. Table 2 shows the creep-rate of the corresponding sample number. It is shown that the highest value of both pressure and temperature during the creep test showing the significant difference, and hence defining the significant mechanism during the creep test. Although several samples show a bit different (S5 & S6) behavior, it is not changing the creeps behavior at a higher rate.

Another way to quantitatively evaluate the creep behavior is comparing them in terms of the curve of creep rate vs. pressure and then finding out the trendline in which some regression method could be used. Figure 5 shows the pressure vs. creep rate curve combining with its trendline. A more significant slope of tangent can be seen directly in this curve. Although the influence of both temperature and pressure is already discussed, and as other comparison techniques have been presented then the more its behaviour occur since the number of samples of each parameter is limited (only 3 x 3) then it is possible to find out the other behaviour bellow and upper from the presented parameter here at this paper.

Table 2 Creep rate of corresponding sample number.

Sample No.	Creep Rate (S-1)
S1	6.85×10^5
S2	3.01×10^6
S3	6.08×10^6
S4	3.41×10^6
S5	4.7×10^6
S6	4.33×10^6
S7	1.16×10^7
S8	1.58×10^7
S9	3.72×10^7

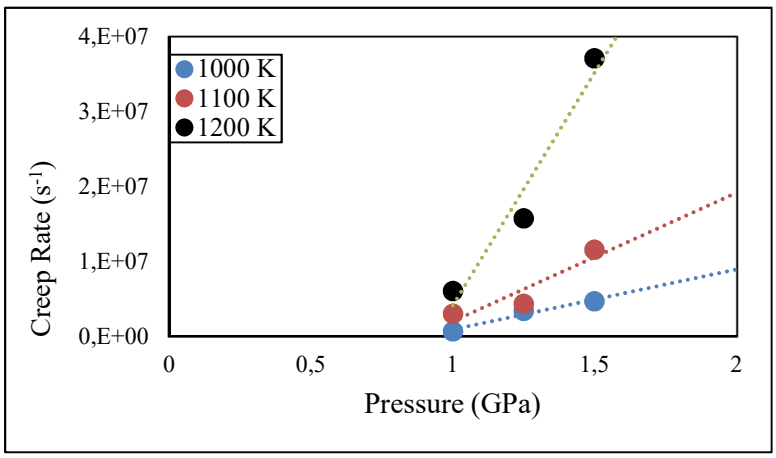

Figure 5 Pressure vs. creep-rate and its corresponding trendline for various temperature tests.

CONCLUSIONS

The conclusion can be drawn as Temperature and pressure have a significant impact on the creep-mechanism; The time evolution shows that dislocation of the atoms, especially those near the grain-boundary causing the mechanism of the creep; and Quantitative analysis shows the same behavior and that a higher level of temperature, pressure, and creep-rate makes the difference more significant.

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