Short Communication

A Study on the Role of Clay Nanoparticles and Temperature on the Creep Behavior of Polyethylene Matrix Nanocomposite

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Abstract

Objective: In the current research, the creep behavior of polyethylene (PE) matrix nanocomposites reinforced with different content of clay nano particles (i.e., 0, 1, 3, 5, 10wt%) at different temperatures (25 and 50°C) has been investigated.

Methods: In order to produce composite samples of pure PE reinforced with different percentages of clay (0, 1, 3, 5 and 10% by weight), a twin extruder was used in the temperature range of 200-230°C. Also, to produce the standard samples, an injection machine was used using in the temperature range of 220-230°C. The samples were subjected to a constant load, typically around 0.7 to 0.8 times the yield strength, at 25 and 50°C , and at constant strain rate of $0.1min^{-1}$.

Results: The creep investigations showed that for PE and its nanocomposites the creep trend could be divided into primary, secondary, and tertiary stages. The length of each stage depended strongly on both temperature and the stress level at which the samples tested as well as the clay content. It was observed that adding nano particles in small amounts (1wt%) led to an increase in the length of the creep life of PE and it also improved this property in both temperatures.

Conclusion: It was observed that increasing test temperature from causes to decrease the creep strength as well as a rapid onset of plastic deformation for all materials including PE and its nanocomposites. PE nanocomposites with higher than 1wt% clay exhibited a noticeable drop in creep strength particularly at 50°C.

Keywords: polyethylene, clay, nanocomposite, creep behavior

1 INTRODUCTION

Polyethylene (PE) is still widely used in various industries. Its excellent resistance to chemicals makes it suitable for storage tanks and pipes used in the chemical industry. Its high toughness and low temperature resistance also make it ideal for applications in the packaging industry, including bottles, containers, and films. But the low melting point of PE restricts its usage in some applications where high temperatures are involved, as it can easily deform and lose its shape. Additionally, the poor thermal properties of PE make it susceptible to heat degradation, further limiting its application in industries that require high thermal stability. To improve the mechanical and physical properties and for promotion of service temperature of PE, the addition of secondary particles such as talc, mica, and calcium carbonate and clay to it is suggested[1-8]. PE-clay nanocomposites are new type of materials that have gained recognition only in recent times. These nanocomposites consist of PE with added clay particles, which serve to enhance various properties of the material. In fact the addition of clay particles to PE can improve the toughness and strength. Furthermore, it can also enhance the thermal properties and conductivity. Additionally, the presence of clay can provide barrier properties that surpass them from the original polymer[8-12].

Creep behavior in polymers is a result of their unique molecular structure and flexibility. Unlike metals or ceramics, which have rigid atomic structures, polymers are made up of long, chain-like molecules that can easily slide past each other. Under the influence of a constant load or stress, these polymer chains or molecular segments can slowly move and rearrange themselves, leading to deformation over time. The temperature at which creep behavior occurs in polymers is often above their glass transition temperature. The glass transition temperature is the temperature at which the polymer segments do not have enough energy to move or vibrate. Above this temperature, the polymer chains have more molecular mobility and can move more freely, making them more prone to creep[13].

PE has been extensively studied in polymer science due to its structural simplicity and wide range of applications. The mechanical creep behavior of PE is of particular interest because of its increasing use in engineering applications, such as water and gas pipes. Therefore, it is crucial to examine the creep behavior of PE under a fixed stress to characterize its mechanical nature within the framework of viscoelasticity. Based on the literature survey done by the authors there are only a few studies concentrated on the creep behavior of polyolefin nanocomposite polymers. For example, Alghamdi[14] focused on the creep resistance of PE-based nanocomposites. The results showed that the addition of just 0.5wt% nanoclay caused a significant increase in the creep resistance of the PE blend. Conversely, the addition of Carbon Black or Carbon Nanotubes caused a reduction in the creep resistance. Wilhelm et al.[15] reported the development and application of nano creep test which allows for a direct control of the experimental parameters especially that of the applied stress, and combines it with an extraordinary high resolution of both the stress and the strain measurement. Laguna-Gutierrez et al.[16] analyzed the mechanical properties and effective diffusion coefficient under static creep loading of low-density foams based on PE / clays nanocomposites. They analyzed the behavior of the gas diffusion coefficient of the cellular materials when they are subjected to a static load. Drozdov et al.[17] derived a new constitutive equations in viscoelasticity and viscoplasticity of polypropylene / clay nanocomposites. Also Drozdov et al.[18] investigated the viscoelasticity of PE / montmorillonite nanocomposite melts. They presented an experimental investigation and constitutive modeling of the time-dependent response of hybrid nanocomposite melts with polymer matrices reinforced with organically-modified nanoclay.

This current study attempted to investigate the role of both clay content and temperature on the creep behavior of PE matrix nanocomposite. For this purpose, creep tests were performed at two stress level and two temperatures.

2 MATERIALS AND METHODS

2.1 Materials and Sample Preparation

The used PE in the current research was produced by Arak Petrochemical Company, Iran, and its specifications are summarized in Table 1. The specific clay utilized in the research is Montmorillonite DK4, manufactured by Nanoline Company in China. The detailed specifications of the clay are presented in Table 2.

In order to produce composite samples of pure PE reinforced with different percentages of clay (0, 1, 3, 5 and 10% by weight), a twin extruder was used in the temperature range of 200-230°C. Additionally, standard samples were produced by using an injection machine in the temperature range of 220-230°C.

2.2 Creep Test

To assess the durability of the PE and its nanocomposite under constant load, creep tests were commonly employed. This information provided valuable insights for engineers regarding the lifespan and performance of the component under the given conditions. In this study, the tested samples were subjected to two constant loads (0.7 and 0.8 times the yield strength of the sample), at 25 and 50°C at constant strain rate of 0.1min⁻¹. The creep strength of the sample was calculated based on the time it took to reach the neck. The EMEC 2088 series creep

Table 1. Specifications of Used Polyethylene

Specification	Unit	Standard ASTM	Value
Melt Flow Index at 230℃ and 2.16Kg	g/10min	D1238/L	7
Softening Temperature, 10N	C°	D1525	152
Heat Deflection Temperature 0.46N/mm²	C°	D648	88
Density	g/cm³	-	0.97

Table 2. Specifications of Used Clay

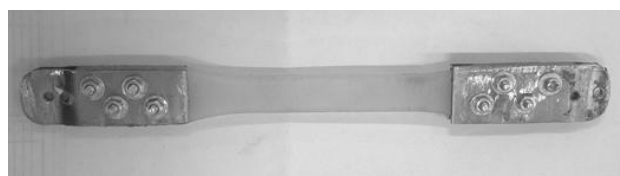| Specification	Unit	Value
Montmorilionite	%	96
Crystalline silicon	%	1
Humidity	%	3
Density	g/cm³	1.7

Figure 2. The prepared sample for creep test.

workpiece. Figure 2 depicted the prepared sample for testing.

3 RESULTS AND DISCUSSION

Figure 3A-3E showed the comparison of the creep behavior of the pure PE and the nanocomposite samples, under a load of 0.7 yield point in temperature conditions of 25℃ and 50℃. As seen in the figures, the creep deformation in PE and its nanocomposites could be divided into three stages, the so called primary, secondary, and tertiary stages. In fact, during primary creep, the rate of strain accumulation is highest at the beginning and gradually decreases over time as the material undergoes molecular rearrangement, until it reached a steady-state or equilibrium. This stage was often referred to as the transient or initial creep. Secondary creep was the stage where the strain rate became relatively constant, relatively low, and remained constant over an extended period of time. This stage was often referred to the steady-state or uniform creep. Tertiary creep was the final stage characterized by an accelerating creep rate. This stage was often associated with the onset of significant damage accumulation and leads to eventual failure of the material.

Based on the conducted creep tests and the comparison of material behavior at 25 and 50℃, it was observed that there was a significant decrease in creep strength and a rapid onset of plastic deformation due to the increase in temperature for all materials including PE and its nanocomposites. The sensitivity to the increase in temperature and the reduction in creep resistance showed a direct relationship with the content of clay. Particularly, percentages above 1% exhibited a noticeable drop in creep strength under high temperature conditions. The creep rate, represented by the slope of the creep curve in the stable region ($d\varepsilon/dt$), played an important role in understanding the deformation behavior of materials under creep conditions. Initially, the creep rate increased rapidly as the specimen lengthens. However, over time, the creep rate decreased
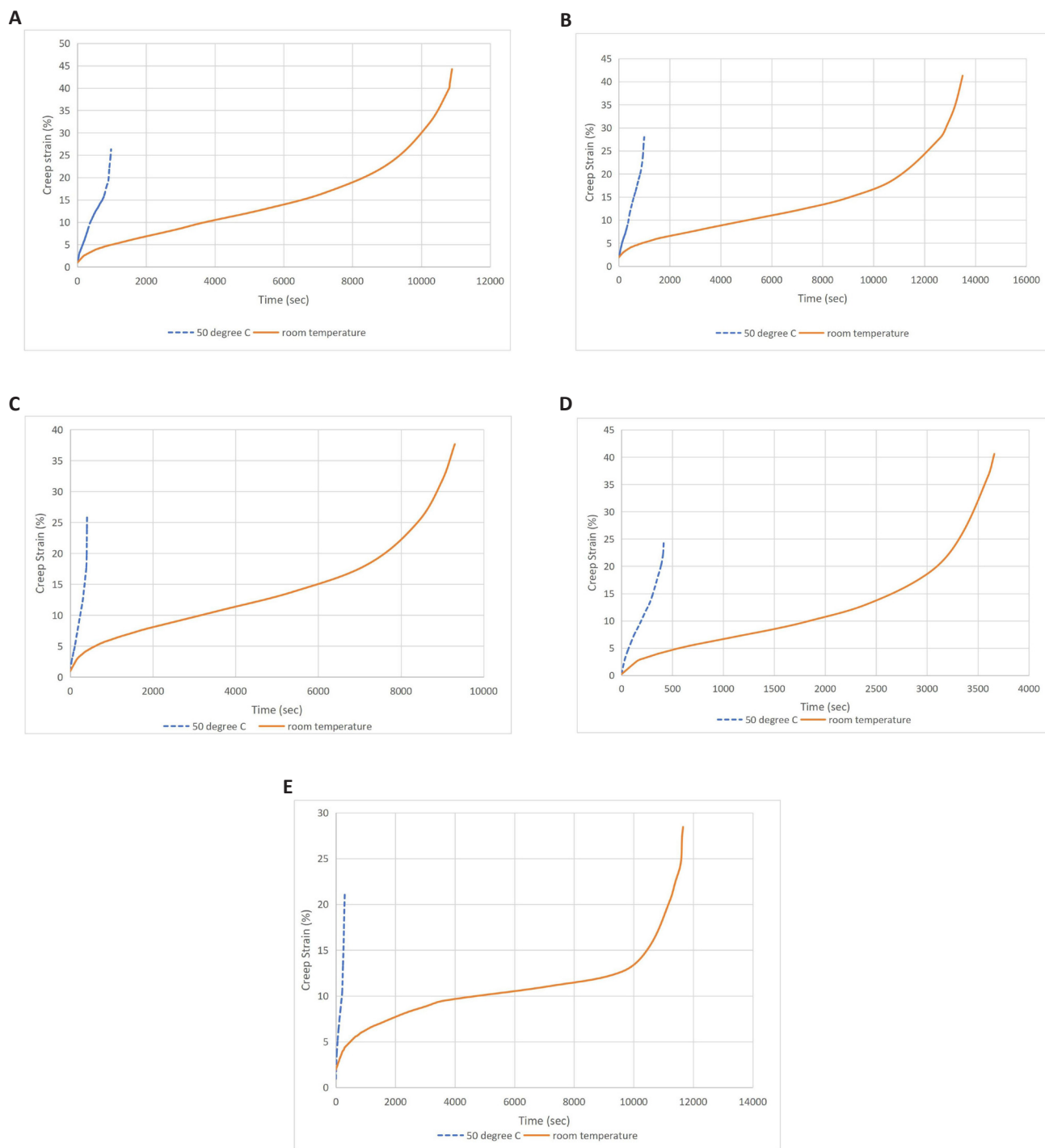

Figure 1. The used apparatus for creep test.

device, belonging to Shiraz University and manufactured in England, was utilized for the test. The temperature conditions were achieved using an electric resistance heat treatment furnace. The heat treatment furnace consisted of a cylindrical holding chamber made of plain carbon steel, which housed the sample and isolated it from the surrounding environment temperature fluctuations. To ensure uniform heat distribution inside the chamber, glass wool insulation was applied around the chamber openings. Heat was supplied by a thermal element connected to the local electricity source, utilizing electrical resistance to generate heat within the element. The temperature was monitored using a thermometer placed inside the chamber. Figure 1 displayed an image of the used creep device.

The used samples for the creep test were equipped with a 1mm thick steel sheet mold and secured with 8 bolts and nuts, four bolts were placed in the upper part and four in the lower part of the sample. The mold served the purpose of holding the sample inside the device and transferring the load from the metal-polymer interface to the central areas of the workpiece. This was done to prevent premature failure of the sample caused by stress concentration in the punched areas of the

Figure 3. Comparison of creep behavior under 0.7 yield load at different temperatures. A: Pure PE; B: PE / 1% clay; C: PE / 3% clay; D: PE / 5% clay; E: PE / 10% clay.

and eventually reached a steady state. Eventually, as failure approached, the creep rate increased rapidly. To further analyze and compare the creep behavior of the PE and its nanocomposites, Figure 4A-4C provided more detailed perspective at different loading and temperatures. As it could be seen in the curves the creep behavior of PE was influenced by temperature, stress level and the presence of clay. Higher temperature generally accelerated the creep rate, as the increase in thermal energy facilitates molecular motion and rearrangement. In fact, increasing temperature generally increased the free volume of polymers, making

PE chains move easily from one point to another. The reason of this effect could be attributed the fact that increasing temperature break weak bounds between chains, enabling them past each other even at a low level stress. Clearly, this behavior became much significant as temperature increased, particularly at temperatures far from glass transition temperature[19,20]. The stress level (0.7 and 0.8 yield strength) applied to the polymer also played a critical role in creep behavior. Higher stress levels accelerated creep deformation, leading to faster degradation and failure of PE. Actually, there were two types of bounds in PE the
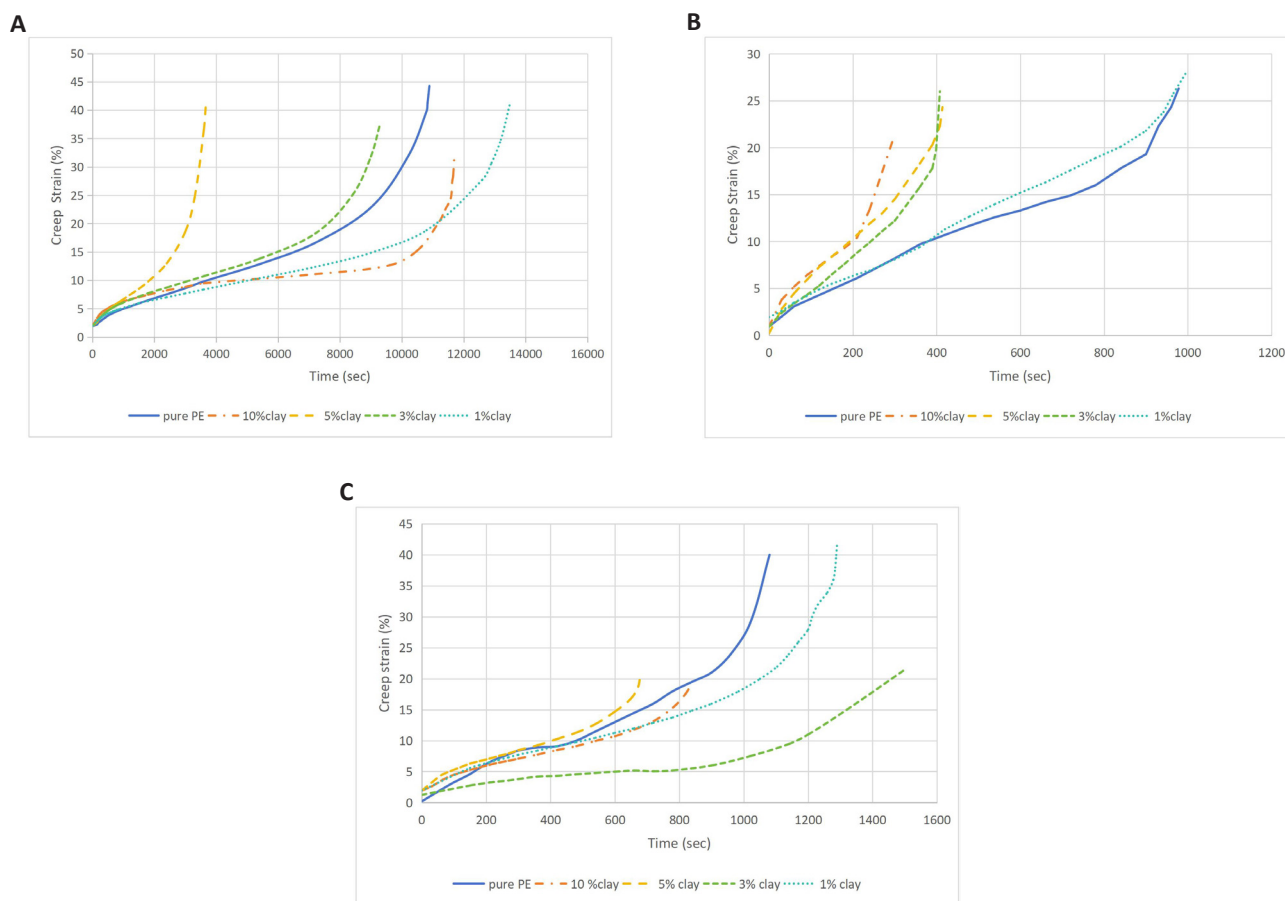
Figure 4. Comparison of the creep behavior of polyethylene and its nanocomposites. A: At 25℃ and the load of 0.7 yield strength; B: At 50℃ and the load of 0.7 yield strength; C: At 25℃ and the load of 0.8 yield strength.

so called primary and secondary bounds. The very strong primary bounds formed between the ethylene monomers, and stress lower than tensile strength couldnot break them. The secondary bounds which formed between the PE chains were very weak, and broke due to the applied stress even less than tensile strength. It was clear that at constant temperature, as the level stress increased, the number of secondary broken bounds increasesd. In such case polymer chains past each other and the creep rate increased. The addition of clay improved the creep resistance of PE. These fillers could hinder molecular motion and provide additional structural support, reducing the propensity for creep deformation. The exact reason of variation in Figure 4B was the synergistic effect of temperature and clay content. On the one hand, creep rate increased because of increasing temperature, on the other hand, the free volume of the polymer decreased because of the presence of clay[21,22].

Upon observing and analyzing the comparative behavior of pure PE and the nanocomposite samples, it was evident that the PE / 1wt% clay sample exhibited the highest creep strength in various workpiece conditions. This specific nanocomposite sample demonstrates a longer time before reaching the threshold of non-uniform plastic deformation, or in other words, the onset of necking in the sample than the other samples. This implied a higher

resistance of the material against creep deformation compared to the rest of the samples. Figure 5A-5E provided schematic representation of this observation. It was clear that one of the most important role of clay in PE matrix was to tolerate load and in theory increasing loading content caused to tolerate the stress at higher level. But the subject should be taking account was the negative effect of agglomeration and high temperature on creep strength. Indeed, role of temperature and clay content was opposite on creep rate. If particles were not agglomerated, they could compensate the negative effect of increasing temperature (as it happened in sample reinforced with 1%). But for the sample reinforced with 3wt% clay, higher temperature caused a weaken to the sample because of agglomeration of particles. This was why the creep strength of the PE reinforced with 3% clay was almost better than the other samples at 0.8 yield strength just at 25℃.

4 CONCLUSION

In the current research, creep test at different stress levels and temperatures performed on the PE and its nanocomposites. The results were remarked as follows:

● The creep trends in PE and its nanocomposites could be divided into primary, secondary, and tertiary stages.

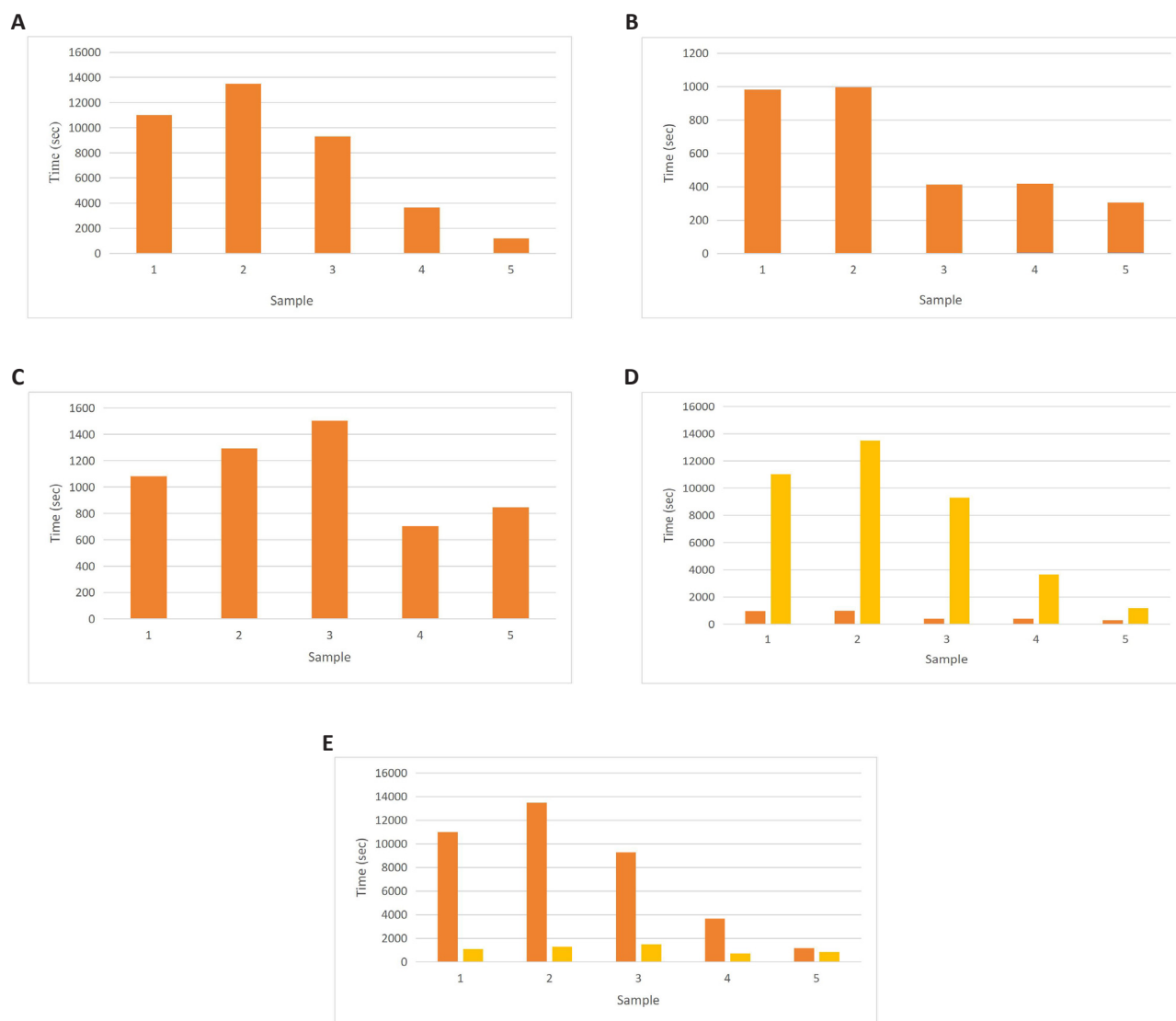● It was observed that an increase in temperature leads

Figure 5. Comparison of the time to reach the necking of the samples. A: At 25℃ and the load of 0.7 yield strength; B: At 50℃ and the load of 0.7 yield strength; C: At 25℃ and the load of 0.8 yield strength; D: The load of 0.7 yield strength and different temperatures (yellow 25 and orange 50 ℃); E: At 25℃ and different load (yellow 0.7 and orange 0.8 yield strength).

to a notable decrease in creep strength and a rapid initiation of plastic deformation in the used materials, including PE and its nanocomposites. The reason for this effect could be attributed to the fact that an increase in temperature leads to the weakening of the bonds between the chains. As a result, the chains are able to slide past each other even under low levels of stress.

● At a constant temperature, when the stress level increases from 0.7 to 0.8, the creep rate also increases. This was due to the promotion of secondary bond breakage, which leads to an increased number of broken bonds. As a result, the polymer chains exhibited an enhanced mobility and demonstrate a higher degree of inter-chain sliding, facilitating easier movement between the polymer chains.

● PE nanocomposite with higher than 1wt% clay exhibited a noticeable drop in creep strength particularly at 50℃.

Conflicts of Interest
The authors declared no conflict of interest.

Author Contribution
Valojerdi MA participated in conducting experiments. Zebarjad SM and Moghim MH were involved in data analysis. Zebarjad SM contributed to the final writing. All authors contributed to the manuscript and approved the final version.

Abbreviation List
PE, Polyethylene

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