Investigation on the serrated flow behavior of bulk metallic glasses based on machine learning

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Abstract
This study proposed a long short-term memory (LSTM) model for predicting the serrated flow behaviors of bulk metallic glasses (BMGs) under nanoindentation. A series of load-controlled nanoindentation tests were conducted on a Pd40Cu30Ni10P20 BMG. The LSTM model was introduced to establish a neural network for predicting the serrated flow at different loading rates, and was verified by the comparisons of experimental data with predictive results. Further investigation based on the predictive serrated flows under different loading rates showed that the serrations exhibit a significant self-organized critical (SOC) phenomenon at different loading rates. The SOC phenomena of the serrations under a lower loading rate were more obvious than that under a higher loading rate.

1. Introduction

Bulk metallic glasses (BMGs) have received a great deal of attention due to their excellent properties, such as high yield strength, high elasticity limitation, large wear and corrosion resistance, and a measure of plastic deformation [1–3]. Therefore, BMGs are widely used as structural and functional materials in many high technology fields, such as precision machinery, aeronautics and astronautics, and military weapons [4–7]. Nevertheless, the application of BMGs is still greatly limited by the overall plasticity at room temperature [8]. In order to improve the workability of BMGs, enormous efforts have been made for its plastic deformation mechanism [8–12]. Generally, the plasticity of bulk metallic glasses is strongly related to the activities of shear bands of bulk metallic glasses during the loading process [10, 12]. However, it is difficult to obtain the plastic deformation mechanism of BMGs by monitoring the nucleation and expansion of shear bands. In nanoindentation, serration flow behavior has been observed in many BMGs, which is significantly associated with the plasticity of BMGs [13–15]. Considering that the serrated flow is accompanied by the plastic deformation of bulk metallic glasses, the plasticity of BMGs can be further explored by analyzing the serrated flow. In literatures, the effect of many factors, including loading rates, ductility, and chemical effects, on the serration flow behavior of BMGs has been investigated [16–19]. However, the serrated flow behavior of BMGs at a large indentation depth cannot be fully investigated due to the limitation of experimental techniques. In order to resolve this problem, Long Short-Term Memory (LSTM) neural network can be adopted as a suitable approach due to its long-term ability in self-studying and forecasting [20–22].

A series of load-controlled nanoindentation tests were carried out on a Pd-based BMG (Pd40Cu30Ni10P20). The serrations were identified and separated by using the proposed approach. Furthermore, long short-term memory was introduced to establish a neural network for predicting the serrated flow at different loading rates. The proposed LSTM model was verified by the comparisons of experimental and predictive results. Furthermore, the self-organized critical phenomena are further investigated based on the predictive serrated flow under different loading rates.
2. Experimental

The experimental material used in the study is Pd$_{40}$Cu$_{30}$Ni$_{10}$P$_{20}$ bulk metallic glass (abbreviated as Pd-based BMG) [23, 24]. The sample is a rod approximately 3 mm in diameter and 40 mm in length, which was cut into 5mm-thick cylinders. Load-controlled nanoindentation tests were performed on TI900 TriboIndenter system (Hysitron Inc.) with a Berkovich indenter. The resolution of load and displacement are 3 nN and 0.2 nm respectively. The peak load, $P_{\text{max}}$, was maintained at 100 mN, and the loading rates, $dP/dt$, were selected at 0.5, 5 and 10 mN/s. The holding time is 10 s at $P_{\text{max}}$. At least three measurements for each loading condition were performed to ensure the reliability of the experiment results. The experimental data used in this paper gained from earlier experiments, and the data have also been used in our previous studies [25].

3. The LSTM model

Machine learning methods have been widely used in the field of materials science [26, 27]. Recurrent Neural Networks (RNN) is one of machine learning methods which have been widely adopted to investigate variable-length sequence [26–28]. However, the vanishing gradient and exploding gradient may encounter while long-term dependencies are optimized by using RNN [29]. To resolve these problems, a lot of variations of RNNs such as BiRNNs, EMI-FastGRNN and MSC-RNN have been proposed [28, 30, 31]. The Long Short-Term Memory is one of the most effective variations having ability to deal with long-term dependencies with a special gated architecture [20–22]. A representative LSTM model consists of a forget gate, an input gate, an output gate, and a memory cell, as shown in figure 1.

In LSTM model, the forget gate can select the memory cell state at the previous step. Then, the input gate is adopted to control the component of memory cell state affected by the inputting vectors $x_t$. Finally, the output gate can influence the targeted outputs by the memory cell state. For a special input time series, where $x(t)$ is the time-dependent serration sequence in the current study, the numerical calculation cycle in the LSTM model is updated as follows:

$$
\begin{align*}
  f_t &= \sigma(W_f[h_{t-1}, x_{t-1}] + b_f) \quad \text{(Forget gate)} \\
  i_t &= \sigma(W_i[h_{t-1}, x_{t-1}] + b_i) \quad \text{(Input gate)} \\
  C_t &= \tanh(W_c[h_{t-1}, x_{t-1}] + b_c) \quad \text{(Candidate memory)} \\
  C_t &= f_t C_{t-1} + i_t C_t \quad \text{(Current memory)} \\
  o_t &= \sigma(W_o[h_{t-1}, x_{t-1}] + b_o) \quad \text{(Output gate)} \\
  h_t &= o_t \tanh(C_t) \quad \text{(Current hidden state)}
\end{align*}
$$

(1)

where $x_{t-1}$ refers to the previous input value. $h_{t-1}$ and $h_t$ are the previous and the current hidden states, respectively. $C_t$ and $C_{t-1}$ are previous and current cell memories. $W_f$, $W_i$, $W_c$, and $W_o$ denote the weight matrix of forget gate, input gate, candidate memory and output gate, respectively. $b_f$, $b_i$, $b_c$, and $b_o$ denote the bias matrix of
forget gate, input gate, candidate memory and output gate, respectively. $\sigma$ represents the sigmoid function which is defined as $\sigma(x) = 1/(1 + e^{-x})$, and $\tanh$ function is expressed as $\tanh(x) = (e^x - e^{-x})/(e^x + e^{-x})$.

4. Results

4.1. Experimental results

The representative curves of load versus displacement ($P-h$) at various loading rates in nanoindentation were plotted in figure 2. In this figure, the origin of each curve has been offset, so that the multiple curves can be distinguished. As shown in figure 2, an obvious serrated flow on the $P-h$ curves during loading periods was observed. Generally, the serrated flow of BMGs is strongly related to the activity of shear band [32, 33]. In nanoindentation, the nucleation of a shear band in BMGs is intermittent during loading process, leading a serrated flow in $P-h$ curves. In addition, it is also clear that the serrations significantly depend on loading rates, which is consistent with the experimental observations reported by Schuh et al [25, 34]. The number of recorded points in the serration process is much fewer than that in process, indicating the higher velocity of serrated flow.

An acceptable prediction on serrated flow acquired by machine learning method needs a typical serration dataset which has significant features. However, it is difficult to directly capture the details of serrations from the
$P-h$ curves under nanoindentation. The curves of displacement versus time ($h-t$) were taken as a breakthrough to analyze [35]. Firstly, obtained $dh/dt$-t curves by differentiating the $h-t$ curves using two-point forward-derivative algorithm, as shown in figure 3. The serration events shown as sharp bursts in the curve of $dh/dt-t$, and many fluctuations with higher frequencies but smaller amplitudes from the instrument and environment noise were also observed. Then eliminated the instrument and environment noise using a method of curve-fitting and statistical analysis, but preserve the original shape of the deformation curve. Lastly, the clear $dh/dt$-t curves at different loading rates can be obtained, as shown in figure 4. It can be observed that the $h-t$ curves obtained by integrating the $dh/dt$-t curves are very close to the experimental results, which indicates that these results are capable of supporting the identification and separation of serrations during nanoindentation.

Figure 4. The $dh/dt$-t curves without the noise fluctuations. The raw curves of $h-t$ were compared with the curves which obtained by the integral of the processed data of time derivative of displacement: (a) 0.5 mN s$^{-1}$, (b) 5 mN s$^{-1}$, (c) 10 mN s$^{-1}$. Reprinted from [35], Copyright (2017), with permission from Elsevier.
4.2. LSTM model prediction results

The processed experimental results were used as datasets for the LSTM model. The proportion of training, testing and validation datasets were chosen as 70%, 15% and 15%, respectively. In addition, the number of hidden layers and nodes can significantly affect the performance of the LSTM model. After numerous experiment sessions, the single hidden layer with 200 hidden nodes was employed in the model due to the minimum absolute percentage error. The learning rate set as 0.0001, epochs as 300, the full batch learning was adopted then the mini-batch size was set as 8000.

![Figure 5. The predicted \( \frac{dh}{dt} \) results by LSTM model compare to experimental results.]
The $dh/dt$-t responses of Pd-based BMG under nanoindentation at various loading rates was simulated by the proposed LSTM model. The comparisons between the predicted and experimental results were illustrated in figure 5. As can be seen, the predicted results are very close to the experimental results, which implies that the proposed LSTM model can effectively inherit the intrinsic features of the original dataset obtained from laboratory experiments, and precisely characterize the serrated flow of Pd-based BMG under nanoindentation.

Figure 6. The $h$-t plots of experimental and LSTM model results at: (a) 0.5 mN s$^{-1}$, (b) 5 mN s$^{-1}$, (c) 10 mN s$^{-1}$.

The $dh/dt$-t plot can be obtained by integrating the $dh$-t plot, as shown in figure 6. The coefficient of determination ($R^2$) of the predicted and the experimental results for three various loading rates 0.5 mN s$^{-1}$, 5 mN s$^{-1}$ and 10 mN s$^{-1}$ are 0.925, 0.906, and 0.912, respectively. That indicates the proposed LSTM model can accurately predict the serrated flow of Pd-based BMG under nanoindentation. The predicted results obtained by
multi-step predictive output of the LSTM model present a similar tendency as the earlier period. The serrations can be observed at a large indentation depth either.

5. Discussions

The distribution of serration size ($\Delta h$) with time obtained from LSTM model predicted results were illustrated in figure 7. Statistical distributions of $\Delta h$ were illustrated in figure 8 as bar charts, the cumulative distributions of

Figure 7. The scatter plot of serration size versus time at various loading rates: (a) 0.5 mNs$^{-1}$, (b) 5 mNs$^{-1}$, (c) 10 mNs$^{-1}$. 
\[ \Delta h \text{ were calculated and inseted in figure 8. The cumulative distribution curve can be well fitted by the empirical relation } [36–38]: \]

\[ F = A \Delta h^{-\alpha} \exp \left[ -\left( \frac{\Delta h}{\Delta h_c} \right)^2 \right] \]

(2)

where \( A \) and \( \alpha \) represent the normalization parameter and scaling parameter, respectively, and \( \Delta h_c \) is the critical size of serrations [39]. From the equation (2), it can clearly notice that the serrations size which is smaller
than \(\Delta h\) obeys the power-law distribution well, while the serrations size which is larger than \(\Delta h\) is constrained by the exponential decay.

The power-law relation is usually indicative of the self-organized critical (SOC) phenomena in dynamics. As seen in figure 8, the fitted value of \(\Delta h\) means that the serration size smaller than \(\Delta h\) follow the SOC behavior. The ratio of \(I = \frac{n_{soc}}{n_{serration}}\) in further introduced to quantitatively characterize the intensity of SOC phenomena at different loading rates. As \(n_{soc}\) refers to the number of serrations following the SOC behavior, \(n_{serration}\) refers to the total number of serrations. It is clear that the serrations exhibit a significant SOC behavior at different loading rates. In addition, the value of the index for three different loading rates (0.5 mN s\(^{-1}\), 5 mN s\(^{-1}\), 10 mN s\(^{-1}\)) are 0.953, 0.744, and 0.731, respectively, thus implying that the SOC phenomena under a lower loading rate is more obvious than that under a higher loading rate. SOC reflects the stability of the deformation process, larger \(I\) value means the better stability and plastic deformation capacity of BMG deformation process under nanoindentation. It is reasonable to assume that the loading rate affecting the formation and evolution of flow units in BMGs.

6. Conclusion

In this study, a series of load-controlled nanoindentation tests were carried out on a Pd-based BMG. The serrations of experimental results were identified and separated. The experimental results were used to training and validating a LSTM neural network model for predicting the serrated flow of BMGs at a large indentation depth during nanoindentation. The self-organized critical phenomena are further investigated based on the predictive serrated flow under different loading rates. It is found that the serrations exhibit a significant self-organized critical phenomenon at different loading rates, and the self-organized critical phenomenon of the serrations under a lower loading rate is more obvious than that under a higher loading rate. Although we only present a preliminary analysis and discussion of the serrated flow of BMGs in this paper, it could also provide reference and help for further study of the mechanical properties of these alloys.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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References

[1] Yavari A R, Lewandowski J J and Eckert J 2007 Mechanical properties of bulk metallic glasses MRS Bull. 32 635–8
[2] Greer A L and Ma E 2007 Bulk metallic glasses: at the cutting edge of metals research MRS Bull. 32 611–9
[3] Huang Y J, Shen J and Sun J F 2007 Bulk metallic glasses: smaller is softer Mater. Res. Express 4 064402
[4] Ponnambalam V, Poon S J, Shi P Y, Huang Y J, Shen J and Sun J F 2007 Bulk metallic glasses: smaller is softer Mater. Res. Express 4 064402
[5] Greer A L and Ma E 2007 Bulk metallic glasses: at the cutting edge of metals research MRS Bull. 32 611–9
[6] Cearbhaill E D, Byrne J H and Browne D J 2016 Bulk metallic glasses for implantable medical devices and surgical tools Adv. Mater. 28 5753–62
[7] Rahaman M L, Zhang L, Liu M and Liu W 2015 Surface roughness effect on the friction and wear of bulk metallic glasses Wear 332–333 1231–7
[8] Sun Y et al In vitro and in vivo biocompatibility of an Ag-bearing Zr-based bulk metallic glass for potential medical use J. Non. Cryst. Solids. 419 2013 82–91
[9] Chen M, Inoue A, Zhang W and Sakurai T 2006 Extraordinary plasticity of ductile bulk metallic glasses Phys. Rev. Lett. 96 245502
[10] Park E and Kim D 2006 Phase separation and enhancement of plasticity in Cu–Zr–Al–Y bulk metallic glasses Acta Mater. 54 2597–604
[11] Men H, Pang S, Inoue A and Zhang T 2003 New Ti-based bulk metallic glasses with significant plasticity Mater. Trans. 46 2218–20
[12] Yang Q, Mota A and Ortiz M 2006 A finite-deformation constitutive model of bulk metallic glass plasticity Comput. Mech. 37 194–204
[13] Rashidi R, Malekan M and Hamishebahar Y 2019 Serration dynamics in the presence of chemical heterogeneities for a Cu-Zr based bulk metallic glass J. Alloys Compd. 775 298–303
[14] Wang Z, Qiao J W, Tian H, Sun B A, Wang B C, Xu B S and Chen M W 2015 Composition mediated serration dynamics in Zr-based bulk metallic glasses Appl. Phys. Lett. 107 201902
[15] Tessler M M and Thadhani N N 2010 Mechanical properties of bulk metallic glasses Prog. Mater. Sci. 55 759–839
[16] Pi J H, Wang Z Z, He X C and Bai Y Q 2018 Hardness and modulus of Cu-based bulk metallic glasses via nanoindentation Rare Met. Mater. Eng. 47 479–84
[17] Bei H, Xie S and George E P 2006 Softening caused by profuse shear banding in a bulk metallic glass Phys. Rev. Lett. 96 105503
[18] Guo F Q, Poon S J and Shiflet G J 2005 Enhanced bulk metallic glass formability by combining chemical compatibility and atomic size effects J. Appl. Phys. 97 013512
[19] Shao L 2020 Effect of chemical composition on the fracture toughness of bulk metallic glasses Mater. Sci. Eng. A 753 138001
[20] Hochreiter S and Schmidhuber J 1997 Long Short-Term Memory Neural Comput. 9 1735–80
[21] Gers F A, Schmidhuber J and Cummins F 2000 Learning to forget: continual prediction with LSTM Neural Comput. 12 2451–71
[22] Fan E 2000 Extended tanh-function method and its applications to nonlinear equations Phys. Lett. A 277 212–8
[23] Inoue A, Nishiyama N and Masumoto T 1996 Preparation of bulk glassy Pd40Cu30Ni10P20 alloy of 40 mm in diameter by water quenching Mater. Trans., JIM 37 181–4
[24] Xu F, Li K F, Deng X H, Zhang P and Long Z L 2016 Research on viscoelastic behavior and rheological constitutive parameters of metallic glasses based on fractional-differential rheological model Acta Phys. Sin. 65 046101
[25] Schuh C A, Argon A S, Nieh T G and Wadsworth J 2003 The transition from localized to homogeneous plasticity during nanoindentation of an amorphous metal Philos. Mag. 83 2585–97
[26] Ren B, Long Z and Deng R 2021 A new criterion for predicting the glass-forming ability of alloys based on machine learning Comput. Mater. Sci. 189 110259
[27] Peng L, Long Z L and Zhao M S Z 2021 Determination of glass forming ability of bulk metallic glasses based on machine learning Comput. Mater. Sci. 195 110480
[28] Palidán M, Maïmaitiyiyu, Yang W Z and Wushouer S 2019 Uyghur sentiment rhythm phrase attention model based on BiRNN J. Univ. Electron. Sci. Technol. China. 48 1–8
[29] Lukschvičius M and Jaeger H 2009 Reservoir computing approaches to recurrent neural network training Comput. Sci. Rev. 3 127–49
[30] Roy D, Srivastava S, Kusupati A, Jain P, Varma M and Arora A 2021 One size does not fit all: multi-scale, cascaded RNNs for radar classification ACM Trans. Sens. Networks 17 1–27
[31] Wu J, Woo H, Yamura Y, Moro A, Massaroli S, Yamashita A and Asama H 2019 Pedestrian trajectory prediction using BiRNN encoder–decoder framework Adv. Robot. 33 956–69
[32] Qiao J W, Zhang Y and Liaw P K 2010 Serrated flow kinetics in a Zr-based bulk metallic glass Intermetallics 18 2057–64
[33] Dubach A, Dalla Torre F H and Löffler J F 2007 Deformation kinetics in Zr-based bulk metallic glasses and its dependence on temperature and strain-rate sensitivity Philos. Mag. Lett. 87 695–704
[34] Schuh C A, Lund A C and Nieh T G 2004 New regime of homogeneous flow in the deformation map of metallic glasses: elevated temperature nanoindentation experiments and mechanical modeling Acta Mater. 52 5879–91
[35] Liao G, Long Z, Zhao M, Zhong M, Liu W and Chai W 2017 Serrated flow behavior in a Pd-based bulk metallic glass under nanoindentation J. Non. Cryst. Solids. 460 47–53
[36] Wang G, Chan K C, Xia L, Yu P, Shen J and Wang W H 2009 Self-organized intermittent plastic flow in bulk metallic glasses Acta Mater. 57 6140–55
[37] Sun B A, Pauly S, Tan J, Stoica M, Wang W H, Kühn U and Eckert J 2012 Serrated flow and stick–slip deformation dynamics in the presence of shear-band interactions for a Zr-based metallic glass Acta Mater. 60 4160–71
[38] Sun B A, Yu H B, Jiao W, Bai H Y, Zhao D Q and Wang W H 2010 Plasticity of ductile metallic glasses: a self-organized critical state Phys. Rev. Lett. 105 035501
[39] Bian X L, Wang G, Chan K C, Ren J L, Gao Y L and Zhai Q J 2013 Shear avalanches in metallic glasses under nanoindentation: Deformation units and rate dependent strain burst cut-off Appl. Phys. Lett. 103 101907