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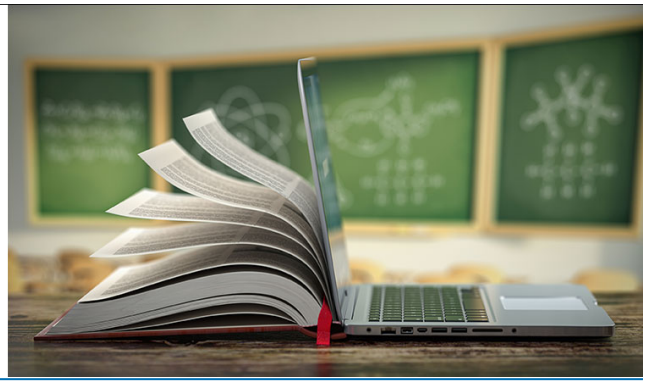

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The Use of Finite Mixture Models and EM-Algorithm to Analyze Grain Structure in HPT-Nanostructured Metallic Materials

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Abstract. Analysis of grain size distribution's histograms of Nb and Ni subjected to high-pressure torsion at cryogenic temperatures and Nb_3Sn layers formed in Nb/Cu–Sn composite wires after the diffusion annealing has been carried out using statistical analysis method based on the application of finite mixture models and using an expectation–maximization algorithm with the estimation of fitting accuracy by the Bayesian information criterion. It has been established that the approximation by the model with a single component of logarithmic standard distribution is the most suitable for all examined experimental distributions in contrast to the model with two components. Besides, the use of the proposed approach allows to practically eliminate an influence of the additional errors in the experimental data which seem to be introduced at transmission electron microscopy image processing and constructing histograms of grain size distributions.

1. Introduction

In the last decades materials with ultrafine-grained (UFG) structure attract great attention of the researchers due to their unique physical and mechanical properties. UFG materials possess a grain size below 1000 nm and exhibit significantly higher strength, unusual mechanical behavior and a complex of unique properties in comparison to their coarse-grained counterparts [1–3].

The application of severe plastic deformation (SPD) enables obtaining bulk UFG materials. A number of various techniques of SPD have been developed by now [4], and one of the earliest and most common among them is high-pressure torsion (HPT) [5,6]. This method makes it possible to produce a structure close to nanocrystalline [7–14] at room temperatures, and HPT at cryogenic temperatures allowed the authors of [15–19] to obtain a true nanocrystalline structure in pure metals.

It was shown [20–22] that grain boundaries as an element of structure playing a determinative role in UFG materials markedly differ from those in coarse-grained polycrystals and are referred to as "non-equilibrium" or deformation-modified boundaries. Besides, such grain boundaries are characterized by high density of defects and, accordingly, enhanced energy.

Since the grain structure is the most important factor responsible for the special properties of such materials, in the studies devoted to investigations of structure in nanostructured materials [7–10,13–16,18,19] transmission electron microscopy (TEM) with a subsequent processing of TEM images is

used. However, in a lot of these studies, the statistical analysis of structure usually deals with the grain sizes distributions described by a logarithmic standard (normal) distribution. At the same time, it was shown [23] that the histograms of the grain size distribution in the nanostructured materials both of the HPT and of the nanostructures of recrystallization origin are not always described only by the logarithmic standard distribution. Besides, the different models can be used to fit the experimental distributions. That may not lead to the same results because of different accuracy of fitting techniques and models [24, 25]. Since experimental distributions may be a mixture of several log-normal distributions with different values of the mean, standard deviation and volume fraction, a small amount of one of distributions may be estimated as contamination. Moreover, a logarithmic normal distribution is not symmetrical that additionally complicates the process of fitting.

Therefore, the aim of this work is to propose a robust statistical approach to analyze experimental grain sizes distributions.

2. Results and discussion

The results of studies [15, 18] devoted to nanostructuring of pure metals by HPT at cryogenic temperatures had been used. It is assumed that an effect of recrystallization both during deformation and after that was completely eliminated. The data on the nanostructure formed during the annealing of highly deformed composites is used [26] as well.

The analysis of grain sizes distributions was performed in terms of that the experimental distributions of grains are described by lognormal function [27]. Pearson's mixture model-based approach known as finite mixture models was chosen to describe the experimental distributions [26]. An expectation-maximization (EM) algorithm [28, 29] has been used for fitting the experimental distributions.

In addition, it is suggested that the experimental distribution may be both single distribution and a sum of separated logarithmic standard distributions, so the expression of the model is as follows:

$$f(x)=\sum_{i=1}^{N} A_i \cdot \frac{1}{x} \cdot \frac{1}{\sqrt{2\pi}\cdot\sigma_i} \cdot e^{-\frac{(\ln x-\mu_i)^2}{2\sigma_i^2}}, \quad (1)$$

where μ_i and σ_i are the mean and the standard deviation of the variable of the natural logarithm distribution; A_i is a scale factor, which is the estimation of volume fraction of the grain group in the experimental distribution; i – grain group number; N – the amount of distributions in the model.

EM-algorithm is common technique used to determine the parameters of a mixture. At the same time, EM-algorithm is of particular appeal for finite normal mixtures with an a priori given number of components [28, 29]. Therefore, in order to use EM-algorithm with the most efficiently it is necessary to describe experimental distribution as normal distribution's mixture.

Since, a log-normal distribution is a continuous probability distribution of a random variable whose logarithm is normally distributed, the model (1) can be represented in logarithmic scale as:

$$f(x)=\sum_{i=1}^{N} A_i \cdot \frac{1}{\sqrt{2\pi}\cdot\sigma_i} \cdot e^{-\frac{(\ln x-\mu_i)^2}{2\sigma_i^2}}, \quad (2)$$

Due to μ_i and σ_i have logarithmic scale, these parameters should be turned into normal scale by follows expressions:

$$M_i = e^{\mu_i+\frac{\sigma_i^2}{2}}, \quad (3)$$

$$S_i = \sqrt{\left(e^{\sigma_i^2}-1\right)\cdot e^{2\cdot\mu_i+\sigma_i^2}}, \quad (4)$$

where M_i and S_i are the mean and the standard deviation of the standard distribution in the normal scale, respectively.

It should be noted that the approach assumes that initial data for processing is a raw set of grain sizes. In order to process experimental grain sizes distributions, the distributions should be decomposed into a raw set of grain sizes in contrast to fitting of distributions by curves in accordance with the technique of work [23–25]. The next step of processing is to represent the set of grain sizes on the logarithmic scale. Then the data was processed by EM-algorithm with different the amount of distributions (N) in accordance with the model. The advantage of the technique is the use of EM-algorithm with enough amount of random start points to avoid incorrect results. Moreover, modern implementations of EM-algorithm for fitting of a Gaussian mixture model allow to compute the Bayesian information criterion (BIC) [30] to assess the number of clusters in the data. That can be used to select the number of components (N) in a model in an efficient way.

In Table 1, the results of the calculation are summarized, as well. It can be seen that the approximation by the model with a single component (N = 1), in accordance with the calculated BIC values, is the most suitable for both the grain structure of Nb and Ni subjected to HPT at cryogenic temperatures and Nb_3Sn structure (Composite) formed in Nb/Cu–Sn composite wires after the diffusion annealing in contrast to the model with two components (N = 2).

Table 1. Results of approximation.

Material	***M*** **(nm)**	***S*** **(nm)**	A_i	**BIC**
Approximation by the model with a single component (N = 1)				
Nb (HPT 80K)	77.42	22.16	1	95.81
Ni (HPT 80K)	92.62	29.17	1	53.69
Composite	72.87	20.19	1	343.59
Approximation by the model with two components (N = 2)				
Nb (HPT 80K)	76.06	25.21	0.62	107.71
	79.55	14.47	0.38	
Ni (HPT 80K)	91.10	31.96	0.78	60.59
	97.62	7.78	0.22	
Composite	71.97	20.59	0.89	359.85
	79.85	11.39	0.11	

In addition, the data processing results of experimental grain sizes histograms from works [15,18,26] with using the most suitable model are represented in Figure 1 where the experimental histograms and calculated curves in both normal and logarithmic scale are shown.

Earlier [23] it was shown that the fitting of the experimental grain sizes histograms by only single log-normal distribution gives significant imprecision for all these materials. Whereas, use of additional normal distribution in the fitting model allowed to describe the experimental data with much greater accuracy in accordance with the calculated chi-square values. Besides, the fraction of the additionally introduced standard distribution in all cases was considerable and even exceeded the fraction of the logarithmic distribution in the case of Nb.

It can be concluded that the common technique based on the analysis of TEM image with a subsequent processing and constructing histograms of grain size distribution introduce additional normally distribution errors. This fact seems to be the main reason for appearing the additional normal

distribution in experimental histograms. At the same time, the use of statistical analysis to describe the experimental distributions in terms of finite mixture models with using EM-algorithm allowed to practically eliminate an influence of the additional errors in the experimental data.

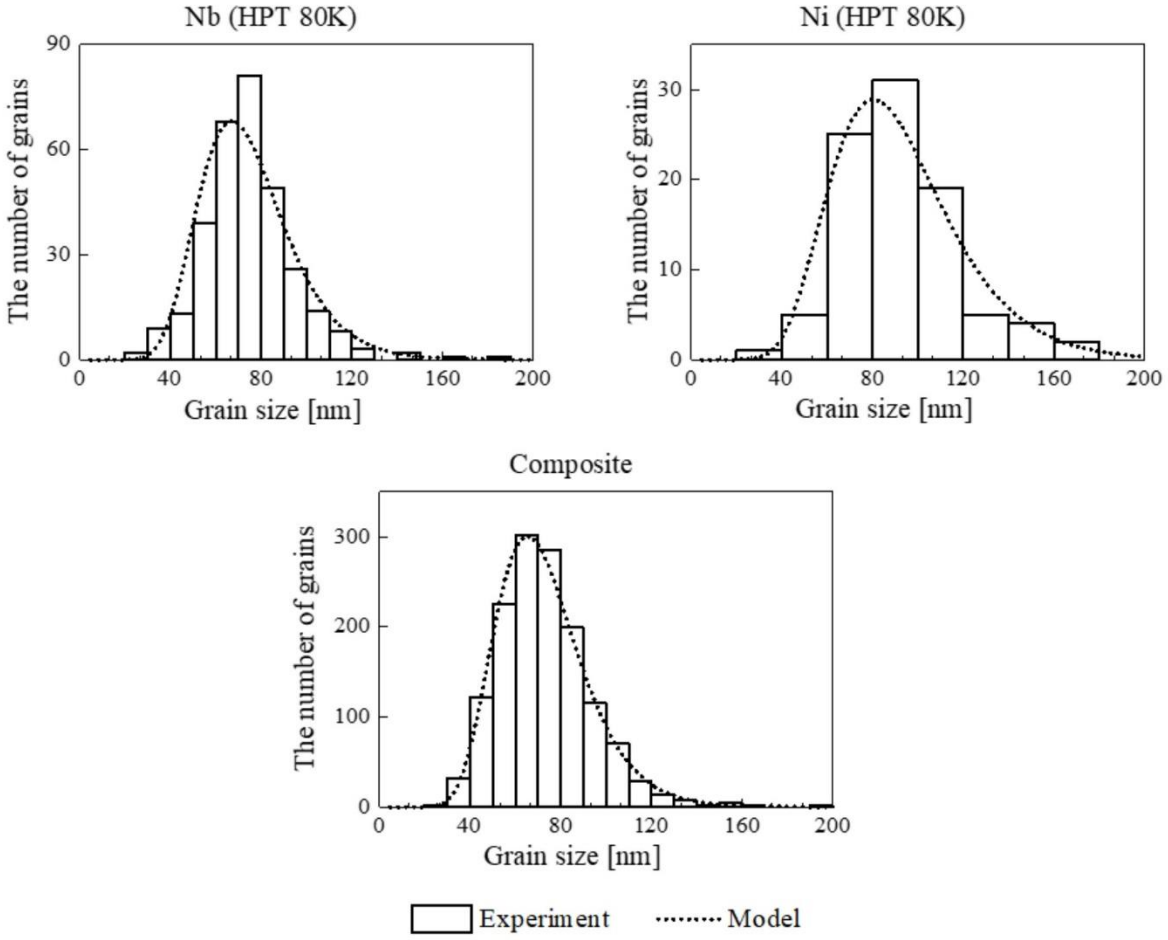

Figure 1. Experimental histograms of grain size distribution [15,18,26] and approximation results.

3. Summary

The analysis of grain sizes distributions was performed in terms of that the experimental distributions of grains are described by the lognormal function. Pearson's mixture model-based was chosen to describe the experimental distributions. An expectation-maximization algorithm has been used for fitting the experimental distributions of the grain structure of Nb and Ni subjected to HPT at cryogenic temperatures and Nb_3Sn structure formed in Nb/Cu–Sn composite wires after the diffusion annealing.

The analysis of fitting accuracy results was shown that the approximation by the model with a single component is the most suitable for all examined experimental distributions in contrast to the model with two components. Besides, the use of the proposed approach allows to practically eliminate an influence of the additional errors in the experimental data which are introduced at TEM image processing and constructing histograms of grain size distributions.

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