Structural changes in aluminium based metallic glasses subjected to severe plastic deformation

Thesis

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Introduction

The dawn of the 21st century is characterized by the appearance of new demands, at the field of energy management, environmental technologies, information technology, human medicine and space research. The new needs require the creation of new materials, structures, properties and technologies.

The task of materials science is to produce and characterize these materials. Classical materials science works in a closed system of disciplines (thermodynamics, optics, statistical physics, transport phenomenon, atomic and nuclear physics etc…), in which the production determines the structure and the properties in order to be best tailored for the appropriate applications. This concept has led to the development of the so-called "advanced" materials that are consciously produced and have controlled structure with well-defined properties.

Due to their low density, aluminium based alloys are widely used in engineering applications, particularly in the field of aerospace and automotive industry. Applications are only limited by their low yield strength that couldn't be increased over 500-600 MPa by the classical strengthening mechanisms. In order to improve the mechanical properties new mechanisms and structures have to be developed. As a result of this research aluminium alloys with non-periodic atomic structure have been discovered in the late 1980's, posessing a yield strength close to 1 GPa. The 1 GPa limit has been exceeded by the homogeneous precipitation of nanoscale α-Al particles into the amorphous matrix. Such nanoscale crystalline particles can be induced either by heat treatments or plastic deformation. One aim of the thesis is to describe and compare the structural changes occurring during the above mentioned two different methods.

Today, beside the excellent mechanical properties, the need for downsizing can increasingly be felt. Materials can be manipulated at nanoscale or in some cases even at the atomic dimensions. Recent research activities tend to develop new technologies and materials, that can meet the challenges occurring in this size range. Because of the lack of crystal defects small metallic glass pieces can be shaped into very smooth surfaces, which can be used as raw materials of microscopic molds, micro-gears or mico-sensors.

Aims

Since amorphous alloys - and especially Al-based metallic glasses – can only be produced in limited sizes, some kind of compaction of small metallic glass pieces needs for practical application. Unfortunately, the traditional compaction methods themselves can undermine the excellent mechanical properties of metallic glasses. Therefore, compaction as a new alternative way has been carried out by high pressure torsion, applying severe plastic deformation at low temperature. In this thesis structural changes, occurring in metallic glasses during severe plastic deformation methods will be investigated. Deformation induced crystallization of metallic glasses is a well known phenomenon, but despite of the many experimental results, its basic mechanism is still unclear.

In the present work, deformation induced structural relaxation and crystallization of metallic glasses is examined in detail during severe plastic deformation. Some experimental results show similarities but others are contrary to the properties of thermally activated crystallization. Our goal is to understand the formation of structures obtained by deformation in order to tailor the desired mechanical properties.

Investigating the deformation induced crystallization from the classical perspective of crystallization, crystal nucleation and growth has been analyzed separately. The effect of high pressure, occurring usually during severe plastic deformation methods, on crystal nucleation and large shear deformation on crystal growth has also been studied. In addition, it has been investigated how deformation influence the glass transition phenomenon, in order to get a better understanding of the deformed glassy state.

Methods

Fully amorphous $Al_{85}Y_8Ni_5Co_2$, $Al_{85}Ce_8Ni_5Co_2$, $Al_{85}Gd_8Ni_5Co_2$ alloys have been obtained using a single roller melt spinning technique in inert atmosphere. As-quenched ribbons were cut into small pieces (flakes) and then compacted by high pressure torsion at low temperature, resulting in a disk-shaped sample. For comparison, the impact of isothermal heat treatment, and another severe plastic deformation method (ball milling) has also been examined.

Morphological properties were characterized by Scanning Electron Microscopy and the microstructure by Transmission Electron Microscopy and X-ray diffraction. Some portion of X-ray diffraction measurements were carried out at the ID-11 beamline of the European Synchrotron Radiation Facility (ESRF), where the disks were mapped by taking two-dimensional diffraction patterns along a couple of diameters with a step size of 300 μm.

Isothermal annealings and thermal stability investigations have been performed on Differential Scanning Calorimeter at the Eötvös Unicersity (ELTE).

Mechanical properties of the samples have been characterized by dynamic hardness measurements. To perform the measurements the small samples have been embedded into an epoxy medium.

Thesis points

1. Near to the glass transition temperature the change between the different crystallization routes is clearly seen during isothermal heat treatments of thermally unstable $Al_{85}Ce_8Ni_5Co_2$ and $Al_{85}Gd_8Ni_5Co_2$ alloys. In this temperature range the transition in atomic mobility plays crucial role in the crystallization kinetics. Accordingly, we have shown that below the glass transition temperature, where the percolation of free volume is blocked, the growth of crystalline phases with low molar volume is preferred, whereas the growth of phases with high molar volume is restricted because of the variance of diffusion coefficient with the local density fluctuations. [S4], [S5], [S7]

2. As a new, alternative method high pressure torsion has successfully been applied to produce bulk Al-based metallic glasses ($Al_{85}Y_8Ni_5Co_2$, $Al_{85}Ce_8Ni_5Co_2$ and $Al_{85}Gd_8Ni_5Co_2$). Low-porosity, bulk disks have been obtained from the as quenched ribbons, which possess higher hardness than the initial states. Due to severe plastic deformation structural transformations takes place, resulting crystalline precipitations with significantly different size. Crystalline particles wits submicron or micrometer size couldn't only be formed by a simple heat treatment, so the applied deformation process gives new opportunities for customizing aluminium alloys. [S1], [S2], [S3], [S6], [S7]

3. Crystal nucleation and growth during severe plastic deformation can significantly differ from the crystallization occurring during thermal activation. The reasons for the differences have been discussed from different aspects, such as the effect of high pressure on nucleation. With the supplement of the classical nucleation theory, we have shown that there exists a critical pressure in the Al-Ce system, above which the precipitation of α-Al is preferred, instead of the intermetallic $Al_{11}Ce_3$. In addition, the presence of the large submicron or even micometer sized particles is explained by the increased atomic mobility induced by shear deformation. The effect of shear deformation on the growth of smaller crystalline nuclei has been revealed and experimentally verified in subsequent heat treatments by the shift of thermally activated relaxation procedures to lower temperature. The results obtained on $Al_{85}Y_8Ni_5Co_2$ and $Al_{85}Gd_8Ni_5Co_2$ alloys clearly show that the effect of severe plastic deformation couldn't be substituted by a simple heat treatment, but it holds the sign of many different annealings simultaneously. [S5], [S6], [S7]

4. Based on heat capacity measurments we have showed that the effect of structural changes occured during high pressure torsion persists even in the supercooled liquid state. General behavior of metallic glasses subjected to high pressure torsion has been described based on free volume theory and concrete numerical calculations have been performed in the case of the most commonly used Zr-based bulk metallic glass. According to the numerical results, dynamic equilibrium evolves in the sample independently of the deformation rate. The detailed investigation of the equilibrium states reveals direct connection between the experimentally measurable torque and other parameters of the sample, like temperature increase, that allows us to draw in-situ conclusions about the conditions occurring during deformation. [S8], [S9]

Publications

S1. P. Henits, Á. Révész, A.P. Zhilyeav and Zs. Kovács
"Severe plastic deformation induced nanocrystallization of melt-spun $Al_{85}Y_8Ni_5Co_2$ amorphous alloy"
Journal of Alloys and Compounds **461** (2008) 195

S2. Zs. Kovács, P. Henits, S. Hóbor and Á. Révész
"Nanocrystallization process in amorphous alloys during severe plastic deformation and thermal treatments"
Reviews on Advanced Materials Science **18** (2008) 593

S3. P. Henits, Zs. Kovács and Á. Révész
"Crystallization of amorphous $Al_{85}Ce_8Ni_5Co_2$ HPT-alloy"
Reviews on Advanced Materials Science **18** (2008) 597

S4. P. Henits, Zs. Kovács, L.K. Varga and Á. Révész
"Nanocrystallization in $Al_{85}Ce_8Ni_5Co_2$ amorphous alloy induced by heat treatment and severe plastic deformation"
Journal of Physics Conference Series **144** (2009) 012095

S5. P. Henits , Á. Révész, L.K. Varga and Zs. Kovács
"The evolution of the microstructure in amorphous $Al_{85}Ce_8Ni_5Co_2$ alloy during heat treatment and severe plastic deformation: A comparative study"
Intermetallics, *submitted*

S6. P. Henits, Zs. Kovács, E. Schaffler, L. K. Varga, J.L. Lábár and Á. Révész,
"Nanocrystallization in $Al_{85}Ce_8Ni_5Co_2$ amorphous alloy obtained by different strain rate during high pressure torsion"
Journal of Alloys and Compounds, *in press*
(doi:10.1016/j.jallcom.2010.03.145)

S7. P. Henits, Á. Révész, E. Schafler, P.J. Szabó, J. L. Lábár, L.K. Varga and Zs. Kovács
"Correlation between microstructural evolution during high pressure torsion and isothermal heat treatment of amorphous $Al_{85}Gd_8Ni_5Co_2$ alloy"
Journal of Materials Research **25** (2010) 1883

S8. Á. Révész, P. Henits and Zs. Kovács
„Structural changes in Zr-based bulk metallic glasses deformed by high pressure torsion"
Journal of Alloys and Compounds **495** (2010) 338

S9. P. Henits, Á. Révész and Zs. Kovács
"Steady state daformation in metallic glasses based on free volume theory"
submitted