Available online at www.sciencedirect.com
ScienceDirect
Trans. Nonferrous Met. Soc. China 30(2020) 2749−2756
Transactions of Nonferrous Metals Society of China
www.tnmsc.cn

Microstructure and mechanical properties of fine grained uranium prepared by ECAP and subsequent intermediate heat treatment

Dong-li ZOU, Li-feng HE, Da-wu XIAO, Ya-wen ZHAO, Zhi-cong QIU, Chao LU, Fan LIU

Institute of Materials, China Academy of Engineering Physics, Jiangyou 621907, China

Received 14 November 2019; accepted 7 August 2020

Abstract: The microstructure and mechanical properties of fine grained uranium prepared by equal channel angular pressing (ECAP) and subsequent intermediate heat treatment were investigated systematically by the confocal laser scanning microscope (CLSM), electron backscatter diffraction (EBSD) and split Hopkinson pressure bar (SHPB). The results show that the initial coarse grained uranium was refined from about 1000 to 6.5 μm prepared by ECAP at 3 passes and subsequent heat treatment, and the corresponding dynamic yield strength increased from 135 to 390 MPa. For the ECAPed uranium samples, the relationship between grain size and yield strength could be described by classical Hall−Petch relationship, and the fitting Hall−Petch relationship for the fine grained uranium samples prepared by ECAP was drawn.

Key words: depleted uranium; equal channel angular pressing (ECAP); heat treatment; grain refinement; severe plastic deformation

1 Introduction

Equal channel angular pressing (ECAP), as a widely used severe plastic deformation (SPD) processing technique, is the most efficient method to refine grains and enhance the comprehensive properties [1]. In this technique, the specimen is pressed through a die with two intersecting channels equal in cross-section and deformed via a simple shear at the intersection of angular channels. At present, there have been many relevant literatures about fine grained materials produced by ECAP process at room temperature, including face centered cubic (FCC) Al [2−5], Cu [6,7], steel [8,9], hexagonal close packed (HCP) Mg [10,11], Ti [12−14], and Zr [15,16], and the corresponding microstructure and mechanical properties of fine grained materials prepared by ECAP have been characterized and measured.

Depleted uranium (DU) is an orthorhombic crystal structure (space group of *Cmcm*), which is made up of corrugated sheets of uranium atoms parallel to the (010) or *ac* planes. The depth or amplitude of corrugation within each sheet is along the [010] direction and the width of corrugation is along the [001] direction, and the separated distance of the corrugated planes is $b/2$ (b is lattice parameter along the [010] direction) [17]. The low symmetry and aeolotropy of uranium lead to the deformation twins, which is considered as the dominant plastic deformation mechanism under quasi-static loading [18,19], and uranium exhibits several different twinning systems, including type I, type II twins, and interacted compound twins. To our knowledge, the preparation of fine grained uranium has been reported limitedly in the literature due to its low symmetry, high density, high hardness and strength. REN et al [20,21] prepared the fine grained U−5.5wt.%Nb by cold and heat treatments, and found that the strength of fine grained alloy was lower than that of coarse grained

Foundation item: Project (51401187) supported by the National Natural Science Foundation of China; Projects (2014B0301046, 2015B0301066) supported by the Science Development Fund of China Academy of Engineering Physics
Corresponding author: Dong-li ZOU; Tel: +86-816-3626748; E-mail: dlzou1980@163.com
DOI: 10.1016/S1003-6326(20)65417-6

sample. GARLEA et al [22] manipulated the grain size in cast uranium by shock deformation and annealing techniques, and significant grain size reduction to ~92 μm was obtained. Four deformation twin modes were indentified, and the shock loading technique was proposed as an attractive alternative approach for controlling cast uranium grain size and its mechanical properties. Though several works have been performed on the grain refinement and mechanical properties of uranium, limited results associated with the fine grained uranium with grain size reduction to several micrometers were available in previous references. Thus, it is necessary to investigate the micro-structure evolution and mechanical properties of uranium with different grain sizes prepared by ECAP at different passes.

The objective of the present work is to investigate the preparation process of fine grained uranium by ECAP and subsequent intermediate heat treatment, and to evaluate the mechanical properties of fine grained uranium. The microstructure of uranium prepared by ECAP at every pass was characterized, and the corresponding mechanical properties of uranium with different grain sizes were measured.

2 Experimental

DU with a diameter of about 4 mm and a length of about 10 mm was selected as the original sample, and the original uranium billet had the coarse grains with diameter ranging from several hundreds micrometers to several millimeters.

ECAP was conducted at room temperature adopting a die with an internal angle Φ of 120° and an outer curvature angle ψ of 60°, as shown in Fig. 1. The imposed effective strain ε of ~0.6 in present ECAP die at each pass can be calculated by following equation [15]:

$$\varepsilon=\frac{1}{\sqrt{3}}\left[2\cot\left(\frac{\Phi}{2}+\frac{\psi}{2}\right)+\psi\operatorname{cosec}\left(\frac{\Phi}{2}+\frac{\psi}{2}\right)\right] \quad (1)$$

Molybdenum disulphide (MoS_2) was used as a lubricant, and the ram speed was set to be 0.2 mm/min for all of the experiments. Firstly, the uranium billet was pressed by ECAP at 1 pass, and then annealed at 500 °C for 15 min. Furthermore, the pressed sample was inserted into a die at a rotation angle of 90° for the second pass and the third pass, and the same processing procedure was performed for the uranium billet at 2 and 3 passes.

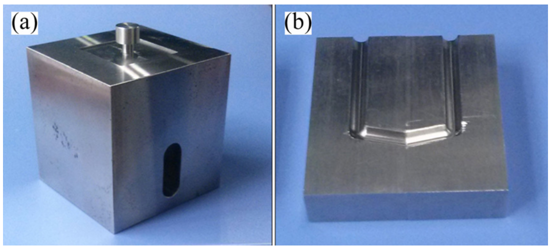

Fig. 1 Die illustration (a) and inner channel angle (b) of ECAP

The optical characterization samples of uranium pressed by ECAP were cut from the flow plane, then mechanically polished and electrochemically etched in a 5% phosphoric acid water solution operated at 5 V for 30 s. The metallographic microstructure was observed by a confocal laser scanning microscope (CLSM, MRC−1000). The specimens were prepared for electron backscatter diffraction (EBSD) observation using the procedure reported in detail in Ref. [23], and the automated EBSD data collection was performed using a TSL camera attached to a dual beam FIB (FEI Helios NanoLab DualBeam system) at a voltage of 25 kV and a beam of 2.5 nA.

The dynamic compression experiments were used to investigate the mechanical properties of the pressed uranium by ECAP at different passes, and the sample of compression experiments was cut from the ECAPed uranium with a size of d4 mm × 4 mm. The dynamic compressive properties were tested by split Hopkinson pressure bar (SHPB), and the strain rates were set at the mean values of 2000 s^{-1}.

3 Results and discussion

3.1 Initial uranium

CLSM and EBSD images of the microstructure in original uranium are shown in Fig. 2. The original uranium mainly composed of coarse grains is observed, and the grain size with a large fluctuation from several hundreds micrometers to several millimeters is found. In a coarse grain, high density subgrains can be observed, and the measured misorientation between the sugrains is lower than 15°, belonging to low angle grain boundary. Besides the subgrians in coarse grained uranium, low density and coarse

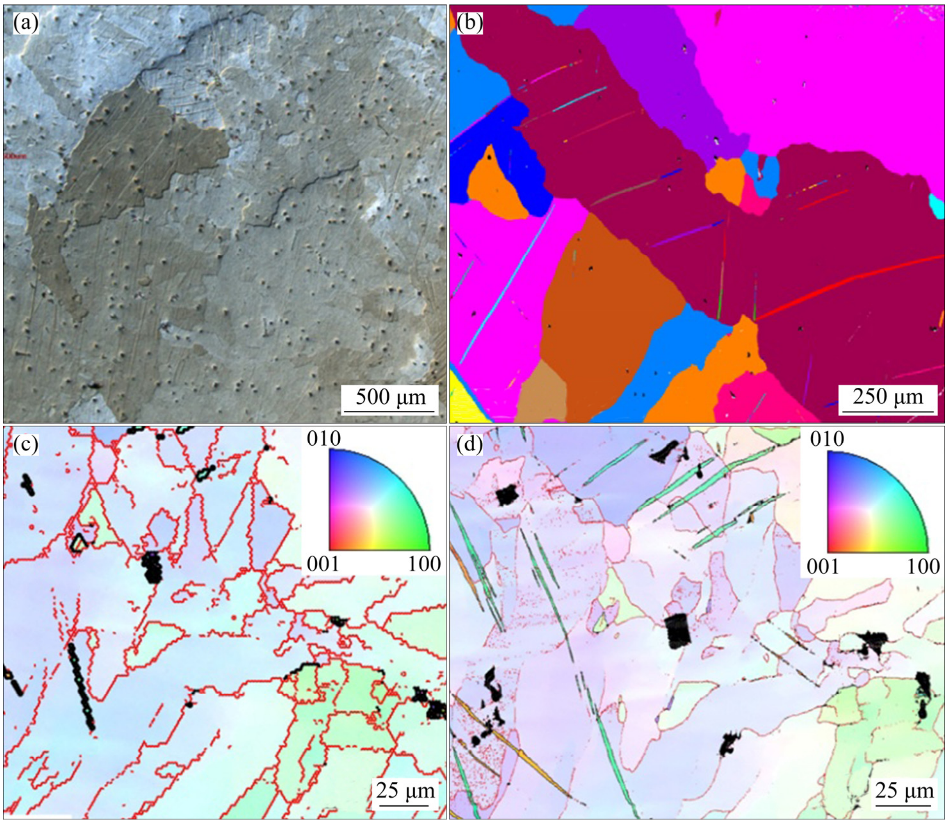

Fig. 2 Microstructures of original uranium sample: (a) CLSM image; (b) All Euler image; (c) Inverse pole map (black line: high angle boundary (>15°); red line: low angle boundary (2°−15°)); (d) Inverse pole map (twin laths)

primary twin laths are observed in undeformed uranium, and the main twin mode {130} is identified and confirmed.

3.2 Uranium pressed at 1 pass

CLSM and EBSD images of the microstructure in uranium sample pressed by ECAP at 1 pass and subsequent intermediate heat treatment are shown in Fig. 3. The pressed uranium samples show severe deformation and elongated grains, and high density deformation twins can be observed in the pressed grains. Then, the pressed samples are annealed at intermediate heat treatment, and the severe deformation and elongated grains in samples are transformed into fine and equiaxed grains, indicating that the recrystallization has occurred in heat treatment process. The melting point of uranium is 1123 °C, and the intermediate heat treatment temperature of uranium sample is 500 °C, which is about 0.55 times melting point, leading to recrystallization occurring to form the fine and equiaxed grains. The measured mean sizes of grains in uranium samples at 1 pass under intermediate heat treatment are about 25 μm. Compared with original samples, the grain size of uranium samples under ECAP and subsequent intermediate heat treatment is decreased about two orders of magnitude. Meanwhile, high density deformation twins and low angle boundary in pressed grains almost disappear, further indicating that the defect density decreases dramatically, and the recrystallization has occurred.

3.3 Uranium pressed at 2 passes

The microstructure of uranium sample pressed by ECAP at 2 passes and subsequent intermediate heat treatment is shown in Fig. 4. The fine and equiaxed grains prepared by ECAP at 1 pass and subsequent heat treatment transform into the deformed and extruded grains caused by ECAP at 2 passes, and the severe fragmentized crystal nucleus and medium density deformation twins can be observed in the pressed samples at 2 passes. After intermediate heat treatment, the severely deformed and extruded grains are evolved into fine and equiaxed grains, and the statistical grain size

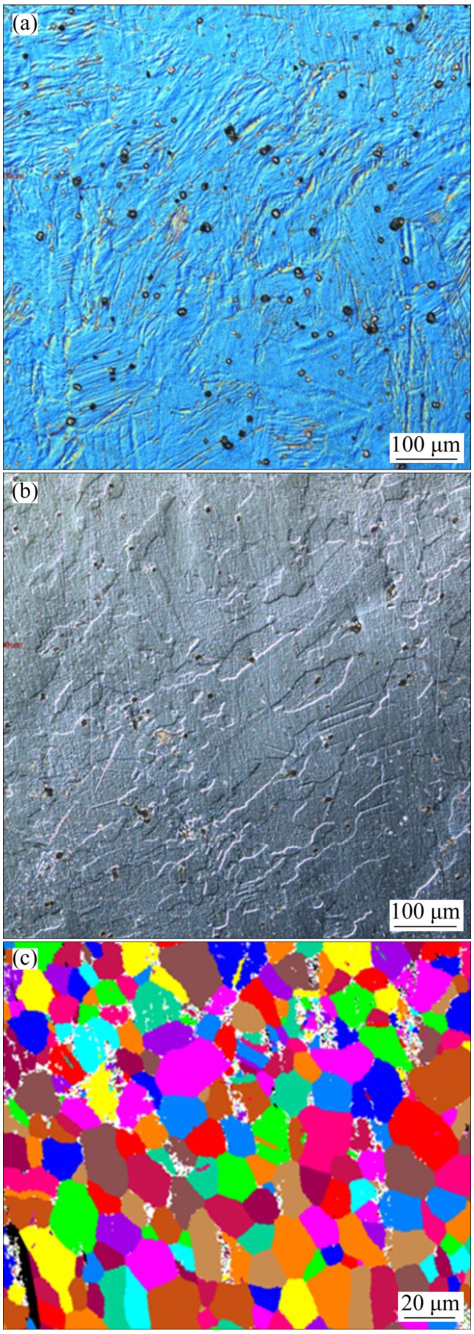

Fig. 3 Microstructures of uranium sample pressed by ECAP at 1 pass and subsequent intermediate heat treatment: (a) CLSM image of pressed sample; (b) CLSM image of annealed sample; (c) All Euler map of annealed sample

prepared by ECAP at 2 passes and heat treatment is 16 μm, which is much smaller than that of the grains formed by ECAP at 1 pass and heat treatment. The low angle boundary and deformation twins decrease dramatically in the grains formed by ECAP at 2 passes and heat treatment, indicating that the formation of fine and equiaxed grains with low defect density is caused by the recrystallization.

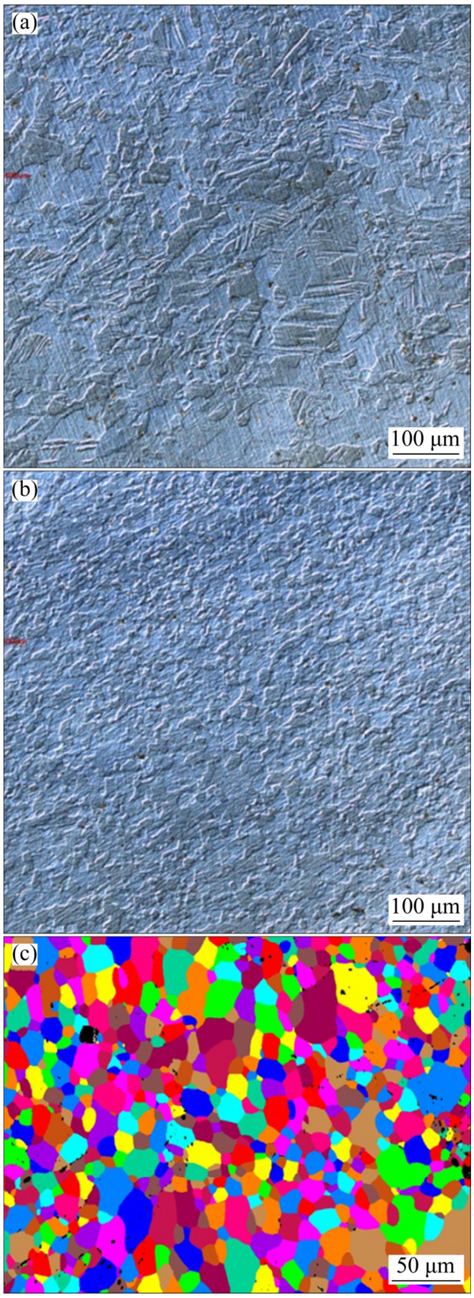

Fig. 4 Microstructures of uranium sample pressed by ECAP at 2 passes and subsequent intermediate heat treatment: (a) CLSM image of pressed sample; (b) CLSM image of annealed sample; (c) All Euler map of annealed sample

3.4 Uranium pressed at 3 passes

The microstructure of uranium sample pressed by ECAP at 3 passes and subsequent heat treatment is shown in Fig. 5. The fine and severe deformation crystal nucleus can be observed in samples pressed by ECAP at 3 passes, and low density deformation twins are found in localized fragmentized grains. The formation of low density deformation twins is associated with the fragmentized crystal nucleus

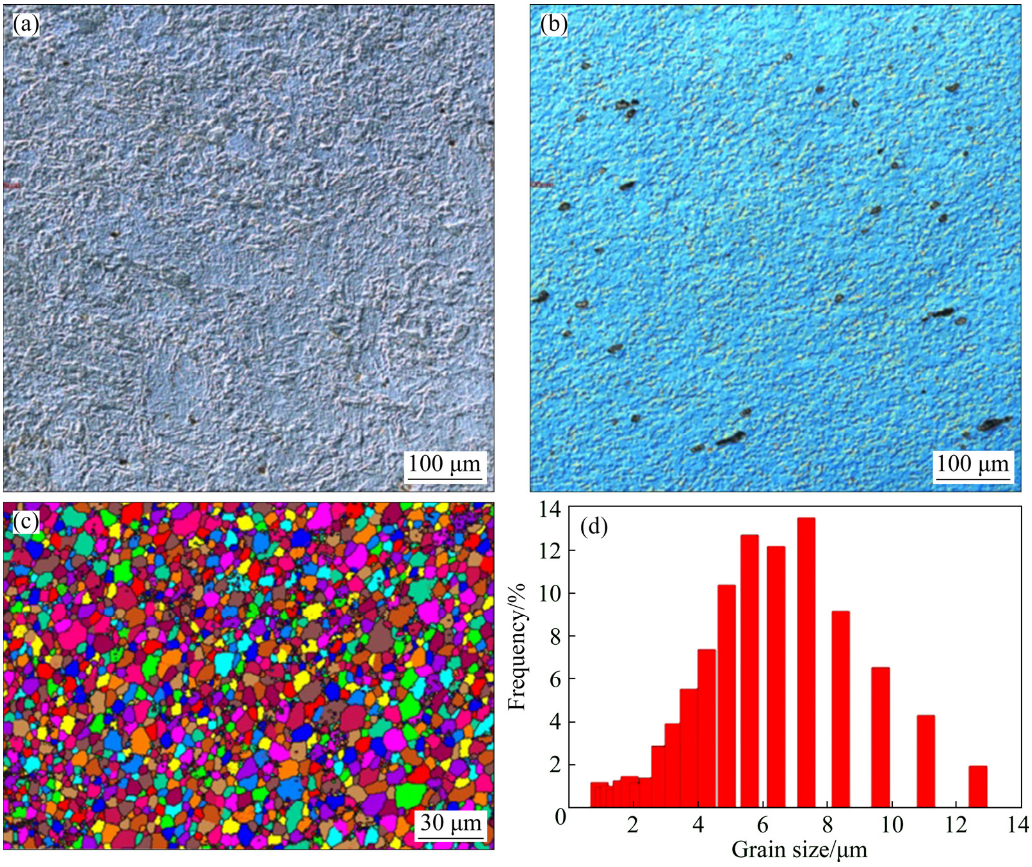

Fig. 5 Microstructures of uranium sample pressed by ECAP at 3 passes and subsequent intermediate heat treatment: (a) CLSM image of pressed sample; (b−d) CLSM image (b), all Euler map (c) and grain size distribution (d) of annealed sample

size. As the crystal nucleus size decreases, the critical shear stress for the formation of deformation twins increases, leading to difficult formation of deformation twins. Thus, the density of deformation twins in fragmentized crystal nucleus decreases with increasing ECAP passes. After the extruded samples are annealed at 500 °C for 15 min, the severe fragmentized crystal nucleus is evolved into fine and equiaxed grains. The distribution of fine and equiaxed grains becomes much more uniform, and the statistical mean grain size prepared by ECAP at 3 passes and subsequent heat treatment is 6.5 μm with a standard deviation ±3.5 μm.

3.5 Grain size−yield strength relationship

The calculated effective strain ε in the present ECAP die at each pass is 0.6, and the relationship between the statistical grain size and effective strain can be seen in Fig. 6. Except the experimental data of original sample, the grain size prepared by ECAP at different passes decreases linearly with the increasing effective strain, showing that the

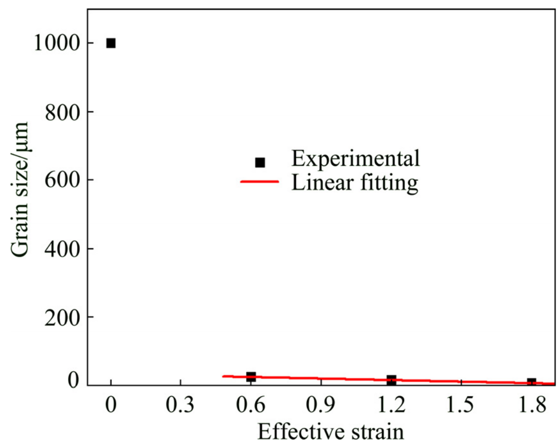

Fig. 6 Relationship between statistical grain size and effective strain

decreasing tendency of the grain size is linearly associated with the ECAP passes and annealing technology. Thus, ECAP passes assistant with annealing can be used to predict the uranium grain size in a certain scope.

The dynamic mechanical properties of

uranium sample prepared by ECAP at different passes and subsequent intermediate heat treatment are shown in Fig. 7. The yield strength of the coarse grained uranium calculated by linear fitting is 135 MPa deformed at a strain rate of 2000 s^{-1}, and the corresponding yield strengths of uranium samples prepared by ECAP at 1 pass, 2 and 3 passes and subsequent heat treatment are 285, 309 and 390 MPa, respectively, indicating that grain refinement leads to the yield strength increasing obviously. As reported in Ref. [24], the yield strength of materials is closely related to the grain size, due to the fact that grain refinement increases the number of grain boundaries, hindering the dislocation slipping and twin nucleation, and leads to the yield strength increasing.

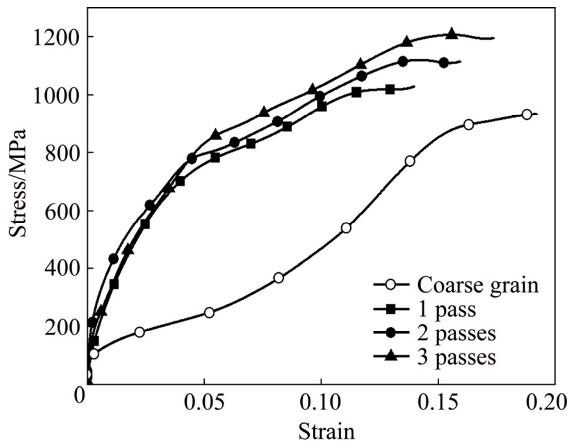

Fig. 7 Dynamic mechanical properties of uranium prepared by ECAP at different passes and subsequent heat treatment

The classical Hall−Petch relationship between the yield strength and the grain size of uranium sample is plotted in Fig. 8. It can be seen that the yield strength of uranium sample is linearly fitted with the grain size, showing that the classical Hall−Petch relationship can be used in uranium grain refinement. The fitting Hall−Petch relationship of uranium samples can described as follows:

$$\sigma_s=\sigma_0+kd^{-1/2} \tag{2}$$

where σ_s is the yield stress, σ_0 is equal to 125 MPa for uranium sample and often identified with “friction stress” needed to move individual dislocations during deformation, k is often referred to as Hall−Petch slope and material dependent and is equal to 708, and d is the average grain size. The correlation coefficient between the yield strength and grain size for uranium sample is 0.99. The well-known Hall−Petch relationship is derived from dislocation strengthening theory [24]. For coarse grained uranium, twinning is considered as the dominant plastic deformation mechanism, and dislocation slipping plays an assistant role [18]. Thus, the Hall−Petch relationship is applied limitedly in coarse grained uranium. With the grain size decreasing to tens of micrometers, the critical shear stress for the deformation twin formation increases obviously, and a few deformation twins are observed in uranium sample pressed by ECAP at 1 pass, 2 and 3 passes, indicating that the transformation of mainly plastic deformation mechanism takes place, twinning developing into dislocation slipping. Thus, the Hall−Petch relationship can be applied in uranium sample pressed by ECAP at different passes with grain size ranging from several micrometers to tens of micrometers.

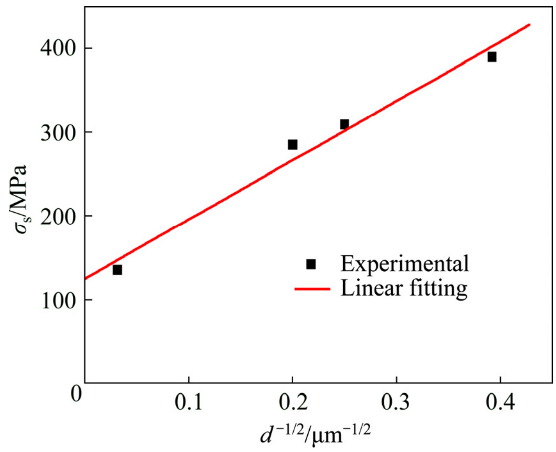

Fig. 8 Relationship between yield strength and grain size of uranium sample

4 Conclusions

(1) ECAP technique is an efficient way to produce fine grained uranium rods, and fine grained uranium rods with diameter of about 4 mm and length of about 10 mm were prepared by ECAP with a die channel angle of 120°.

(2) The coarse grained uranium samples are refined by ECAP at different passes and subsequent heat treatment, and the initial grains with diameter of about 1000 μm are refined into 25, 16 and 6.5 μm by ECAP at 1 pass, 2 and 3 passes, respectively.

(3) Grain refinement leads to the dramatic increase of dynamic yield strength of uranium samples, from initial 135 to 390 MPa for the sample prepared by ECAP at 3 passes.

(4) The yield strength and grain size of uranium sample can be described by classical Hall−Petch relationship, and the fitting Hall−Petch relationship of uranium samples is drawn as $\sigma_s=125+708d^{-1/2}$.

Acknowledgments

The authors wish to thank YIN Jia-qing for discussion, LIAO Yi-chuan for EBSD sample preparation, ZENG Rong-guang for microscope observation, and TANG Xian-e for metallographic assistance.

References

[1] LEI Wei-wei, WANG Hong-xia, LIANG Wei, HUANG Zhi-jun. Microstructure and mechanical properties of Fe-canned pure Mg after a single ECAP at room temperature [J]. Materials Science and Engineering A, 2017, 700: 163−167.

[2] LEWANDOWSKA M, CHROMINSKI W, LIPINSKA M, OLEJNIK L, ROSOCHOWSKI A. Incremental ECAP as a method to produce ultrafine grained aluminium plates [J]. Key Engineering Materials, 2016, 710: 59−64.

[3] CARDOSO K R, TRAVESSA D N, BOTTA W J, JORGE A M Jr. High strength AA7050 Al alloy processed by ECAP: Microstructure and mechanical properties [J]. Materials Science and Engineering A, 2011, 528: 5804−5811.

[4] DAMAVANDI E, NOUROUZI S, RABIEE S M, JAMAATI R. Effect of ECAP on microstructure and tensile properties of A390 aluminum alloy [J]. Transactions of Nonferrous Metals Society of China, 2019, 29: 931−940.

[5] LELAA M I A. 3D FEM simulations and experimental validation of plastic deformation of pure aluminum deformed by ECAP and combination of ECAP and direct extrusion [J]. Transactions of Nonferrous Metals Society of China, 2017, 27: 1338−1352.

[6] WEI W, WANG S L, WEI K X, ALEXANDROV I V, DU Q B, HU J. Microstructure and tensile properties of Cu−Al alloys processed by ECAP and rolling at cryogenic temperature [J]. Journal of Alloys and Compounds, 2016, 678: 506−510.

[7] ABIB K, BALANOS J A M, ALILI B, BRADAI D. On the microstructure and texture of Cu−Cr−Zr alloy after severe plastic deformation by ECAP [J]. Materials Characterization, 2016, 112: 252−258.

[8] VINCENTIS N S D, KLIAUGA A, FERRANTE M, AVALOS M, BROKMEIER H G, BOLMARO R E. Evaluation of microstructure anisotropy on room and medium temperature ECAP deformed F138 steel [J]. Materials Characterization, 2015, 107: 98−111.

[9] DOBATKIN S V, SKROTZKIC W, RYBALCHENKO O V, TERENT'EV V F, BELYAKOV A N, PROSVIMIN D V, ZOLOTAREV E V. Structural changes in metastable austenitic steel during equal channel angular pressing and subsequent cyclic deformation [J]. Materials Science and Engineering A, 2018, 723: 141−147.

[10] MINARIK P, VESELY J, KRA I R, BOHLEN J, KUBA S J, JANEC C E M, STRA S J. Exceptional mechanical properties of ultra-fine grain Mg−4Y−3RE alloy processed by ECAP [J]. Materials Science and Engineering A, 2017, 708: 193−198.

[11] POGGIALI F S J, SILVA C L P, PEREIRA P H R, FIGUEIREDO R B, CETLIN P R. Determination of mechanical anisotropy of magnesium processed by ECAP [J]. Journal of Materials Research and Technology, 2014, 3: 331−337.

[12] YANG Xi-rong, ZHAO Xi-cheng, FU Wen-jie. Deformed microstructures and mechanical properties of CP-Ti processed by multi-pass ECAP at room temperature [J]. Rare Metal Materials and Engineering, 2009, 38: 955−957.

[13] EFTEKHARI M, FARAJI G, NIKBAKHT S, RASHED R, SHARUFZADEH R, HILDYARD R, MOHAMMADPOUR M. Processing and characterization of nanostructured Grade 2 Ti processed by combination of warm isothermal ECAP and extrusion [J]. Materials Science and Engineering A, 2017, 703: 551−558.

[14] NASERI R, KADKHODAYAN M, SHARIATI M. Static mechanical properties and ductility of biomedical ultrafine-grained commercially pure titanium produced by ECAP process [J]. Transactions of Nonferrous Metals Society of China, 2017, 27: 1964−1975.

[15] LEE B S, KIM M H. Annealing behavior of Zr702 fabricated by ECAP [J]. Materials Science and Engineering A, 2006, 416: 150−154.

[16] LEE B S, KIM M H, HWANG S K, KWUN S I, CHAE S W. Grain refinement of commercially pure zirconium by ECAP and subsequent intermediate heat treatment [J]. Materials Science and Engineering A, 2007, 449−451: 1087−1089.

[17] ZOU Dong-li, GUO Ya-kun, SHUAI Mao-bing, XIAO Da-wu. Microstructure evolution of depleted uranium impacted by steel projectile at velocity of 50 m/s [J]. Transactions of Nonferrous Metals Society of China, 2017, 27: 1043−1051.

[18] FIELD R D, MCCABE R J, ALEXANDER D J, TETER D F. Deformation twinning and twinning related fracture in coarse-grained uranium [J]. Journal of Nuclear Materials, 2009, 392: 105−113.

[19] MCCABE R J, CAPOLUNGO L, MARSHALL P E, CADY C M, TOME C N. Deformation of wrought uranium: Experiments and modeling [J]. Acta Materialia, 2010, 58: 5447−5459.

[20] REN Zhi-yong, HU Gui-chao, MA Rong, WU Jun, WANG Zhen-hong, LUO Chao. Recrystallization and grain growth of U−5.5wt%Nb alloy [J]. Journal of Nuclear Materials, 2017, 494: 284−293.

[21] REN Zhi-yong, MA Rong, HU Gui-chao, WU Jun, WANG Zhen-hong, LUO Chao. Grain refinement and mechanical response of U−5.5wt%Nb alloy produced by cold rolling and heat treatment [J]. Journal of Nuclear Materials, 2017, 494:

72−78.

[22] GARLEA E, BRIDGES R L, GARLEA V O, CARPENTER D A, HEMPHILL M A, MORRELI J S. Characterization of a grain size refinement process in cast uranium [J]. Materials Science and Engineering A, 2013, 559: 210−216.

[23] CLARKE A J, FIELD R D, MCCABE R J, CADY C M, HACKENBERG R E, THOMA D J. EBSD and FIB/TEM examination of shape memory effect deformation structures in U−14at.%Nb [J]. Acta Materialia, 2008, 56: 2638−2648.

[24] KOIZUMI T, KURODA M. Grain size effects in aluminum processed by severe plastic deformation [J]. Materials Science and Engineering A, 2018, 710: 300−308.

等通道挤压及热处理制备细晶铀的显微组织及力学性能

邹东利，何立峰，肖大武，赵雅文，邱志聪，路 超，刘 帆

中国工程物理研究院 材料研究所，江油 621907

摘 要：采用等通道挤压及热处理相结合的技术制备细晶金属铀，采用激光共聚焦显微镜、电子背散射衍射、霍普金森压杆系统地研究细晶金属铀的显微组织及力学性能。结果表明，粗晶金属铀经 3 道次等通道挤压及后续热处理后晶粒尺寸由约 1000 μm 细化为 6.5 μm，相应的动态屈服强度由 135 MPa 增加至 390 MPa。对于等通道挤压铀样品，用经典 Hall−Petch 公式能描述晶粒尺寸与屈服强度之间的关系，同时获得等通道挤压技术制备细晶金属铀的 Hall−Petch 拟合关系。

关键词：贫铀；等通道挤压；热处理；晶粒细化；严重塑性变形

(Edited by Wei-ping CHEN)