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Strength and fracture mechanism of a magnesium alloy for medical applications after equal-channel angular pressing

G. V. Klevtsov[†,1], R. Z. Valiev[2], N. A. Klevtsova[1], M. V. Fesenyuk[3], O. B. Kulaysova[2], I. N. Pigaleva[1]

[†]Klevtsov11948@mail.ru

[1]Togliatti State University, Togliatti, 445020, Russia
[2]Ufa State Aviation Technical University, Ufa, 450008, Russia
[3]Joint Stock Company "Production Association "Strela", Orenburg, 460005, Russia

In this paper, we study the strength and fracture mechanism of the cast magnesium alloy Mg-Zn-Ca (1.0% Zn, 0.2% Ca) for medical applications after equal-channel angular pressing (ECAP) in comparison with the initial condition (after homogenization annealing). Mechanical tests included tensile and torsion tests of samples with a diameter of 3 mm and impact strength tests of samples with a V-shaped stress raiser. The microrelief of all fractures was studied in a Phenom ProX G6 scanning electron microscope (SEM). The results of tensile testing of the samples showed that the strength of the alloy after ECAP increased by a factor of 1.5–1.8, and the elongation increased by a factor 2.6. The torsional strength of the alloy increased by a factor 1.1–1.3, and the relative shear of the alloy decreased by factor a 1.3 as compared with the alloy after annealing. In contrast to static loads, the magnesium alloy Mg-Zn-Ca after ECAP is poorly resisting one-time impact loads. Analysis of the results of this study allows us to conclude that the magnesium alloy Mg-Zn-Ca after ECAP is more promising for the manufacture of medical products that experience static loads during operation, as compared with the alloy after annealing.

Keywords: magnesium alloy, equal-channel angular pressing, ultrafine-grained structure, mechanical properties, strength, fracture.

1. Introduction

The use of modern "preserving" operating technologies in dentistry, maxillofacial surgery and traumatology, that minimize trauma during surgery and shorten the period of postoperative rehabilitation, involves miniaturization of medical products, for example, various implants, plates for bone osteosynthesis, pins and screws for the fixation of plates and bone fragments. During operation the products may experience different types of stresses, both in terms of value and loading type, namely static and cyclic [1–5]. Therefore, the task of miniaturization of medical products cannot be solved without the use of materials having a high biocompatibility and a high set of mechanical properties under different types of loading [2, 7]. These requirements are fully satisfied by a new class of bulk nanostructured metallic materials produced by the severe plastic deformation (SPD) processing, in particular, by equal-channel angular pressing (ECAP) [7–12]. Numerous recent studies convincingly indicate that the processing of metallic materials by SPD, in addition to grain refinement, leads to a high density of defects in the crystal structure and other structural changes [7–10]. The hardness and strength of ECAP-processed materials increases, but ductility declines [2, 6, 7–13], which severely limits their use in many advanced technologies. However, several research groups have reported metallic nanomaterials with a good ductility and a high strength [8, 9]. Authors [14] presented a review of experimental and theoretical studies on a good combination of increased strength and ductility in metallic nanomaterials. Particular attention is paid to the main strategies for optimizing their strength and ductility, the use of fundamental deformation mechanisms, structural architecture at one or more length scales, texture, and chemical composition. The authors highlighted key points that are of particular interest for future research on the ductility of high-strength nanostructured metals and alloys [14]. In addition to tensile testing, materials intended for the manufacture of implants are subjected to other types of loading, both static and dynamic. A positive effect of ECAP processing on the torsion resistance of such materials as titanium, austenitic steel was noted [6,13,15]. However, ECAP processing has an ambiguous effect on other characteristics of materials, such as fatigue strength, impact strength, and static crack resistance. This fully applies to magnesium alloys, which are widely used in dentistry, maxillofacial surgery, traumatology and other areas of medicine [13–21]. Therefore, the issue of the effect of ECAP processing on the fracture resistance of a magnesium alloy under various types of loading remains relevant.

An equally important aspect at this stage is the study of the fracture mechanisms of materials after ECAP at various scale levels [22]. So far, such studies are, as it were, background studies in the evaluation of mechanical properties. However, it is obvious that without studying the fracture mechanisms it is impossible to understand the physical nature of the strength and ductility of this class of materials. Study of the fracture process using a set of physical methods, the most important of which is scanning electron microscopy, provides ample opportunities in this area.

The aim of the present study is to evaluate the strength and fracture mechanisms under static (tensile, torsional) and impact loading of a magnesium alloy for medical applications after ECAP in comparison with the alloy in the initial condition.

2. Materials and methods

We selected the magnesium alloy Mg-Zn-Ca (1.0% Zn, 0.2% Ca), widely used in medicine, as the material to be studied. The alloy in the initial condition was investigated after homogenization annealing of cast billets at a temperature of 450°C, 24 hours, cooling in water. Then the homogenized alloy was subjected to ECAP via the following regime [7]: route Bc, $\varphi = 120°$, $n = 2$ at a temperature of 430°C + $n = 1$ at a temperature of 400°C + $n = 1$ at 350°C.

The structure of the alloy was studied using a JEM-6390 scanning electron microscope (SEM). The fine structure of the UFG alloy was studied using a JEM-2100 transmission electron microscope (TEM). To measure the grain size, the method of "Arbitrary secants" was used. The static tension test of the cylindrical specimens with a diameter of 3 mm was carried out at a temperature of 20°C using an H50KT universal testing machine (Tinius Olsen, Redhill, UK). The tension speed was 5 mm/min. The torsion test of specimens with a diameter of 3 mm was carried out using a KTS-405-20-0.5 testing machine. The "torque - angle of twist" diagrams were recorded, and according to their results the tensile mechanical properties of the alloy were calculated [23]. The impact toughness tests (KCV) of the specimens 10×10×55 mm in size with a V-shaped stress raiser were performed over a wide temperature range using a TSKM-50 pendulum impact testing machine. The fracture microrelief was examined using a Phenom ProX G6 scanning electron microscope (SEM).

3. Results

In the initial condition (after annealing), the structure of magnesium alloy Mg-Zn-Ca samples consists of α-Mg grains with a mean size of 415 μm (Fig. 1a). Particles (presumably $Ca_2Mg_6Zn_3$) with a size of up to 1 μm were found inside the grains; the size of these particles at the boundary of the grains was equal to 4 μm. The volume fraction of the particles at the grain boundaries was 1%. After homogenization, the mean grain size was large, and the alloy under investigation had an HCP lattice, resulting in the material's low ductility in the homogenized condition. The aim of the ECAP processing was structure refinement, therefore the pressing temperature of 430°C was selected empirically. We were able to implement 2 ECAP passes at this temperature without fracture of the billets. ECAP processing for 2 passes at a deformation temperature of 430°C produced a structure with a mean grain size of 80 μm, which is an order of magnitude smaller than that in the homogenized condition. However, this ECAP-processed condition did not provide a satisfactory level of properties, therefore, further processing was performed at lower temperatures (1 pass at 350°C and 1 pass at 300°C). The approach of grain structure refinement, in HPC metals

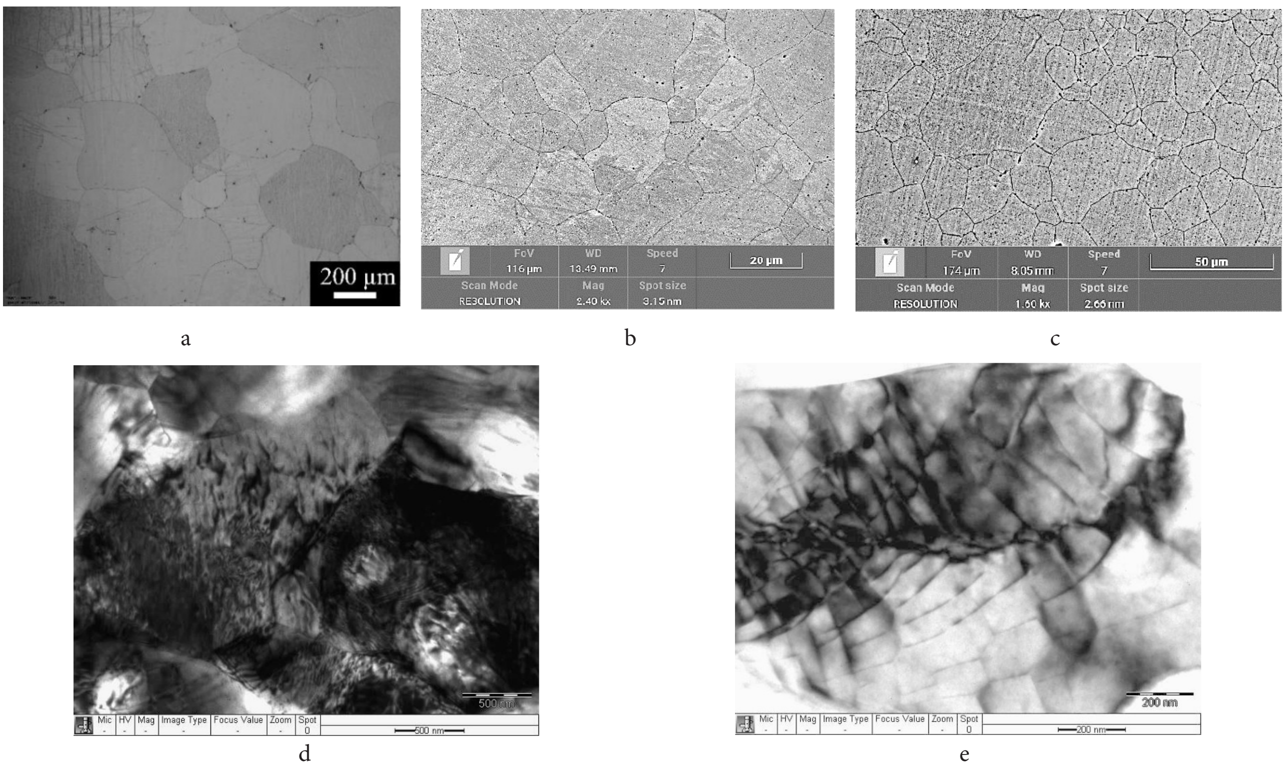

Fig. 1. Microstructure (SEM) of the magnesium alloy Mg-Zn-Ca after annealing (a) and after ECAP: longitudinal direction (b), transverse direction (c); dislocation structure inside the grains (d, e) (TEM).

as an example, through plastic deformation with a gradual temperature decrease had been previously applied many times [24,25]. In the ECAP-processed magnesium alloy, a bimodal structure was formed. The structure also contains large grains with a size of 35–40 μm and small grains with a size of 5 μm (Fig. 1b,c). A high density of dislocations inside the grains is observed (Fig. 1d,e). This structure provides both higher tensile strength properties of the alloy and a higher ductility (Table 1).

The results of the torsion tests of the specimens showed that the torque of the magnesium alloy after ECAP via the above regime increased from 13.8 to 15 N·m; the angle of twist decreased from 210 to 140 degrees (Fig. 2) as compared to the homogenized condition of the alloy.

The mechanical properties of the alloy under torsion calculated from the "torque-angle of twist" diagram (Fig. 2) showed that, after ECAP, the torsional strength and torsional yield strength increased, respectively, by factors of 1.1 and 1.3, and the relative shear decreased by a factor of 1.3, as compared with the alloy after annealing (Table 2).

On the surface of the obtained fractures of the magnesium alloy in the initial condition (Fig. 3a), the central part, which has a high roughness, as well as the transitional and peripheral parts, which have a smoothed macrorelief, are clearly visible (Fig. 3a). Microfractographic analysis showed that in the central part of the sample, fracture occurred by delaminating along the slip planes; visible areas of relief collapse from contact with the response surface of the fracture (Fig. 3b). The microrelief in the transitional and peripheral parts of the fracture is almost completely worn out (Fig. 3c,d) as a result of a mutual friction between the mating fracture surfaces. The fracture surface of the magnesium alloy after ECAP is uniform and rougher (Fig. 3e) than in the previous case. In the central region, fracture of the sample occurred according to the quasi-cleavage mechanism (Fig. 3f). In the transitional part of the fractures, the microrelief is partially worn out (Fig. 3g), and in the peripheral part it is almost completely worn out (Fig. 3h).

The fracture surface microrelief reflects the process of specimen fracture during torsion. Fracture under torsional stresses starts from the peripheral region. During the further torsion of the specimen the microrelief in the peripheral and transitional parts of the fracture becomes worn out as a result of a mutual friction between the mating fracture surfaces

Table 2. Mechanical properties of the magnesium alloy Mg-Zn-Ca.

Condition	τ_k, MPa	$\tau_{0.3}$, MPa	g, %
After annealing	264±9	102±5	33±0.3
After ECAP	283±15	132±10	25±0.7

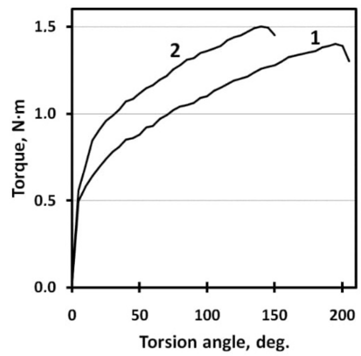

Fig. 2. "Torque-angle of twist" diagram based on the torsion tests of the magnesium alloy samples after annealing (1) and after ECAP (2).

Table 1. Mean grain size and tensile mechanical properties of the magnesium alloy Mg-Zn-Ca.

Condition	d_{mean}, μm	$\sigma_в$, MPa	$\sigma_{0.2}$, MPa	δ, %
After homogenization	$d_{mean}=415$	119±9	65±5	9±0.3
After ECAP	Bimodal structure $d_{large\ grains}=40$ $d_{small\ grains}=5$	210±10	97±7	23±0.5

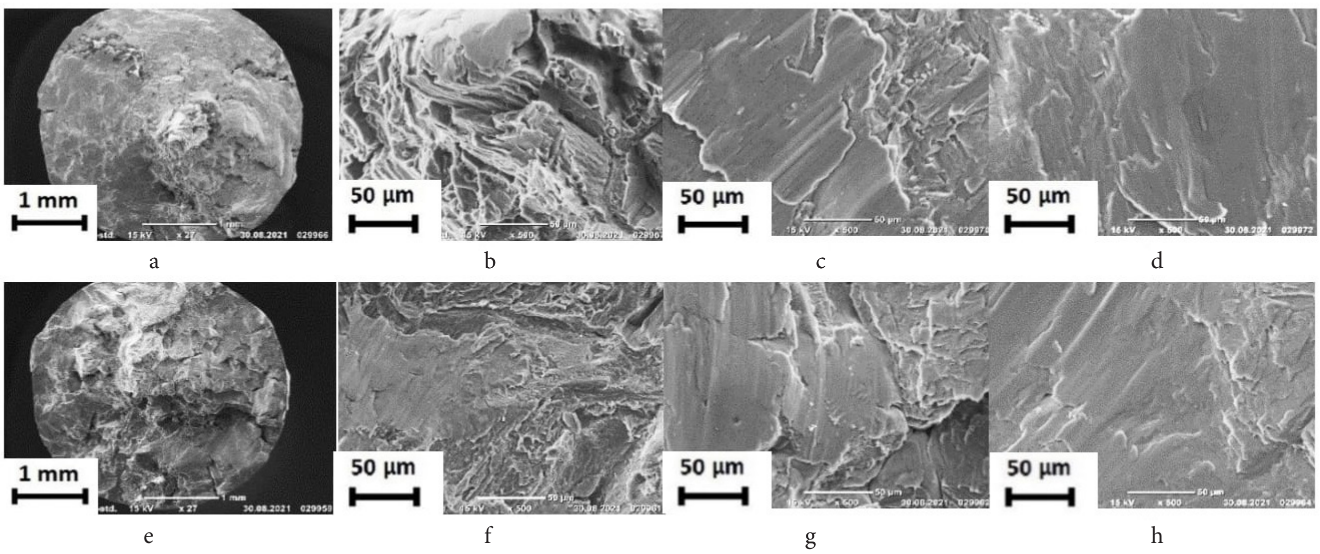

Fig. 3. Typical view (a, e) and microrelief (b–d, f–h) of the fractures of the specimens for torsion with a diameter of 3 mm from the magnesium alloy Mg-Zn-Ca after annealing (a–d) and after ECAP (e–h) in the central part of the fractures (b, f), the transitional part (c, g), and the peripheral part (d, h). ×27 (a, e); ×500 (b–d, f–h).

under normal rupture stresses. In the central part, fracture occurred under normal rupture stresses as evidenced by a fully or partially preserved microrelief.

The results of the impact toughness tests of the samples showed (Fig. 4) that the magnesium alloy after ECAP at all test temperatures under study has lower values of impact toughness (KCV) as compared to the alloy in the initial condition. Moreover, the impact toughness of the alloy in the initial condition increases with increasing test temperature, and the impact toughness of the alloy after ECAP remains almost constant in the temperature range from −196°C to 200°C (Fig. 4).

All of the produced fractures of the magnesium alloy had a homogeneous, fine-grained structure. The fracture surfaces of the alloy after ECAP have a higher surface roughness, as compared to the alloy in the initial condition, at all test temperatures. The magnesium alloy in the initial state is destroyed by a quasi-cleavage with the formation of areas separated by secondary cracks at the temperature of −196°C (Fig. 5a). The delamination along the slip planes is a dominant mechanism of impact fracture of the magnesium alloy at higher test temperatures (Fig. 5b, c). Viscous ridges are visible along the grain boundaries at the test temperature of 300°C (Fig. 5c). Impact fracture of the alloy after ECAP at all test temperatures occurred according to the quasi-cleavage mechanism with the formation of secondary cracks (Fig. 5d–f). Secondary cracks are visible on the fracture surface at low and room test temperatures (Fig. 5d, e). Areas similar in structure to dimples appear on the fracture surface at the test temperature of 300°C (Fig. 5f). It is possible that the reason for the appearance of dimpled areas is associated with crystallization processes in the alloy at a given temperature. The fracture of the magnesium alloy after ECAP by the quasi-cleavage mechanism predetermined a constant and low level of impact toughness at all test temperatures. It is known [13,17] that the quasi-cleavage fracture of magnesium alloys indicates a low value of impact toughness.

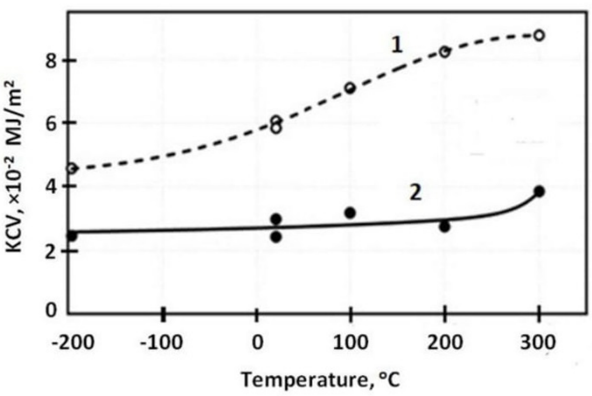

Fig. 4. Temperature dependence of the impact toughness (KCV) of the magnesium alloy Mg-Zn-Ca in the initial condition (1) and after ECAP (2).

4. Discussion

As noted above, in the process of operation, medical devices (plates, screws, pins, etc.) experience loads large in value and various in type. Therefore, when selecting a material for their manufacture, it is necessary to take into account a whole range of mechanical properties under various types of loading. In this regard, it is interesting to compare a set of mechanical properties of the magnesium alloy Mg-Zn-Ca, widely applied in medicine, in the ECAP-processed condition with the respective properties of the alloy in the initial condition.

As shown above, after ECAP processing the alloy's tensile strength increases by a factor of 1.5–1.8, and ductility increases by a factor of 2.6. This, of course, is a positive factor in relation to medical devices, for example, plates for bone osteosynthesis which are often deformed during operation when adjusting to the individual characteristics of patients.

Analysis of the operational damages of medical devices shows [3, 5] that a large percentage of screws for the fixation of plates and bone fragments in maxillofacial surgery and

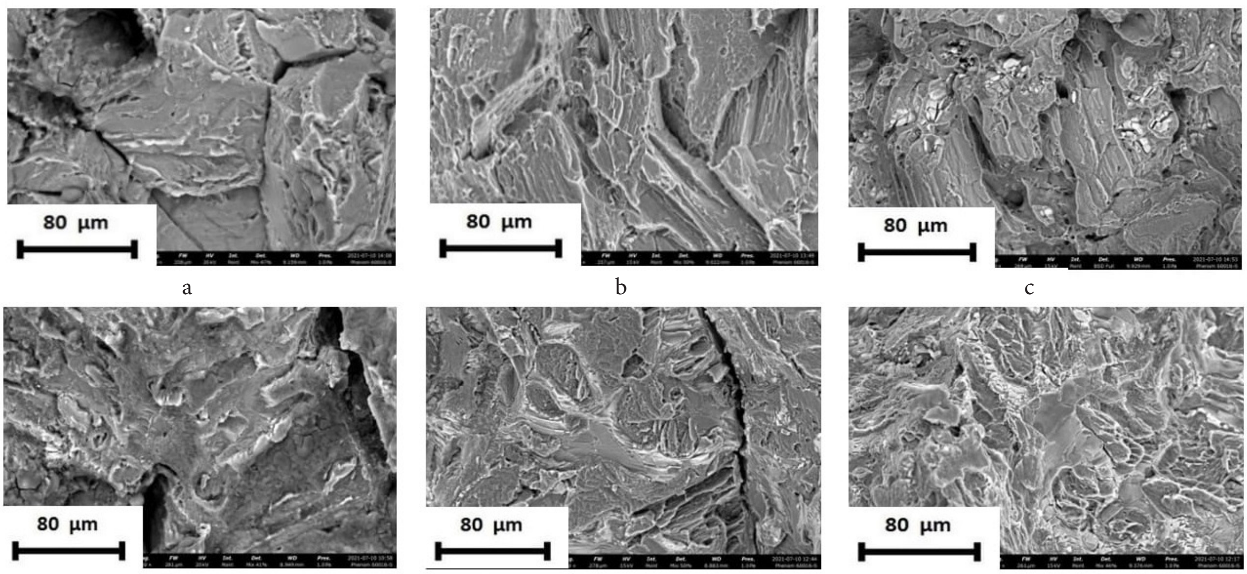

Fig. 5. Impact fracture surface microrelief of the central part of the magnesium alloy Mg-Zn-Ca in the initial condition (a–c) and after ECAP (d–f). The fracture surfaces of specimens tested at temperatures of −196°C (a, d), 20°C (b, e), 300°C (c, f). ×2000 (a–c, e); ×1850 (g, d).

traumatology are destroyed by twisting in the smooth area of the screw between the threaded part and the head. The bone fragments fixed by the plate with screws are not subjected to a high load, because the plate with screws takes care of this function. Therefore, in areas of the bone in which there is no load bearing, bone resorption develops, which often leads to pathological fractures. The longer the stay of the fixed plate, the deeper degenerative-destructive processes develop in the bone. Therefore, the plate with screws should be removed shortly after the fusion of fragments before the development of secondary changes in the bone. However, by this time, as a rule, the bone has already fused with the threaded part of the screw, which makes it difficult to unscrew the screw. Since the design of bone screws assumes that a torsional force is applied to the screw head when they are loosened, the maximum torsional stresses occur in a local area at the border between the head and neck of the screw. When unscrewing the screws where the bone has fused with the threaded part of the screw, an additional load is required, which increases the maximum torsional stresses in the local area at the border between the screw head and neck. The number of destroyed screws during an attempt to unscrew them can reach 30–60%, depending on the degree of fusion of the bone with the threaded part of the screw. The subsequent removal of the screw part remaining in the bone is associated with additional trauma to the patient. The torsional tests of the specimens from the magnesium alloy Mg-Zn-Ca showed that, after ECAP, the tensile strength in torsion and the yield strength in torsion increased by factors of 1.1 and 1.3, respectively. The relative shear decreased by a factor of 1.3 as compared to the alloy after annealing. The increased strength properties of the magnesium alloy are a favorable factor that increases the fracture resistance of bone screws in service, and decreased relative shear can also serve as a favorable factor that reduces the probability of fracture of screws fused to bone when they are unscrewed.

In contrast to static loads, the magnesium alloy Mg-Zn-Ca is poorly resistant to single-impact loads after ECAP. The impact toughness tests (KCV) showed that at 20°C the KCV of the alloy after ECAP is lower than that after annealing ($2.3.10^{-2}$ MJ/m^2 vs. $6.0.10^{-2}$ MJ/m^2). Consequently, although medical implants are not designed for impact loading, single dynamic loading should be avoided if individual fitting of the implants is necessary prior to surgery.

Summarizing the above, we can conclude that magnesium alloy Mg-Zn-Ca after ECAP is a promising material for the manufacture of medical products, which experience various static loads during operation.

5. Conclusions

1. The equal-channel angular pressing (ECAP) of the magnesium alloy Mg-Zn-Ca for medical purposes via the regimes described in this article, due to grain refinement and a high density of crystalline structure defects, increases the tensile strength properties of the alloy by a factor of 1.5–1.8 times and ductility by a factor of 2.6 times in comparison with the annealed condition.

2. The tensile strength and yield strength of the magnesium alloy after ECAP increased by factors of 1.1 and 1.3, respectively, and the relative shear decreased by a factor of 1.3, as compared to the alloy after annealing.

3. The magnesium alloy after ECAP has lower values of impact toughness (KCV) at all test temperatures as compared to the annealed alloy.

4. Thus, the magnesium alloy Mg-Zn-Ca after ECAP is a promising material for the manufacture of medical products that experience various static loads during operation. Having higher tensile and torsional strength properties, this alloy processed by ECAP can provide the possibility of miniaturization of medical products while maintaining the necessary strength characteristics that are currently required from the material.

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