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Проведено порівняльний аналіз існуючих матеріалів для виготовлення імплантатів, представлені їх фізико-механічні властивості, висвітлені переваги і недоліки. Показано, що магнієві сплави є одними з найбільш перспективних біорозчинних матеріалів. Вони біоінертні та біосумісні, але їх використання в остеосинтезі обмежене в основному їх недостатніми механічними властивостями через високу швидкість біодеградації, що вимагає їх поліпшення за рахунок зміни хімічного складу сплаву.

Для розробки нового біорозчинного сплаву на основі магнію, підібрані системи легування, що найкраще відповідають встановленим критеріям.

За допомогою методів планування експерименту вивчено окремий і спільний вплив цирконію, неодиму та цинку на структуроутворення і механічні властивості магнієвого сплаву. Побудовано математичні моделі, що описують вплив легуючих елементів, що досліджуються, на механічні властивості металу. З використанням отриманих рівнянь регресії проведена оптимізація хімічного складу магнієвого сплаву.

Проведено промислову і доклінічну апробації імплантатів з розробленого біорозчинного сплаву. Експерименти на тваринах підтвердили відсутність токсичного впливу продуктів деградації розробленого магнієвого сплаву на живий організм. Дослідження впливу розробленого сплаву на репаративний остегенез в експерименті на кролях показало позитивну динаміку відновлення кісткової тканини без помітних змін в її структурі, що забезпечує надійне зрощування елементів кісток при остеосинтезі.

Встановлено, що імплантати, виготовлені з розробленого сплаву, мають необхідний рівень механічних властивостей, відповідний механічним властивостям кісткової тканини. При цьому, вони нетоксичні і забезпечують надійне зрощення кісткової тканини до повної консолідації перелому. Позитивні результати проведених експериментів дозволяють зробити сприятливий прогноз про можливість застосування імплантатів з розробленого біорозчинного сплаву системи Mg–Zr–Nd у людини

Ключові слова: легувальні елементи, планування експерименту, границя міцності, відносне видовження, хімічний склад, оптимізація

DESIGN AND EXAMINATION OF THE NEW BIOSOLUBLE CASTING ALLOY OF THE SYSTEM Mg–Zr–Nd FOR OSTEOSYNTHESIS

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1. Introduction

A large number of injuries are reported globally every year. Among the injuries of musculoskeletal system, up to 25 % account for open fractures. Treatment of fractures that do not heal without surgical fastening the pieces is performed by using implants made of different materials in the form of various sophisticated structures (pins, needles,

plates, etc.). During operation, such implants are exposed to static and dynamic loads, as well as the effect of biocorrosion. In this regard, materials for implants must be biologically inert and compatible, as well as possess sufficient physical-mechanical properties (tensile strength, modulus of elasticity, and relative elongation). These characteristics should not be lower than the respective physical-mechanical properties of bone tissue.

Upon using, conventional implants are to be removed, as they are foreign bodies and increase the risk of local inflammation. Furthermore, they inhibit the regeneration processes in the body and further treatment. In order to avoid such negative consequences, surgeries are carried out aimed at removing the implants. These surgeries are rather expensive and do not rule out the risk of repeated surgery.

Recently, such implants have been widely used that are able to dissolve when exposed to the internal medium in the body, which makes it possible to eliminate the need for a repeated surgery. However, the insufficient mechanical properties and the rate of biodegradation of those materials that are used for biosoluble implants have necessitated the search for the most suitable material, which remains a relevant task.

2. Literature review and problem statement

The first and most widely used materials for the manufacture of implants were various metals and alloys on their base. These include stainless steels, titanium and its alloys, cobalt-based alloys, zirconium and tantalum [1]. The listed materials generally have good corrosion resistance, high strength and modulus of elasticity (Table 1) [2, 3]. Thus, they can withstand the stresses that implants receive during their application. However, some of these materials, such as stainless steels, cobalt alloys, and, partially, titanium alloys have shortcomings in terms of biocompatibility as they contain highly toxic alloying elements (chromium, molybdenum, and nickel). The long presence of such implants in humans leads to the accumulation of these toxic elements, thereby leading to metallosis [4, 5]. In addition, the high cost and scarcity of pure tantalum and zirconium also limit their application.

Table 1

Properties of various metals and alloys used in the manufacture of implants

Material	Tensile strength, MPa	Modulus of elasticity, GPa	Relative elongation, %
Stainless steels	200	530–1000	20–45
Pure titanium	102	200–550	15–24
Titanium alloys	105–115	750–1,100	10–20
Cobalt alloys	235–240	600–1,793	8–50
Pure zirconium	94.5	330	32
Pure tantalum	186	285–650	5–30
Human bone tissue	30–150	3–20	1.4–3.1

It is worth noting that the mechanical properties of metallic implants are significantly higher than those of the human bone tissue (σ_B=30–150 MPa, E=3–20 GPa, δ= =1.4–3.1 %) (Table 1). This often leads to the effect of "stress shielding" when an implant receives a large part of the

applied load while the bone perceives lower loading, which reduces its density after healing [5].

Biosoluble polymers are a good alternative to metals. In addition to their capability to dissolve in the human body, they also possess excellent biocompatibility, do not cause significant inflammation or toxic reactions, and have lower mechanical properties that prevents the "stress shielding".

Polyglycolic (PGA) and polylactic (PDLA and PLLA) acids and copolymers (PLGA) have become the most widely used biosoluble polymeric materials. These polymers have been approved by the US Food and Drug Administration and do not cause significant inflammation or toxic reactions in humans [6].

These polymers differ mainly by their biodegradation rate. PGA tends to degrade rapidly, but loses its mechanical strength *in vivo* after about 4–7 weeks after implantation [7]. Poly-L-lactide (PLLA) is resistant to hydrolysis due to its high crystallinity and hydrophobicity that predetermines a lower rate of biodegradation. Biodegradation with a complete loss of mechanical properties does not occur *in vitro* within the first 2 years after implantation [8], while the entire period of biodegradation takes up to 5 years [9, 10]. Poly-D-lactide (PDLA) has a lower degree of crystallinity, less resistant to hydrolysis and its strength is lower than that of PLLA [8, 10]. Implants made from PLGA dissolve after about 18 months [11, 12]. The disadvantages associated with PLGA include their synthetic origin and a decrease in pH level at the site of implantation, which leads to sterile sepsis [13].

The polymers of hydroxy derivatives of alkanoic acids (PHA) may also be used at osteosynthesis. Their natural origin provides for the capability to biodegrade and excellent biocompatibility. The PHA biodegradation rate is lower than that of most other polymers, except for PLLA [13].

A general disadvantage of pure polymers is their low and unstable mechanical strength [14]. That renders them unsuitable for the production of bone implants exposed to loads (Table 2) [3, 15, 16].

Table 2

Properties of various polymers used for the manufacture of implants

Material	Tensile strength, MPa	Young modulus, GPa	δ, %
PLA	49.6	3.6	2.4
PDLLA	54	1.5	16
PLLA	28–57	1.2–2.7	23
PGA	55–70	6.5–6.9	1.0
PLGA	41.4–55.2	1.4–2.8	–
PHA	20–43	2.5	5–20

Calcium phosphate ceramics such as natural hydroxyapatite $Ca_5(PO_4)_3OH$ and artificial tricalcium phosphate $Ca_3(PO_4)_2$ can also be used at osteosynthesis [17]. These types of ceramics are similar in their mechanical properties.

Hydroxyapatite (HAP) has excellent biocompatibility (the material is nearly identical to the natural bone material), high biological activity, high osteoconductive characteristics, non-toxic, it does not cause inflammatory reactions and has the capability to bioresorb [18, 19]. Thus, HAP is used in the manufacture of polymer-based composite materials such as PLLA-HA (based on poly-L-Lactide) and POC-HA (based on poly-1.8-octanediol citrate) (Table 3), in order to increase their biocompatibility and the rate of

biodegradation [20]. However, the mechanical properties of such composites are still insufficient to create implants that would perceive loads [21, 22].

Table 3

Mechanical and physical properties of polymer-based composites (POC-HA and PLA-HA)

Characteristic	Value	
	POC-HA	PLA-HA
Tensile strength, MPa	21.4–334.8	35–154.1
Elasticity modulus, GPa	0.023–0.027	1.1–2.6

Note: large spread of values is due to different amount of HAP

Magnesium is a very promising biosoluble material. Its advantage over polymers and ceramics is the higher mechanical properties that are very close to the properties of bone tissue (Table 4) [23, 24]. Additionally, magnesium and its corrosion products have excellent biocompatibility [25].

Table 4

Comparison of mechanical properties of magnesium and human bone tissue

Material	Elasticity module, GPa	Tensile strength, MPa	Relative elongation, %	Density, g/cm^3
Bone tissue	3–20	30–150	1.4–3.1	1.8–2.1
Pure magnesium	41–45	113	2–3	1.74

Despite its advantages, pure magnesium is very fragile and has a lack of tensile strength [26]. In addition, the degradation of magnesium alloys always leads to the release of hydrogen bubbles that can accumulate in the gas pockets near the implant that slows the growth of bone tissue and may lead to necrosis [27, 28]. Biological corrosion of magnesium could lead to an increase in the pH of the body that may cause alkaline poisoning [26, 28].

The listed shortcomings hamper the use of implants made from magnesium. It is possible to improve their physical-mechanical properties by additional doping and the development of an optimal chemical composition for the new alloy.

3. The aim and objectives of the study

The aim of this study is to design and optimize the mechanical properties of magnesium alloy for the manufacture of biosoluble implants, and to perform its industrial and pre-clinical testing.

To accomplish the aim, the following tasks have been set:
– to choose the most suitable system of doping in accordance with the established criteria and to study the effect of the selected alloying elements on the structure formation and the mechanical properties of the alloy;
– to construct mathematical models that would describe the influence of the examined alloying elements on the mechanical properties of the alloy;
– to optimize chemical composition of the alloy by employing the derived dependences;
– to perform industrial and preclinical approbation of implants made from the designed biosoluble alloy.

4. Materials and methods to study the influence of chemical composition on the mechanical properties and the rate of biodegradation of magnesium alloy

The alloy was melted in the crucible induction furnace IPM-500 with the rated capacity of 0.5 tons, power of 140 kW, performance of 230 kg/h, as well as in the gas distributing furnace with the rated capacity of 150 kg, in line with a standard technology. The melt was refined with the flux VI-2 in the distributing furnace; batches of metal were discharged using a bucket, the growing additives of ligatures containing Zr, Nd, Zn were injected, nest we poured standard samples to a sand-clay mold for mechanical tests. The aim was to obtain samples with a different content of alloying elements (Table 5) according to the chosen experiment design (Table 6). Influence of the alloying elements was studied in the following range: 0.4–1.5 % Zr; 2.2–3.4 % Nd; 0.1–0.7 % Zn.

Table 5

Chemical composition of samples

Sample No.	Content Zr, %	Content Nd, %	Content Zn, %
1	0.4	2.2	0.1
2	1.5	2.2	0.1
3	0.4	3.4	0.1
4	1.5	3.4	0.1
5	0.4	2.2	0.7
6	1.5	2.2	0.7
7	0.4	3.4	0.7
8	1.5	3.4	0.7
9	0.95	2.8	0.4
10	0.95	2.8	0.4
11	0.95	2.8	0.4

Table 6

Matrix of experiment design 2^3

Experiment No.	X_1	X_2	X_3
1	−1	−1	−1
2	1	−1	−1
3	−1	1	−1
4	1	1	−1
5	−1	−1	1
6	1	−1	1
7	−1	1	1
8	1	1	1
9	0	0	0
10	0	0	0
11	0	0	0

All samples were exposed to heat treatment using the furnaces Bellevue and PAP-4M, in accordance with the regime T6: hardening from 540±5 °C, over 8 hours, air cooling, and aging at 200±5 °C, over 3 hours, cooled in the air. Tensile strength (σ_B) and relative elongation (δ) of the alloy samples were determined using the universal test machine INSTRUN 2801. The tests were conducted in accordance with acting standards before and after aging in gelofusin (artificial blood substitute) during different periods of time.

Mathematical processing of results from mechanical tests was carried out in accordance with the standard method of experiment design [29].

5. Results of studying the mechanical properties and microstructure of magnesium alloy, industrial and preclinical tests

5. 1. Selecting the alloying system and analysis into the influence of elements on mechanical properties of magnesium alloy

The selection criteria for the alloy's alloying elements are: the capacity to form complex doped solid solutions and intermetallide compounds during thermal processing; the absence of toxic effect on the living organism.

The capability of alloying elements to form solid solutions is determined by the proximity of their atomic radii, which, in accordance with the Hume-Rothery rule, should differ by not more than 15 % [30]. If this ratio is larger, the binding energy of the solvent atoms and alloying elements is reduced, and, as a consequence of the curvature of the crystal lattice, solubility of alloying elements is decreased. Another important condition for the solubility of an element in the base metal, according to [31], is the difference in the electronegativities of elements that should not exceed 0.2...0.4.

Formation of intermetallide compounds and their properties depend on the electron structure of interacting elements included in the composition of the alloy. Such elements as zirconium and rare earth metals (REM) with incomplete d-shells form the intermediate phases with magnesium, and their solubility in the solid magnesium is low.

In accordance with the specified criteria, among known systems of alloying, the most suitable are Mg–Zn–Zr and Mg–Nd–Zr. Their mechanical characteristics are approximately at the same level, but the rate of biodegradation of alloys in the system Mg–Nd–Zr is lower, therefore, it is more acceptable. Typical representatives of alloys in a given system are ML10, its analog NZ30K, as well as WE-43 (Table 7) [32–34].

Thus, taking into consideration the requirements to the mechanical characteristics and the rate of biodegradation, magnesium alloys of the system Mg-Nd-Zr are the most promising materials for the production of biosoluble implants. However, the lasting effect of biocorrosion reduces mechanical properties of these alloys. Aging the samples in gelofusin revealed that after 3 months of use (average time of fracture consolidation) an implant loses half of its strength (Table 8).

Table 7

Physical-mechanical properties of alloys of the system Mg–Nd–Zr

Alloy	Elasticity modules, GPa	Tensile strength, MPa	Relative elongation, %
ML10	44	235	>3
WE-43	44	250	>2
NZ30K	44	230	>3
Bone tissue	3–20	30–150	1.4–3.1

Note: average values

Table 8

Mechanical properties of the alloy Mg Nd Zr (ML10) after aging in gelofusin

Basic	1 month	2 months	3 months
σ_B, MPa	δ, %	σ_B, MPa	δ, %	σ_B, MPa	δ, %	σ_B, MPa	δ, %
235	3.0	178	2.6	146	2.3	115	1.2

An analysis of phase state diagrams of the systems Mg–Nd and Mg–Zr has revealed that the increase in content of neodymium in the alloy leads to the hardening of a solid solution, as well as to the formation of a larger amount of the reinforcing phase $(Mg, Zn)_{12}Nd$ that will have a positive influence on tensile strength (σ_B) [35]. Increasing the amount of zirconium in the alloy will exert a positive effect on plasticity due to the increased number of nucleation centers and subsequent grinding of the grain, thus increasing relative elongation (δ) [36].

We studied both separate and joint effect of the content of both elements on the mechanical properties of the alloy $(\sigma_B$ and $\delta)$. Results from the mechanical tests of samples are given in Table 9.

Table 9

Results from mechanical tests of samples

Sample No.	Content Zr, %	Content Nd, %	Content Zn, %	σ_B, MPa	δ, %
1	0.4	2.2	0.1	230	2.6
2	1.5	2.2	0.1	236	5.4
3	0.4	3.4	0.1	298	2.7
4	1.5	3.4	0.1	258	3.9
5	0.4	2.2	0.7	232	2.8
6	1.5	2.2	0.7	237	5.5
7	0.4	3.4	0.7	300	2.9
8	1.5	3.4	0.7	260	4.1
9	0.95	2.8	0.4	242	3.3
10	0.95	2.8	0.4	232	3.1
11	0.95	2.8	0.4	236	2.9

Note: average values

Neodymium has had a positive effect on the tensile strength of the alloy. Zirconium, in turn, enabled an increase in relative elongation. Zinc has had a minimal impact on the entire set of properties. The joint effect of neodymium and zirconium is not additive in character, which is why at a high content of both elements the mechanical properties of the alloy are compromised.

5. 2. Analysis of microstructures

Metallographic examination of samples has shown that the initial structure of the alloy consists of a solid solution of neodymium, zirconium, and zinc in magnesium and intermetallides $(Mg, Zn)_{12}Nd$ (Fig. 1, *a*). An increase in the content of neodymium resulted in an increase in the number and size of the reinforcing phase (Fig. 1, *b, c*).

The microstructure of samples with a high content of zirconium (Fig. 2) had a finer grain. The effect of grinding the grain intensified with the increased content of zirconium.
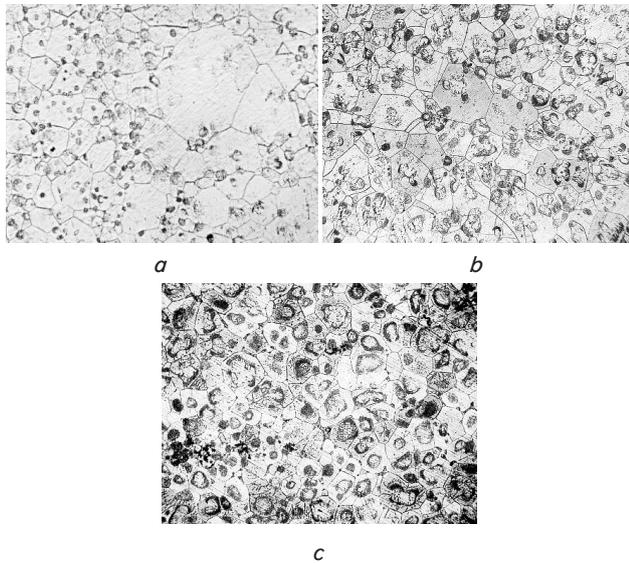
Fig. 1. The microstructure of magnesium alloy samples with different content of Nd, ×350: a — 2.2 % Nd; b — 2.8 % Nd; c — 3.4 % Nd
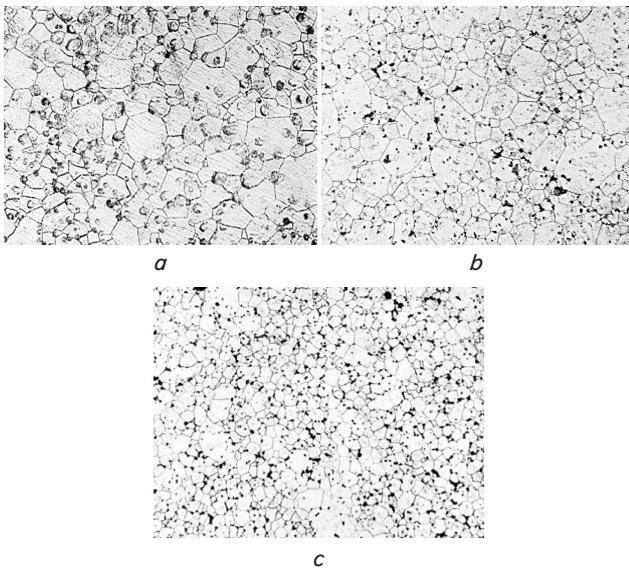

Fig. 2. The microstructure of magnesium alloy samples with different content of Zr, ×100: a — 0.4 % Zr; b — 0.95 % Zr; c — 1.5 % Zr

Thus, the metallographic analysis of samples with different content of alloying elements has shown a positive effect of zirconium and neodymium on the general alloy microstructure.

5. 3. Mathematical processing of experimental data

To study the influence of neodymium and zirconium on tensile strength (σ_B) and relative elongation (δ), we compiled a planning matrix in accordance with design 2^3 (Table 10). The influence of zirconia is encoded by number X_1, neodymium – X_2, and zinc – X_3, respectively. The joint effect of zirconium and neodymium is encoded by number X_{12}, zirconium and zinc – X_{13}, neodymium and zinc – X_{23}, all elements – X_{123}.

The result of mathematical processing of data is the derived regression equation (1), which describes the influence of chemical elements on the alloy's tensile strength:

$$\sigma_B = 256{,}4 - 8{,}625x_1 + 22{,}625x_2 - 11{,}375x_1x_2. \qquad (1)$$

Table 10

Matrix of experiment design 2^3

Experiment No.	X_1	X_2	X_3	X_{12}	X_{13}	X_{23}	X_{123}	σ_B, MPa	δ, %
1	−1	−1	−1	1	1	1	−1	230	2.6
2	1	−1	−1	−1	−1	1	1	236	5.4
3	−1	1	−1	−1	1	−1	1	298	2.7
4	1	1	−1	1	−1	−1	−1	258	3.9
5	−1	−1	1	1	−1	−1	1	232	2.8
6	1	−1	1	−1	1	−1	−1	237	5.5
7	−1	1	1	−1	−1	1	−1	300	2.9
8	1	1	1	1	1	1	1	260	4.1
9	0	0	0	0	0	0	0	242	3.3
10	0	0	0	0	0	0	0	232	3.1
11	0	0	0	0	0	0	0	236	2.9

An analysis of the resulting regression equation has revealed that an increase in the content of neodymium significantly strengthens the alloy while an increase in zirconium reduces its strength. The joint effect of neodymium and zirconium has a negative influence on the tensile strength. The influence of zinc was negligible.

The calculations were also performed for relative elongation. The regression equation takes the following form:

$$\delta = 3{,}7375 + 0{,}9875x_1 - 0{,}338x_2 - 0{,}388x_1x_2. \qquad (2)$$

The resulting regression equation showed that zirconium increases plasticity while neodymium decreases it. The joint effect of neodymium and zirconium negatively affects the plasticity. The influence of zinc was insignificant.

After decoding the regression equations, we derived the following dependences:

$$\sigma_B = 187{,}96 + 16{,}29*Zr(\%) + \\ +27{,}04*Nd(\%) - 8{,}62*Zr(\%)*Nd(\%); \qquad (3)$$

$$\delta = 2{,}96 + 1{,}72*Zr(\%) + \\ +0{,}002*Nd(\%) - 0{,}29*Zr(\%)*Nd(\%). \qquad (4)$$

The derived dependences (3) and (4) take the form of the equation of a plane that makes it possible to represent them graphically in a three-dimensional coordinate system (Fig. 3).
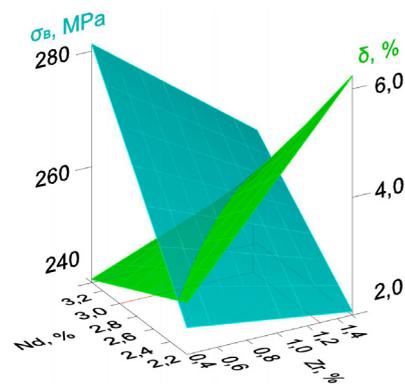

Fig. 3. Dependences of mechanical properties on chemical composition of the alloy Mg Zr Nd in graphical form

An analysis of the derived dependences has allowed us to establish a limit on the content of alloying elements in the designed magnesium alloy (Zr: 1.25–1.3 %, Nd: 2.9–3.1 %), ensuring the maximum combination of plastic and strength properties.

5. 4. Industrial tests

For industrial testing, we fabricated samples in the form of malleolar screws of different designs.

The microstructure of the alloy had a uniform finely dispersed structure and its mechanical properties significantly outperformed the available alloys Mg–Zr–Nd (Table 11).

Table 11

Results from mechanical tests of the alloy with an optimal content of alloying elements

Content Zr, %	Content Nd, %	Content Zn, %	σ_B, MPa	δ, %
1.25	2.98	0.61	266	4.3
1.3	3.05	0.69	271	5.1
1.28	3.1	0.54	274	4.6

Studying the obtained samples after aging in gelofusin for 3 months has shown that the alloy retains the necessary level of properties up to the full consolidation of the fracture (Table 12). Given the result obtained, the alloy is recommended for further preclinical testing.

Table 12

Mechanical properties of alloys with a different chemical composition after aging in gelofusin

Material	Basic		1 month		2 months		3 months	
	σ_B, MPa	δ, %	σ_B, MPa	δ, %	σ_B, MPa	δ, %	σ_B, MPa	δ, %
ML10	235	3.0	178	2.6	146	2.3	115	1.2
Designed alloy	270	4.7	246	4.4	220	4.0	188	3.2

Note: average values

5. 5. Pre-clinical tests

The experiment on rats has established that the products of biocorrosion of the new alloy do not exert toxic effect on the tissues of the body and do not enhance the cellular destruction. This is evidenced by the lack of signs of endogenous intoxication and oxidative damage to functional macromolecules.

Gradual (over seven months) metabolism of metal clamps made from the biosoluble magnesium alloy of the system Mg–Zr–Nd by the body of white inbred male rats was accompanied by the absence of disruption in the physiological manifestations in experimental animals.

We have registered specific and nonspecific symptoms of intoxication related to studying the biological safety of products from the biodegradation of magnesium implants. We detected the absence of proteinuria and the increased content of nitrite in the urine in the dynamics. There was no adverse effect on the overall physical state (there were no pathological changes to eyes, hair, mucous membranes, no changes in body weight). The high mobility and investigative activity, the absence of neurological deficit and abnormalities in the emotional state, indicated no changes in behavior.

The above results testify to the absence of toxic effect from the products of biodegradation of the examined magnesium-based alloy on the body of experimental animals.

In a study that involved rabbits, we found that the use of implants made from the designed alloy did not disrupt the processes of vascularization and angiogenesis in fracture zones as the main factor of differentiation of progenitor cells into osteoblasts. In all cases, we identified a developed micro-vessels network, whose density increased in accordance with the intensity and duration of the reparative process. Already at the early stages after breaking, the formation of reticulofibrous tissue proceeded with the formation of cavities of different sizes, at the inner surface of which the endothelial lining appeared. When using the stainless-steel implants for osteosynthesis, we observed a disruption in blood circulation to the osteon of the compact bone. This led to the death of progenitor cells with subsequent resorption of these sites and their replacement with their fibers of connective tissue. When applying the magnesium implants, no such changes were observed.

Thus, an adequate blood supply defined the gradual and progressive process of regenerative changes that were little different from the normal reparative process.

At late stages of observation (6 months), a section of the bone in the region of the implant made from magnesium alloy did not differ in its structure from the normal bone structure. The newly formed bone callus slightly differed from the bone tissue only by a somewhat disorderly arrangement of bone beams.

6. Discussion of results of designing and studying the new alloy of the system Mg–Zr–Nd

Mathematical processing of experimental data on the influence of neodymium and zirconium on mechanical properties has shown that increasing the amount of neodymium helps strengthen the alloy, while its ductility decreases, which is associated with an increase in the amount and the size of the redundant phase. The admixtures of zirconium to the alloy contributed to increasing the value for plastic properties, while lowering the strength characteristics of the metal. Such results are explained by a change in the microstructure of the alloy as a result of doping. Metallographic analysis shows that zirconium contributes to the grinding of grain and size in the structural components of the alloy; an increase in the content of neodymium led to an increase in the size and amount of the redundant phase.

The mathematical models derived have made it possible to optimize the chemical composition of the alloy in order to achieve the maximum level of mechanical properties. In this case, the maximal estimated values σ_B=260 MPa and δ=4 % are achieved at the following ratio of alloying elements: Zr: 1.25–1.3 %, Nd: 2.9–3.1 %.

During pre-clinical trials, it was found that the chemical elements in the alloy are non-toxic and enhance cellular destruction. The experiments on laboratory rats showed no disruption to their physiological functioning.

Studying the influence of the designed alloy on reparative osteogenesis in the experiment on rabbits showed the positive dynamics of bone tissue recovery without noticeable changes in the bone tissue structure, which ensures reliable cross-linking of bone elements at osteosynthesis.

Thus, the experiments on animals have shown that the implants made from the designed alloy possess the necessary level of mechanical properties that correspond to the mechanical properties of bone tissue. In addition, they are non-toxic and ensure complete healing of the bone tissue until the full consolidation of the fracture.

An analysis of mathematical processing of experimental data on the influence of the studied alloying elements on mechanical properties of the metal has revealed that increasing the content of neodymium in the alloy up to 2.9–3.1 % contributed to its consolidation and lower plasticity. This is predetermined by the additional solid solution hardening with neodymium and the evolution of intermetallide phases following the heat treatment. The admixtures of zirconium to the alloy up to 1.25–1.3 % enhances its plasticity, while the metal's strength characteristics decreased. Such research results are attributed to the grinding of the microstructure of the alloy as a result of doping through the formation of additional centers of crystallization. A further increase in the content of alloying elements in the alloy (Nd≥2.9–3.1 %, Zr≥1.25–1.3 %) led to the formation and excessive evolution of intermetallide phases with complex composition along the grains' boundaries, which contributed to the embrittlement of the metal and a decrease in the entire set of properties.

In order to fabricate implants at osteosynthesis, stainless steels, titanium, cobalt alloys, etc. are widely used at present. These materials are biologically inert and possess a high level of mechanical properties. However, it is necessary to remove the implants made from these materials from the human body after a surgery, which requires repeated surgeries and prolongs the healing process. An alternative to such alloys could be the biosoluble materials. The most common among them are the polyacid-based materials. However, they are expensive while products of their biodegradation are not always absorbed by the body.

A promising material for making implants is magnesium. It is a natural element in the body, it is included into the composition of bone and muscle tissues and is involved in many metabolic processes. However, pure magnesium has a low level of mechanical properties, which makes it difficult to use. A variety of magnesium-based alloys available in machine building have a sufficient level of mechanical properties, but the rate of their biodegradation, toxicity, other medical indicators, made it difficult to use them as implants.

The advantages of the designed biosoluble magnesium alloy:

– the level of mechanical properties of the alloy corresponds to the level of properties of bone tissue;

– the implants made from a given alloy maintain the level of physical-mechanical properties, up to the full consolidation of the fracture;

– non-toxic;

– produce an antibacterial effect.

The performed optimization of chemical composition of the alloy made it possible to establish the rational content of zirconium (1.25–1.3 %) and neodymium (2.9–3.1 %), which ensures the best set of its mechanical properties that distinguishes it from existing analogs. Exceeding these limits changes the structure formation of the alloy and degrades its mechanical properties.

Despite a sufficient accuracy of the results obtained, the models constructed have certain limitations. First of all, they show the dynamics of change in mechanical properties only within the predefined limits: Nd: 0–3.4 %, Zr: 0–1.5 %. In addition, a procedure of mathematical modeling implies the presence of errors in calculations. Consequently, the results derived will be somewhat different from those obtained practically. In addition, the state diagrams for Mg–Zr and Mg–Nd are not examined enough and are slightly different in different sources. Given this, there is an opportunity to further refine the chemical composition, based on practical application.

The positive results of experiments conducted allow us to recommend the use of implants made from the designed biosoluble alloy of the system Mg–Zr–Nd for clinical studies.

7. Conclusions

1. It was established that zirconium and neodymium improve the alloy microstructure and enhance a set of its mechanical properties. These alloying elements are non-toxic and are a promising material for the development of new magnesium-based biosoluble alloys in order to fabricate implants. Metallographic analysis showed the positive effect of alloying elements on microstructure of the alloy. Increasing the content of neodymium to 3.4 % had a positive effect on the tensile strength of the alloy (increased up to 300 MPa) by increasing the amount and size of the reinforcing phase. Increasing the content of zirconium to 1.5 % ensured considerable grinding of the grain, resulting in an increase in relative elongation (to 5.5 %).

2. The result of mathematical processing of experimental data is the derived equations that describe the effect of the examined alloying elements on mechanical properties of the alloy. An analysis of dependences obtained has made it possible to establish the chemical composition of the alloy that ensures the optimal set of properties: Zr: 1.25–1.3 %, Nd: 2.9–3.1 %.

3. The industrial tests that we performed have confirmed the results from mathematical processing. The alloy melted under industrial conditions had a high level of physical and mechanical properties; after 3 months of exposure to gelofusin, the alloy had the following properties: σ_B=188 MPa, δ=3.2 %.

4. The preclinical trials that we carried out have confirmed the absence of toxic effect from the products of degradation of the designed magnesium alloy on the living organism. The positive dynamics were observed in the recovery of bone tissue when studying the influence of the developed alloy on reparative osteogenesis.

Results from the experiments conducted allow us to suggest a favorable prognosis about the possibility of using implants made from the designed biosoluble alloy of the system Mg–Zr–Nd in humans.

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