GLASS FIBERS AND POLYSILOXANES BASED COMPOSITE MATERIALS FOR BIOMEDICAL APPLICATIONS

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Abstract: The study presented deals with development of advanced composite materials to be used in the bone tissue engineering, based on the polysiloxane matrix and glass fibres, and endowed with 3D porous architecture. An extended experimental investigation was necessary for obtaining physical and chemical properties of the composite materials, namely their mechanical properties, porosity, void fraction, surface roughness. All these parameters had to be modified and studied, using both in vitro and in vivo testing, for achieving an optimum bone tissue ingrowth and maturation.

Introduction

Three major groups of materials are used in the bone surgery: metals, ceramics and synthetic polymers. Each of these material types has its own specific advantages and limitations. It is important to produce implants matching mechanical and also other physical properties of the bone and to find non-toxic, non-immunogenic and non-carcinogenic, low-cost, easy available material. Various kinds of composite biomaterials are becoming increasingly important in biomedical practice. Among them also glass composites can be potentially used in orthopedics in the form of substitutive or connective elements [1].

The aim of this study is to design and to produce biomaterial with mechanical and biotolerant properties that should approach as much as possible the properties of the human bone, the strength characteristics should be at least the same or higher and the modulus of elasticity should be close to the value characterizing the human bone [2, 3]. It should have a sufficient porosity, to enhance a bone growth. However, by the biocompatibility is meant now not only a passive biocompatibility or an inertness, i.e., the facilitation of the growth of the tissue around the implant without any signs of toxicity, but especially the bioactivity, i. e. the assurance of a specific biological response on the interface of the material, resulting in the formation of a solid bond between the material and the tissue [4, 5]. In this respect we designed methodology for composite surface modification, prepared samples with various open porosities and studied their biological responses on the interface between the implants and the bone tissue.

Materials and Methods

The composites were prepared from plain-woven cloth V240 (E-glass, VETROTEX, Litomysl, Czech Republic), and from satin-woven fabric 21055 (R-glass, VETROTEX, Saint Gobain, France) and the polysiloxanes LUKOSIL 901 (L901 and LUKOSIL M130 (M130)) as the precursors of matrix, both commercial products of Lucebni zavody Kolín, Czech Republic. The curing temperature was 200-350°C (in nitrogen). The analysis of the properties of the glass composites tested represented the testing of four types of samples in the production of which various combinations of the basic precursors were used.

Analysis of the mechanical, biological and surface properties consisted in testing four types of samples. The samples were produced with different precursors (each sample is a combination of one matrix type and one fibre type) but with the same technology to compare their mechanical and biotolerance properties. It was aimed at the selection of the most suitable composite material.

Mechanical properties of the glass composites were obtained using three methods (see Table 1). Young's modulus *Eres.* and shear modulus in elasticity *Gres.* were measured by means of the electrodynamic resonant frequency tester ERUDITE. Young's modulus *E4p.b.* and the flexural strength *Rm* were determined by a four-point and three-point (for *Rm*) bending arrangement on the material tester INSPEKT. Young's modulus *Estr.* and the compressive strength *Rstr.* were measured on samples with dimensions enabling the strain gauges application, while loading the samples in the parallel direction to the composite laminae in the MTS material tester. To ensure a full contact between the samples tested and the hydraulic jaws, special fixtures were manufactured. The R-glass+L901 samples were discarded from the mechanical testing because of

ISSN: 1727-1983

an extensive delamination of individual layers of the composite.

Table 1: Mechanical Properties (Vf – Volume Fraction of Fibers)

	Vf [%]	Rm [MPa]	Eres. [GPa]	E4p.b. [GPa]	Gres. [GPa]	Rstr. [MPa]
E+M130	51	200.8	25.4	23.7	2.4	80.39
E+L901	52	195.8	27.7	25.7	2.8	119.9
R+M130	65	97.9	16.2	12.2	5.8	52.95

Another important and necessary part of our study is testing of biological properties. Biological properties were observed in in vitro tests – adherence, proliferation and metabolic activity of cells growing on the tested materials, and levels of inflammatory cytokines exprimed during the cultivation into the cell medium. The medium of this cultivation experiment was performed for cytokines *TNF-α*, *IL-1β* detection using immuno-chemiluminescence method of the analyzer Immulite (DCP, Los Angeles, USA), see Table 2.

Table 2: "Ranking" of Biological Properties

Ranking according to: (*1= best, 4= worst*)	R+ M130	R+ L901	E+ M130	E+ L901
The metabolic activity of cells cultured in the medium prepared by using the liquid extracts from tested composites	1.	2.	3.	4.
The metabolic activity of the cells adherent to the tested materials	1.	4.	3.	2.
Cytokine productions	1.	3.	3.	2.

Not only a composite material exhibiting similar mechanical properties in comparison with that of the human bone was looked-for (see Table 1). From the standpoint of interaction with the surrounding bone tissue, bone implants can be designed in two ways, in the form of substitutive and filling elements or supporting elements (plates). In other words, in a form of materials interacting with the bone or in a form of inert materials, which can be placed into human body just for a particular time period and removed after the treatment (e.g., bone plates for osteosynthesis of long bones).

Composites based on polysiloxane resins (produced by Lucebni zavody, Kolin, Czech Republic) promoted colonization with human osteoblasts of the line hFOB 1.19 [6]. However, procedure of glass composites production does not led to a significant increase in the open porosity. The small pore sizes give the hard bone tissue a minimum possibility to down-grow to the glass composites (see Figure 1).

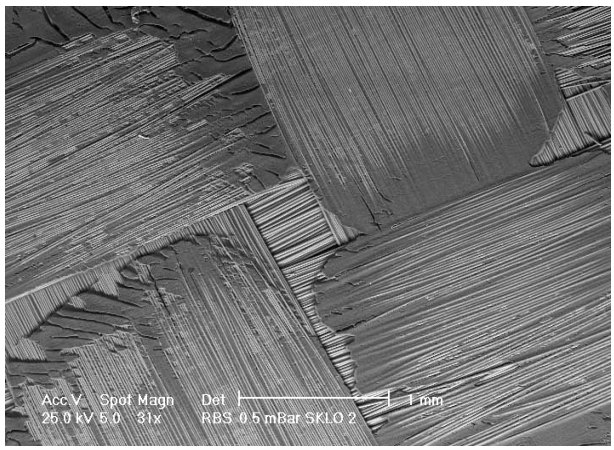

Figure 1: The Primary Surface of Glass Composite

This is possible only in the case of the pores with a minimum size of 0.15 mm [7]. Next step of our project is the study of the surface structure and examination of possibilities of morphology changing (see Figure 2 and Figure 3) due to the bone cells ingrowth.

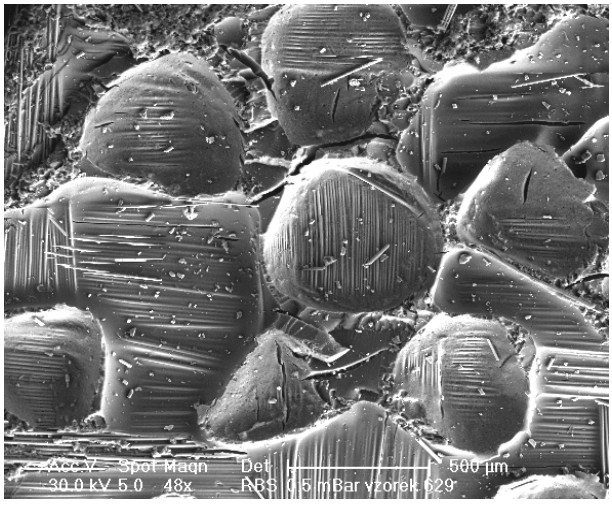

Figure 2: The Treated Surface of The Glass Composite (R-glass+M130, Pores Size 0.45-0.6mm)

Based on previous tests we've chosen the composite material with a sufficient compromise between both mechanical and biological properties (R-glass+M130) and prepared samples with three different pore sizes, namely 0.25-0.45 mm, 0.45-0.6 mm, >0.6 mm. These pores were created by backfilling of a special salt particles with controled dimensions under pressure. The salt particles were further washed thoroughly after the composite curing progress.

Special tests (using rabbits - based on Nakamura's method [8, 9, 10, 11]) were performed. to compare the interfacial bonding strength of composite materials with

ISSN: 1727-1983

different surface structures. The composite samples were shaped into a rectengular plate of size 7.5x10 mm.

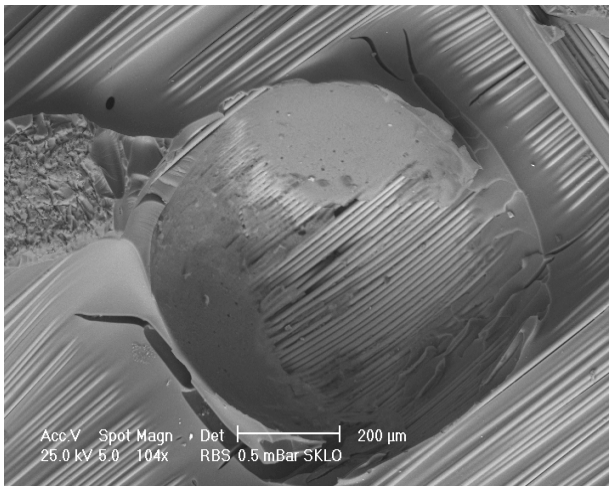

Figure 3: The Treated Surface of Glass Composite, Detail of the Pore (R-glass+M130, Pores Size 0.45-0.6mm

The rabbits were anesthetised and the operations were performed under standard aseptic conditions. The samples were implanted into the proximal condyle of the femur. After seven weeks each sample was removed with surrounding segment of the bone (see Figure 4, left). The bonding strengths in the bone-composite interface were examined (see Figure 4, right, and Figure 6).

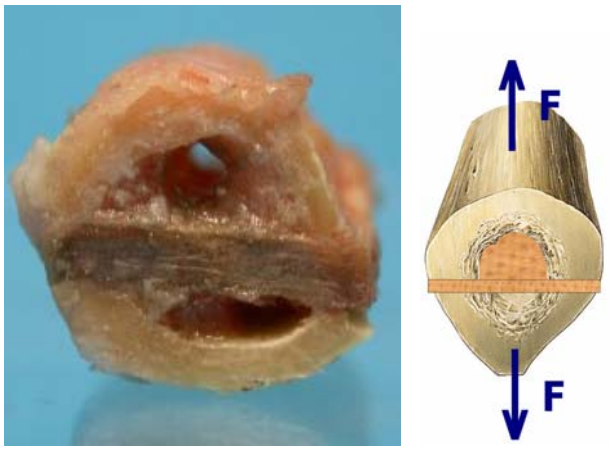

Figure 4: The Tested Sample – A Rabbit Femur with Ingrowthted Composite (left), The Scheme of Pull-off Test (right)

The objective of these tests was to determine the influence of material pore sizes to the bone ingrowing (some results are listed in Figure 5), i.e., those of the optimum „bone-friendly“ surfaces (enabling the bone ingrowth) and those which surfaces are inert for the bone ingrowth.

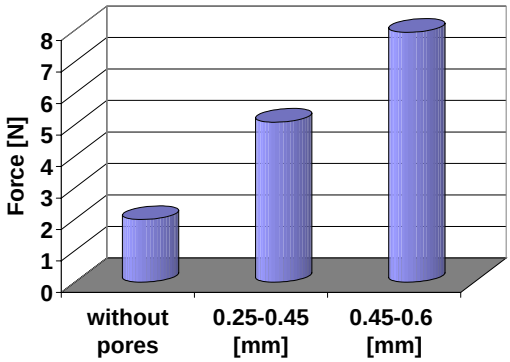

Figure 5: First Results of Pull-off Tests (Applied Force, R-glass+M130)

Another tests will be performed by pure Ringier's solution contact angle measurements. Advancing and receding contact angles will be determined on samples with different surface structure. This will be another description of the surface topography and will help us to find the osteoblast enabling or inert surfaces.

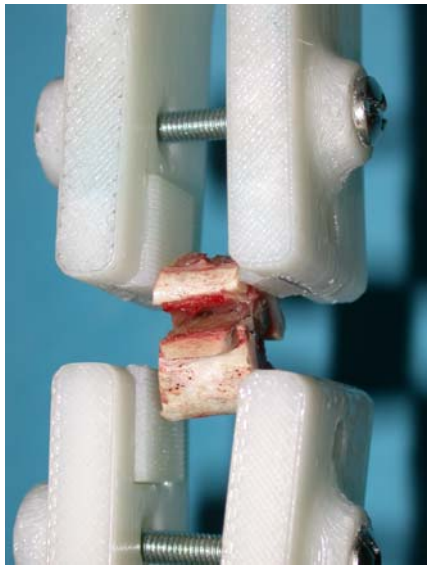

Figure 6: The Ilustration of Pull-off Test (A Bone Segment with the Composite Sample in Special Fixtures After Pull-off)

Results

The objective of this study was to develop an advance composite material to be used in the bone tissue engineering. Another objective was to tend a better understanding of the proportions between material surface properties and the bone ingrowth. Based on these analyses we are able to select a material with sufficient mechanical and biological properties having an inert surface for applications in the form of supporting elements (e.g., bone plates for osteosynthesis of long bones) or a material with sufficient mechanical and biological properties having an open surface contributing to a solid bond between the implant and the

ISSN: 1727-1983

bone tissue for applications in the form of substitutive elements. Another step of our study will be the improvement of material bioactivity by adding of hydroxyapatite into the polysiloxane matrix.

Conclusions

Composites based on glass fibers and polysiloxane resins promise to achieve good biomechanical properties while requiring a significantly lower cost. Depending on their pore sizes, they can be potentially used in the form of supporting, substitutive or filling elements.

Acknowledgements

This study was supported by the GACR under the project No. 106/03/1167 and by the Ministry of Education project: Transdisciplinary research in Biomedical Engineering II., No. MSM 6840770012.

References

[1] RAMAKRISHNA S., MAYER J., WINTERMANTEL E. and LEONG KAM W. (2001): 'Biomedical applications of polymer-composite materials: a review', *Composite Science and Technology*, **61**, pp. 1189-1224

[2] BALIK K., MACHOVIC V., CERNY M., KOLAR F., KRENA J., MATEJKA P. and MIZERA J. (2003): 'Preparation and oxygen resistance of 2D composites based on E-glass, R-glass, and siloxanes', *Materials Chemistry and Physics*, **82**, pp. 458-465

[3] BALIK K., SOCHOR M., SUCHY T. and CERNY, M. (2003): 'Mechanical properties of composites based on glass fibers and siloxane as biomaterial', *Engineering of Biomaterials*, **27**, pp. 3-5

[4] PESAKOVA V., SMETANA JR. K., BALIK K., HRUSKA J., PETRTYL M., HULEJOVA H. and ADAM M. (2003): 'Biological and biomechanical properties of the carbon composite and polyethylene implant materials', *Journal of Materials Science: Materials in Medicine*, **14**, pp. 531-537

[5] PESAKOVA V., SMETANA K., SOCHOR M., HULEJOVA H. and BALIK K. (2005): 'Biological properties of the intervertebral cages made of titatium and containing a carbon-carbon composite covered with different copolymers', *Journal of Material Science:Materials in Medicine*, **16**, pp. 143-148

[6] GUMULA T., BLAZEWICZ S. (2004): 'Study on polysiloxane resin-based composites for bone surgery application', *Polim Med.*, **34**, pp. 49-54

[7] WILLIAMS, P.L., BANNISTER, C.H., BARRY, M.M., COLLINS P., DYSON M., DUSSEK J.E. and FERGUSON M.W.J. (1995): 'Gray's Anatomy', (Churchill and Livingstone, New York)

[8] NAKAMURA ET AL. (1985): 'A new glass-ceramic for bone replacement: Evaluation of its bonding to bone tissue', *Journal of Biomedical Materials Research*, **19**, pp. 94-96

[9] KITSUGI T., NAKAMURA T., OKA M. ET AL. (1996): 'Bone-bonding behavior of plasma-aprazed coatings of Bioglass, AW-glass ceramic, and tricalcium phosphate on titanium alloy', *J. Biomed. Mater. Res.*, **30**, p. 261

[10] TAKATSUKA K., ZAMAMURO T., NAKAMURA T. and KOKUBO T. (1995): 'Bone-bonding behavior of titanium alloy evaluated mechanically with detaching failure load', *J. Biomed. Mater. Res.*, **29**, p. 157

[11] TAMURA J., KITSUGI T., IIDA H. ET AL. (1997): 'Bone bonding ability of bioactive bone cements', *Clin. Orthop.*, **343**, p. 183

ISSN: 1727-1983