LONG-TERM MECHANICAL TESTING OF MULTIFUNCTIONAL COMPOSITE FIXATION MINIPLATES

Karol Gryń*

AGH University of Science and Technology,
Faculty of Materials Science and Ceramics,
Department of Biomaterials and Composites,
Al. Mickiewicza 30, 30-059 Krakow, Poland
*e-mail: kgryn@agh.edu.pl

Abstract

This paper presents the analysis and comparison of the results of mechanical testing of dumbbell-shaped specimens and multifunctional fixation miniplates made via injection forming. Three types of materials were used: a) polylactic acid; b) a composite made of a polylactic acid matrix modified with tricalcium phosphate β-TCP; c) a composite made of a polylactic acid matrix modified with a mixture of bioceramic powders of tricalcium phosphate β-TCP and hydroxyapatite HAp. All the samples were stored in normal conditions, no special treatment was applied. Tests were conducted right after samples were prepared and they were repeated two and four years after preparation. The values of basic mechanical parameters and stress-strain curves were recorded and analyzed. The attention was focused on changes in time of tensile strength and stiffness of materials and implants. It was discovered that having been stored for four years in the open air, without sunlight, with no hermetic sealing, and no sterilization, all the materials (PL38, PL38/TCP, PL38/TCP/HAp) showed slight changes in mechanical characteristics when compared to the data of the initial samples tested after fabrication. These changes were not critical and did not adversely affect either tensile strength or Young's modulus of the implants. All the analyzed miniplates maintained their mechanical properties at an acceptable level, fulfilling requirements for fixation devices for osteosynthesis. Therefore, it was proposed that the expiry date of these implants can be indirectly determined, based on long-term mechanical testing.

Keywords: *multifunctional composites, fixation plates, mechanical testing, implant expiry date, polylactide acid, calcium-phosphate ceramics*

[Engineering of Biomaterials 157 (2020) 20-25]

doi:10.34821/eng.biomat.157.2020.20-25

Introduction

In recent years the development and application of new composite materials for the production of implants have become one of the main topics in the field of biomaterials. The introduction of modifiers into polymeric matrices to improve their strength or increase their stiffness is already a standard procedure [1,2]. There are numerous laboratories around the world where scientists try to provide novel materials with additional therapeutic features and functions. Among them, there are the Department of Biomaterials and Composites (Faculty of Materials Science and Ceramics of the AGH University of Science and Technology in Krakow) and MEDGAL Ltd. (Białystok, Poland). The key achievement of this cooperation were multifunctional composite biomaterials designed to manufacture biodegradable fixation miniplates. These new composite implants made of PLA modified with bioactive ceramic powders (β-TCP and HAp) not only ensure mechanical stability of bone fragments but also support local healing processes and intensify processes of reconstruction of the damaged bone tissue [3].

Besides the standard material and biological description, the most important issues that the manufacturer is obliged to determine are time and conditions of storing the product and its expiry date. In Poland, there are no legal regulations and precise requirements or procedures to determine the usefulness of medical devices, implants or medical equipment. It is generally accepted that the expiry date of reusable products is determined by the quality of packaging and sterilization processes. Thus, it guarantees that the product is safe and sterile until the moment of application. In other words, the expiry date is related exclusively to the biological purity and sterility of the product [4]. Materials commonly used in medicine e.g. metals, alloys or biostable polymers can be treated as reusable products and in these cases "the sterility principle" may be applied. However, the question arises with regard to degradable materials. In most cases, such materials are multiphase systems made of resorbable polymer matrices modified with various additives (drugs, bioactive ceramics, etc.). That is why the variety of possible reactions affecting the final properties of the implants' material may occur [5-10]. A series of complicated, laborious, and expensive structural studies must be performed to assess the behaviour of these materials in time. However, is it necessary in all cases? Is there any other way to verify this behaviour? Based on the presented research results, an inexpensive alternative to estimate the expiry date of an implant is proposed. It is the long-term data obtained through mechanical testing.

Materials and Methods

The samples were made from three types of materials: a) PURASORB PL38 poly(lactic acid), an amorphous resorbable polymer with Food and Drug Administration (FDA) approval for medical applications (PURAC, Europe); b) a composite of the PURASORB PL38 resorbable matrix modified with 8vol% micrometric β-TCP tricalcium phosphate powder (Chema Elektromet, Poland); c) a composite of the resorbable PURASORB PL38 matrix modified with two types of bioceramic additives; total volume fraction did not exceed 8vol%: micrometric tricalcium phosphate powder β-TCP ~7.5vol% and nanometric hydroxyapatite powder HAp ~0.5vol% (Chema Elektromet, Poland).

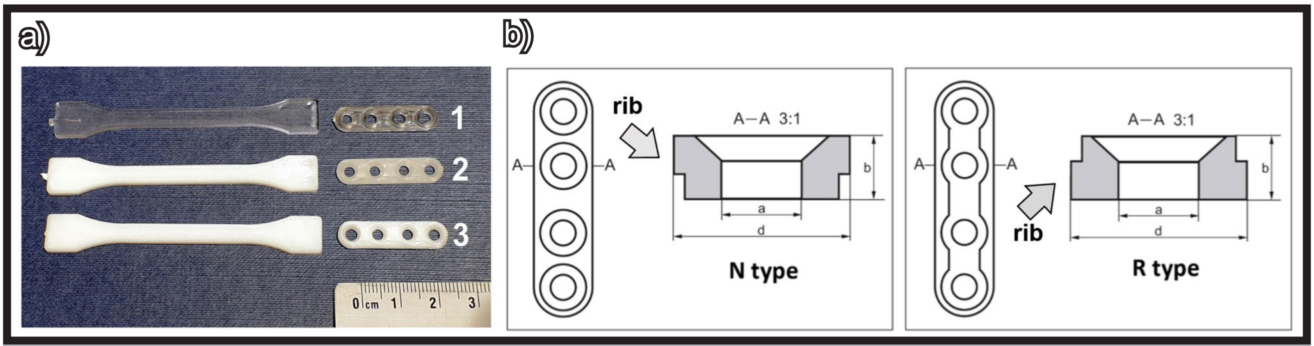

FIG. 1. a) Examples of dumbbell-shaped specimens and fixation miniplates used for testing. 1 - PL38; 2 - PL38/TCP; 3 - PL38/TCP/HAp; b) Schematic sketches of the I-shaped four-hole miniplate: type "N" with reinforcing rib from the bottom; type "R" with reinforcing rib from the top.

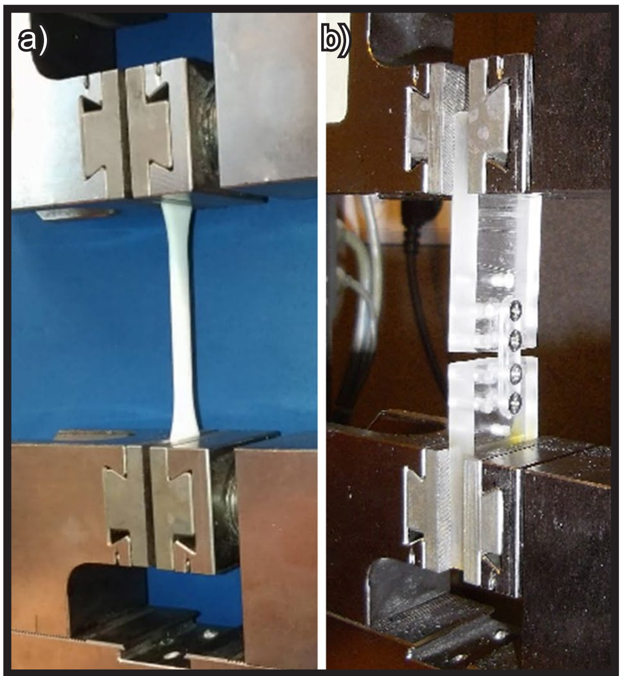

FIG. 2. a) Dumbbell-shaped specimen mounted and ready for testing; b) Simulated model of osteosynthesis with fixation miniplate.

Both powders were approved for medical applications. The samples were made by injection molding on a vertical screw-piston injection molding machine (Multiplas V4-S-15N, Taiwan) at the processing temperature of 180°C, and the pressure in the injection system of 90 kg/cm^2 (~9.0 MPa). Two types of the samples were produced: a) dumbbell-shaped specimens (length: 75 mm, width: 5 mm, thickness: 2 mm) (FIG. 1a); b) I-shaped four-hole miniplates with length: 29 mm, width: 7 mm, thickness: 2 mm, hole diameter: 3 mm (FIG. 1a). Two geometric variants of miniplates were prepared: with a reinforcing rib on the lower surface (type "N"); with a reinforcing rib on the upper surface (type "R") (FIG. 1b). The detailed descriptions of the preparation of dumbbell-shaped specimens and fixation miniplates along with the results from the preliminary tests conducted in 2014-2015 were published in the article: "Mechanical characterization of multifunctional resorbable composite plate for osteosynthesis" [3]. All samples were stored at room temperature, isolated from sunlight. The samples were not sterilized or hermetically sealed. For each type of the samples: dumbbell-shaped, "N" type miniplates and "R" type miniplates, five specimens were examined in the beggining of the experiment, and after two and four years.

All specimens were tested under the same loading scheme and conditions. The static uniaxial tensile tests were performed in accordance with the PN-EN ISO 527-1: 2012 standard. The research was carried out on Zwick 1435 testing machine, coupled with TestExpert v8.1 software. The traverse speed was set at 2 mm/min. The dumbbell-shaped specimens were tested in standard clamps (FIG. 2a). The I-shaped four-hole fixation miniplates were tested in a model simulating osteosynthesis: two blocks of plexiglass (PMMA) measuring 50x25x8 mm were cut. The fixation miniplates were mounted on the blocks using stainless steel screws with nuts (M2.5 A4-70). The gap between the PMMA blocks simulating a bone fracture measured 2 mm in width (FIG. 2b). Screws were tightened with tightening torque M_d = 20 cNm (± 0.01 cNm) with a digital torque wrench (POLTORQUE BMS-150). The values of tensile strength R_m and Young's modulus E were determined. The sets of $\sigma = f(\varepsilon)$ curves were obtained. Additionally, the damaged specimens underwent microscopic examination regarding the fractures.

Results and Discussion

Dumbbell-shaped specimens

When analyzing tensile curves for the dumbbell-shaped specimens (FIG. 3), all the PL38 samples revealed a certain range of plastic deformation. The widest range was observed for the initial specimens (tested right after manufacturing). The local plastic deformation was confirmed during microscopic observations. There was a whitening area around the line of fracture and the small "necking" was found (FIG. 4a). For the composite samples PL38/TCP and PL38/TCP/HAp, the deformation had the elastic-brittle character. For all the materials and all the time intervals, the dumbbell-shaped specimen fracture was located at ~1/5 of the length of the measurement base. The crack line was perpendicular to the long axis of the specimen and the fracture was flat-parallel. Almost no plastic deformation on the tensile curves was recorded, therefore it can be concluded that the composite fracture was brittle (FIG. 4b, 4c).

The comparison of dumbbell-shaped specimens revealed no significant changes in the tensile strength values recorded in time (FIG. 5). For the PL38/TCP/HAp composite, a slight strengthening effect was observed: after 2 years by about 8% and after 4 years by about 10%.

In general, the materials stiffness increased with time (FIG. 5). The biggest changes were recorded for PL38 – for instance, its Young's modulus almost doubled after four years of storage. For the PL38/TCP/HAp composite there were not such significant differences in the value of Young's modulus: less than 20%.

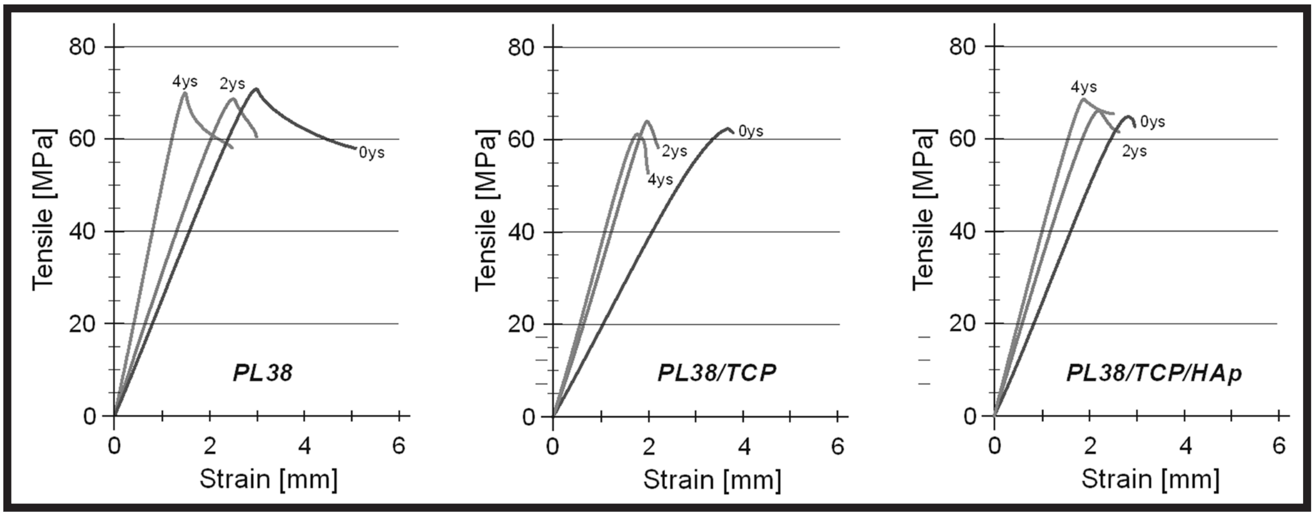

FIG. 3. Examples of tensile curves of dumbbell-shaped specimens.
0ys - initial samples; 2ys - tested after two years; 4ys - tested after four years.

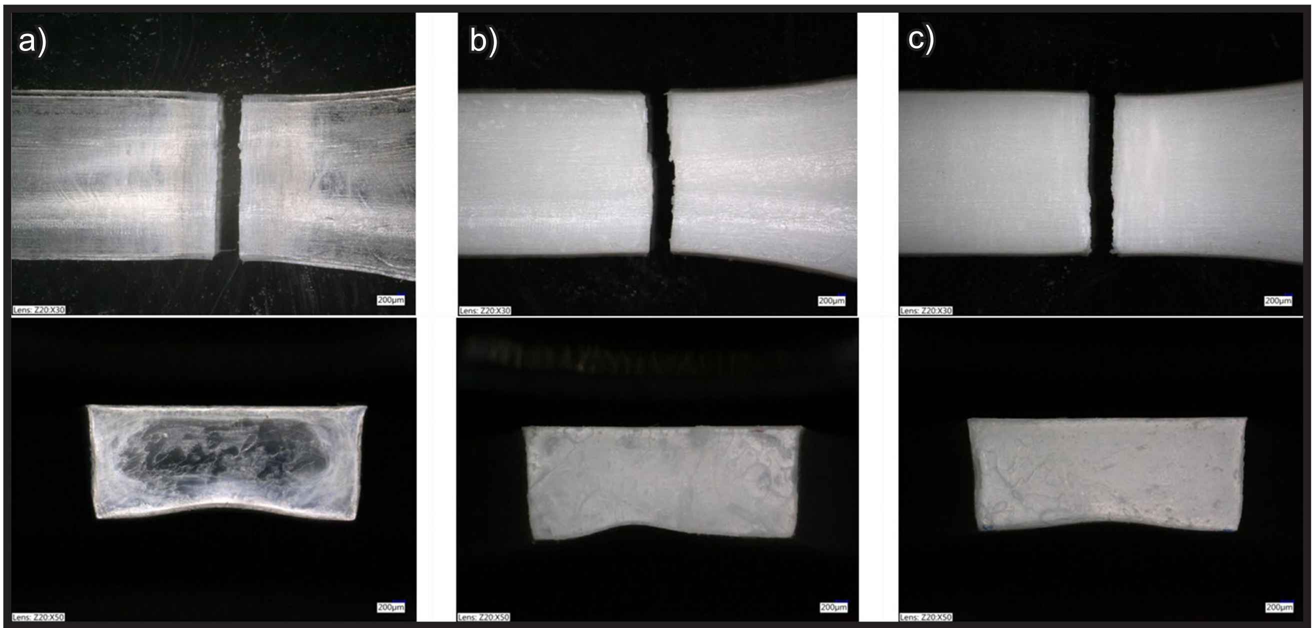

FIG. 4. Example of microscopic images of sample fracture (KEYENCE VHX-500, mag. x30; x50): a) PL38; b) PL38/TCP; c) PL38/TCP/HAp.

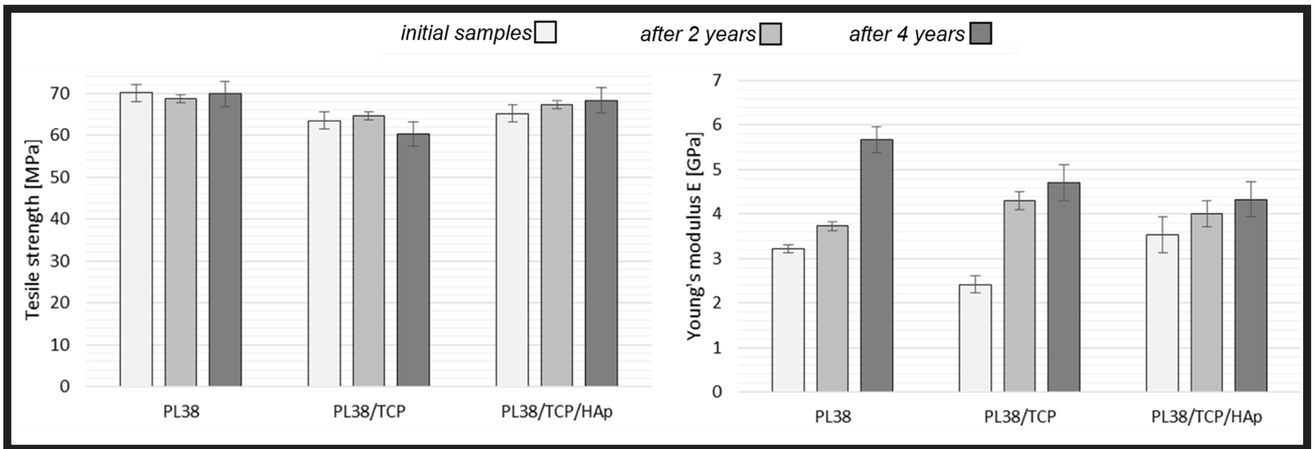

FIG. 5. Comparison of average R_m and E values obtained during mechanical testing of dumbbell-shaped specimens of the initial samples, after 2 and after 4 years.

Fixation miniplates

The results of the mechanical tests of the fixation miniplates are presented as tensile curves (FIGs. 6, 7) and graphic bars (FIGs. 8, 9).

All the curves show the tensile stress values fluctuation area and a subsequent decrease in the graph angle with respect to the strain axis. This might result from localized plastic deformation in the critical cross-section – i.e. the area of the fixation hole – causing changes in the implant geometry. The miniplate thickness might have decreased and the shape of the fixation hole changed from round to elliptic. Thus, the stress state ensuing from the pressure under the fixation screw conical head was no longer uniform. Nevertheless, it was also found that fixation miniplates with the reinforcing rib on the upper surface ("R" type) had better and more stable characteristics. The mechanical properties of the PL38 and PL38/TCP miniplates decreased with time. An interesting phenomenon was observed for the PL38/TCP "N" sample. After two years of storage, the tensile strength dropped by ~45% but after the next two years, it increased again. In the case of the PL38/TCP/HAp miniplates, different behaviour was noted. The longer the samples were stored, the higher tensile strength values were recorded. After two years the stiffness and Young's modulus values significantly increased but after the next two years they slightly decreased. It can be speculated that such behaviour could result from a change in the polymer crystallinity degree. It may also happen when moisture from the environment is absorbed, possibly due to the presence of HAp in the matrix. To confirm this hypothesis, additional structural tests and analysis are necessary.

In general, the obtained values of tensile strength for all types of miniplates were in the range of 35-65 MPa and the materials stiffness expressed by Young's modulus values fluctuated in the range of 7-14 GPa. Regardless of the storage time, the tested fixation miniplates match the characteristics of natural bone, as the bone tensile strength is 1-180 MPa, depending on the type (spongy/bony) and Young's modulus is from 0.007 GPa to 30 GPa [10].

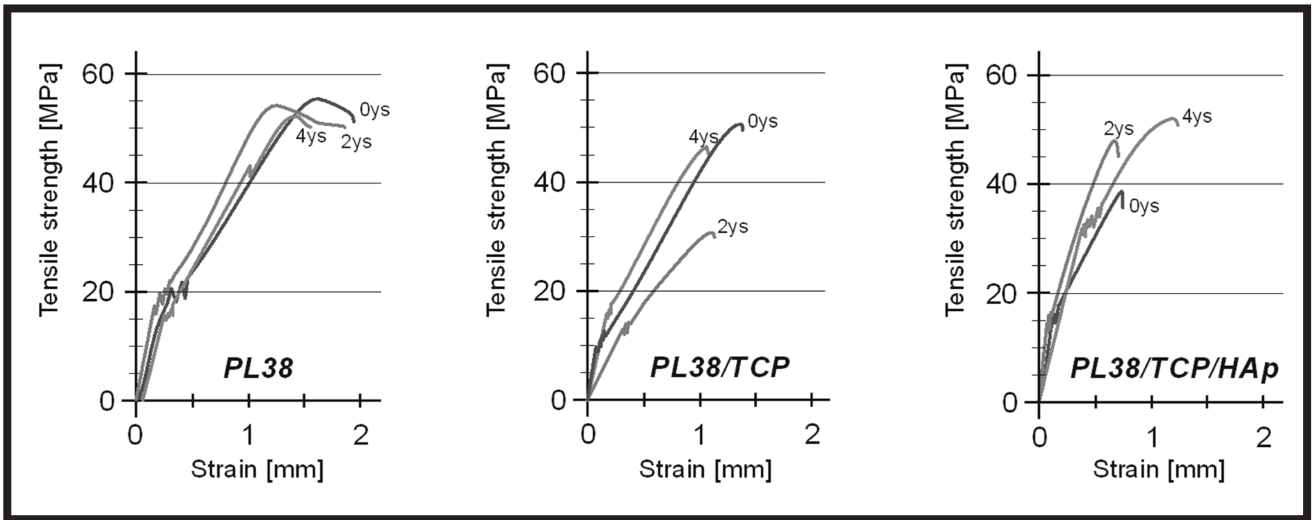

FIG. 6. Example tensile curves of "N" type miniplates.
0ys - initial samples; 2ys - tested after two years; 4ys - tested after four years.

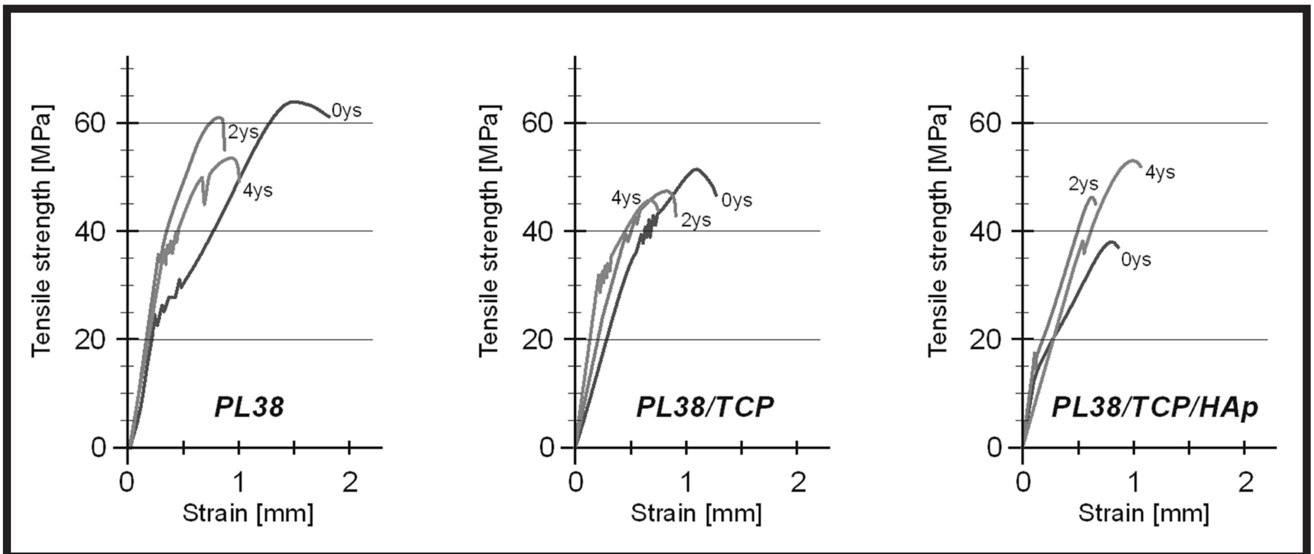

FIG. 7. Example tensile curves of "R" type miniplates.
0ys - initial samples; 2ys - tested after two years; 4ys - tested after four years.

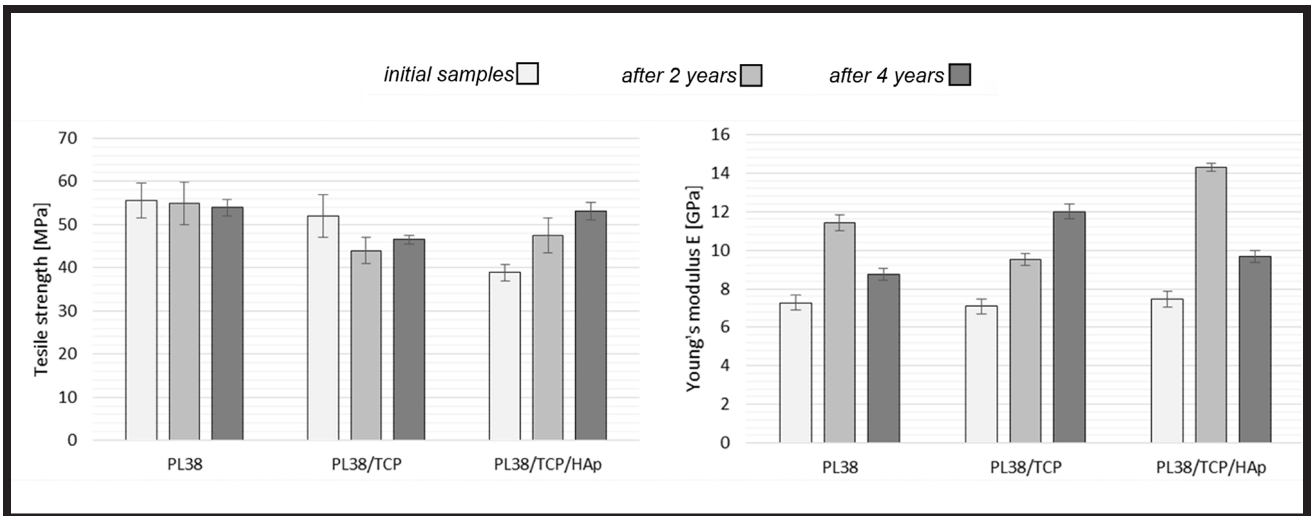

FIG. 8. Comparison of mechanical test results (R_m and E) of the "N" type fixation miniplates of the initial samples, after 2 and after 4 years.

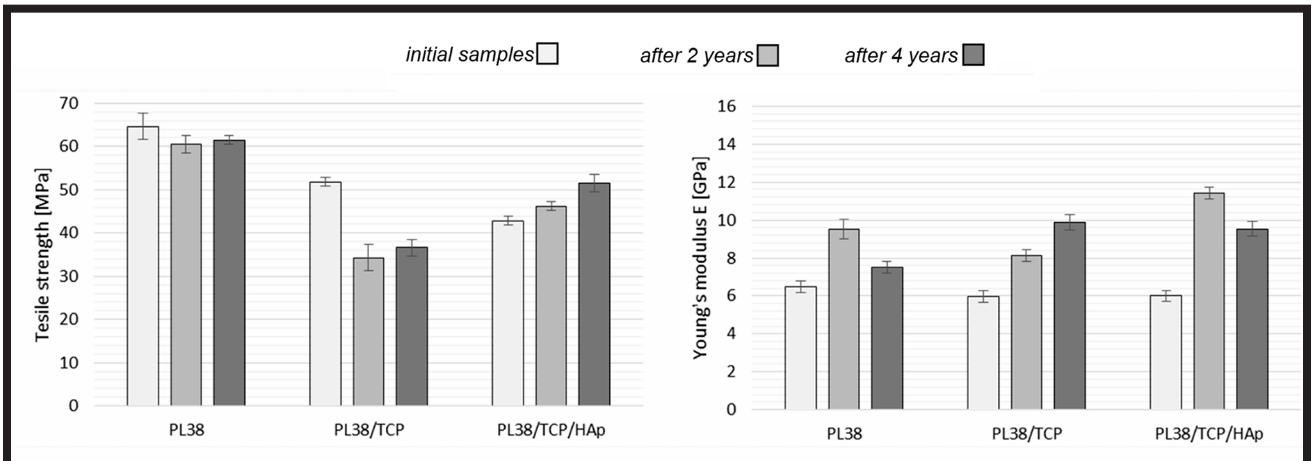

FIG. 9. Comparison of mechanical test results (R_m and E) of the "R" type fixation miniplates of the initial samples, after 2 and after 4 years.

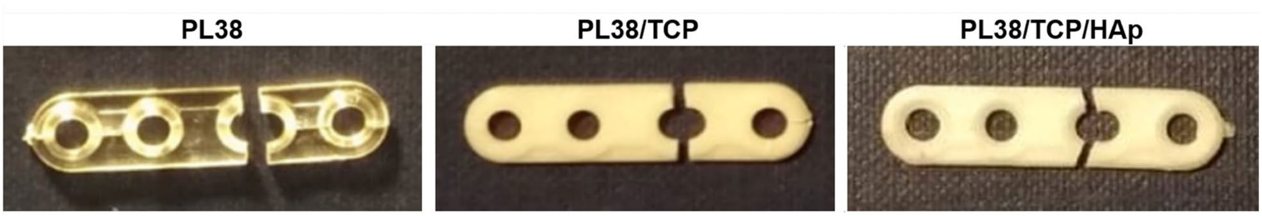

FIG. 10. Examples of miniplates after the mechanical testing. The characteristic manner of fracture can be seen.

The microscopic observation of the damaged fixation I-shaped four-hole miniplates revealed that both "R" and "N" type miniplates fractured in a predictable manner. The fractures occurred close to the first hole proximal to the centre of the miniplate (FIG. 10). The fracture was perpendicular or nearly perpendicular to the plane of tension and the crack was in the first critical cross-section (the weakest part of a miniplate). In the case of PL38, regardless of the storage time, a slight opacity of the specimen was observed in the fracture vicinity. This may indicate a slight local plastic deformation. Due to the white colour and lack of transparency of the PL38/TCP and PL38/TCP/HAp composite miniplates, a similar analysis could not be performed. The fracture lines were in both cases straight and the crack was brittle. This characteristic brittle behaviour was also visible in the stress-strain curves.

Conclusions

Having analyzed the results of the tests, it can be concluded that all the materials (PL38, PL38/TCP, PL38/TCP/HAp) after four years of storage in the open air, without exposition to the sunlight, with no hermetic sealing, and no sterilization, showed slight changes in mechanical characteristics when compared to the data of initial samples tested after their fabrication. These changes might result from the degradation processes of the tested materials (e.g. absorption of moisture from the air, temperature changes). Nevertheless, these changes were not critical and did not affect negatively either the tensile strength or the implants stiffness. All the miniplates maintained their mechanical properties at an acceptable level. Therefore, it can be concluded that even after four years of storage they can be used as a fully valuable product. Considering its tensile strength and Young's modulus, even the weakest miniplate remained within the range required for materials used in osteosynthesis.

In conclusion, the presented methodology can be used to indirectly estimate the expiry date of composite implants. For two years the implant can be safely stored and used in surgery. Since its mechanical properties fulfil the requirements for osteosynthesis fixation device, the implant will surely perform its function properly during this period of time. However, the behaviour of such an "old" implant in the living organism is difficult to predict. Therefore, further studies, especially in a simulated biological environment and the *in vivo* tests will be necessary to verify the results of this work.

Acknowledgements

The study was performed in collaboration with MEDGAL Company within the R&D project: "Elaboration and implementation of the production technology of innovative bone implants made of polymer composites" (POIG.01.04.00-20-003/10).

ORCID iD

K. Gryń: https://orcid.org/0000-0001-6499-2465

References

[1] Marciniak J., Błażewicz S.: Biomateriały, Tom 4, Akademicka Oficyna Wydawnicza Exit, 2016

[2] Lincoln R.L., Scarpa F., Ting V.P., Trask R.S.: Multifunctional composites: a metamaterial perspective. Multifunct. Mater. 2 (2019) 043001.

[3] Gryń K., Szaraniec B., Morawska-Chochół A., Chłopek J.: Mechanical characterization of multifunctional resorbable composite plate for osteosynthesis. Engineering of Biomaterials 133 (2015) 22-33.

[4] Kudzia-Karwowska D.: Czynniki warunkujące okres przydatności wyrobów medycznych. Technika i technologia. Ogólnopolski Przegląd Medyczny 11 (2013) 15-17.

[5] Elsawy M.A., Ki-Hyun K., Akash J.W., Deep A.: Hydrolytic degradation of polylactic acid (PLA) and its composites. Renewable and Sustainable Energy Reviews 79 (2017) 1346-1352.

[6] Kulkarni A., Dasari H.: Current Status of Methods Used In Degradation of Polymers: A Review, MATEC Web of Conferences 144, 02023 (2018).

[7] Rivaton A., Gardette J.L., Mailhot-Jensen B.: Basic Aspects of Polymer Degradation, Macromol. Symp. 225 (2005) 129-146.

[8] Szaraniec B., Gryń K., Szponder T., Żylińska B.: Biodegradable fixation plates for veterinary medicine. Engineering of Biomaterials 125 (2014) 30-36.

[9] dos Santos V., Brandalise R.N., Savaris M.: Engineering of Biomaterials, Springer International Publishing 2017, ISBN 978-3-319-58607-6.

[10] Yuehuei H., Rob. Draughn R.: Mechanical Testing of Bone and the Bone-Implant Interface, CRC Press; 1st edition (November 29, 1999), ISBN-13: 978-0849302664