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COMPOSITES PMMA - CARBON FIBRE AS AN ALTERNATIVE TO CONNECTION OF ENDOPROSTHESIS WITH BONE

Surgical cements, being polymer composites, constitute the substances which fix prostheses to bones. Due to the load which they are expected to bear and responsibility for maintenance of a stable connection of the bone and prosthesis, cements should be characterized by enhanced mechanical strength and biocompatibility. Basic mechanical parameters indicated by PMMA under variable load might affect durability of biomechanical systems, such as artificial femoral joint. For these reasons, analysis of functional properties of bone cements under empirical conditions is of essential importance since it allows for determination of cement's behaviour in human body. Analysis of clinical results demonstrated that bone cements used for hip joint replacement, despite a number of modifications, still do not meet sufficient requirements of biofunctionality, mainly due to high toxicity of liquid monomer. Bone cements are still characterized by insufficient biocompatibility and poor mechanical properties. Furthermore, they show tendency to cracking, chipping and, in consequence, to loosening of prosthesis in bone. This paper aims at determination of the factors which cause destruction of cement layer in human body. Examination of the parts of artificial joint removed from human body because of the destruction of PMMA revealed that cement mass does not adhere to the prosthesis on the whole contact surface but only in some random areas. It was also observed that connection of the cement with bone occurs based on sticking whereas dendrite surfaces of cement show tendency to generate microcracks, which, in the course of time, are progressing and cause destruction of PMMA. The attempts were also made to develop, on the basis of commercial cements, a new composite reinforced with carbon fibre. The investigations aimed at obtaining a cement with limited percentage of toxic monomer, enhanced mechanical strength and porous surface which will allow for osseointegration of composite mass with the bone. It was determined on the basis of the investigations that optimal fraction of carbon fibre fillers in commercial bone cements accounts for 30%. This modification allows for obtaining a composite characterized by higher compressive strength [MPa] as compared to pure cements, from 20 to 26%. The developed PMMA composite with carbon fibre also showed higher impact strength (by approx. 20÷26%), which, in the case of the nature of load it is expected to bear, can considerably extend the time of use inside human body. Another essential advantage of the developed composite is that a porous surface was obtained, which will allow for osseointegration of the modified PMMA with bone and will increase mechanical strength in bonecement arrangement. It is also remarkable that implementation of the filler limits the percentage of monomer in cement mass, which results in a higher biocompatibility in the obtained composite as compared to pure cement.

Keywords: composite, carbon filament, modified PMMA, mechanical properties

KOMPOZYT PMMA - WŁÓKNO WĘGLOWE JAKO ALTERNATYWA POŁĄCZENIA ENDOPROTEZY Z KOŚCIĄ

Cement chirurgiczny, będący kompozytem polimerowym, po związaniu stanowi element spajający protezę z kością. Ze względu na obciążenia, jakie przenosi, oraz na dużą odpowiedzialność w utrzymaniu stabilnego połączenia protezy z kościa powinien charakteryzować się wysoką wytrzymałością mechaniczną oraz biozgodnością. Podstawowe parametry wytrzymałościowe PMMA, jakie wykazuje w warunkach oddziaływania zmiennych obciążeń, mogą mieć wpływ na trwałość układu biomechanicznego, jakim jest sztuczny staw biodrowy. Z tych powodów istotne znaczenie mają badania właściwości użytkowych cementów kostnych w warunkach doświadczalnych, albowiem pozwalają na określenie jego zachowania się w organizmie człowieka. Analiza wyników klinicznych wskazuje, że stosowane w endoprotezoplastyce cementy kostne, pomimo wielu modyfikacji, nadal jeszcze niedostatecznie spełniają stawiane im wymagania oczekiwanej biofunkcjonalności, głównie z powodu dużej toksyczności płynnego monomeru. Cementy kostne wciąż jeszcze charakteryzują się niewystarczającą zgodnością biologiczną, niekorzystnymi właściwościami wytrzymałościowymi. Ponadto posiadają dużą skłonność do pękania, wykruszania, a w konsekwencji do obluzowania protezy w kości. W artykule podjęto próbę określenia czynników powodujących niszczenie płaszcza cementowego w organizmie ludzkim. Na podstawie elementów sztucznego stawu usuniętego z organizmu ludzkiego z powodu zniszczenia PMMA określono, iż przyleganie masy cementowej do protezy nie następuje na całej powierzchni kontaktowej, a jedynie w pewnych przypadkowych Dodatkowo obszarach. zaobserwowano. że połączenie cementu z kością odbywa się na zasadzie przyklejenia, a dendrytyczne powierzchnie cementu mają skłonności do powstawania mikropęknięć, które wraz z czasem postępują, powodując niszczenie PMMA. W badaniach podjęto próbę opracowania na bazie cementów komercyjnych nowego kompozytu wzbogaconego w włókno węglowe. Celem prowadzonych badań jest uzyskanie spoiwa, w którym ograniczony zostanie udział toksycznego monomeru, zwiększy się wytrzymałość mechaniczna oraz uzyska się porowatą powierzchnię, która umożliwi przerost masy kompozytowej kością. Na podstawie przeprowadzonych badań określono, że przy zastosowanych komercyjnych cementach kostnych optymalny udział wypełniaczy z włókna węglowego stanowi około 30%. Modyfikacja taka pozwala na uzyskanie kompozytu charakteryzującego się wyższą wytrzymałością na ściskanie Rc od czystych cementów od 20 do 24%. Opracowany kompozyt PMMA z włóknem węglowym zwiększył również swą udarność o około 20÷26%, co w przypadku charakteru jego pracy może znacznie wydłużyć czas użytkowania w organizmie ludzkim. Kolejną bardzo istotną zaletą opracowanego kompozytu jest uzyskanie porowatej powierzchni, co umożliwi przerośnięcie modyfikowanego PMMA kością i znacznie zwiększy wytrzymałość połączenia kość-cement. Nie bez znaczenie jest również fakt, że wprowadzenie wypełniacza ogranicza procentowy udział monomeru w masie cementowej, przez co uzyskany kompozyt ma wyż-szą biozgodność od czystego cementu.

Słowa kluczowe: kompozyt, włókno węglowe, modyfikacja PMMA, wytrzymałość mechaniczna

INTRODUCTION

PMMA after polymerization is a strong binder that should hold implanted endoprosthesis inside the marrow cavity and bear the load resulting from human motor activity [1]. The application of cements has been shown to provide opportunities of fixing of the prosthesis inside a bone. However, it had to be done in a suitable manner while surgeons must understand how it works. Cement should not be treated as a glue but as a filler-filling the space between a bone and metal and transferring the load to the bone. Insertion of the prosthesis with the cement applied on it is useless as there are neither adhesion forces nor chemical reactions present between the cement and the bone. However, inserting the endoprosthesis into a cavity filled with cement generates the pressure pushing cement particles into the gaps in bone structure. After polymerization, the uniform system of prosthesis and cement is obtained while its external surface is the negative of the endosteal structure [2].

Clinical practice has shown a high percentage of failures of artificial hip joint implantation operations caused by the loss of stability of the mandrel placed inside the femur [3]. The considerable percentage of the endoprosthesis loosening cases is caused by destruction of the acrylic cement being the binder which fixes the mandrel to the bone. The tests demonstrated that the mechanism of transmission of load inside healthy hip joint is very complex. Implantation of the endoprosthesis leads to physiological disturbances in the mechanisms of transmission of load inside the hip joint. Excessive stiffness of the mandrel leads to the load in a closer part of femur, which results in the atrophy of the osseous tissue in the trochanter region being the cause of loosening of the endoprosthesis. In the case of insufficient stiffness of the implant in relation to the bone, the increase in concentration of stresses in proximal part of femur is observed [4].

MATERIAL AND METHODS

Analysis of the structure of polymethyl methacrylate removed from human body revealed that the area of contact of polymethyl methacrylate with the prosthesis is prone to damage, not only due to increased contact stresses resulting from the difference in mechanical strength of prosthesis and PMMA, but also due to uneven and rather random area of adherence (Fig. 1).



Fig. 1. PMMA-implant contact area Rys. 1. Powierzchnia kontaktowa PMMA-implant

Cement mass is not a uniform structure. PMMA surface reveals a dendrite nature of cement combined with porous bone surface. Polymerized cement mass does not have pores to which bone could get in, thus the connection is based on sticking rather than on osseointegration (Fig. 2). Creation of such a complex contact area causes that individual groups of tiny cement 'crystals' have to bear higher load. Uneven shape and dimensions and sharp edges cause intensification of notch effect, which contributes to creation of microcracks, whereas propagation of cracks causes crushing of PMMA.

A number of microcracks can be observed in the analysed surface with the course of time. The crack might then extend and cause chipping of the portions of cement mass. The destruction of the cement coating can be also attributed to a considerable load from the prosthesis on PMMA and that on bone, resulting from human body motor behaviours.



Fig. 2. PMMA-bone contact area Rys. 2. Powierzchnia kontaktowa PMMA-kość

EXPERIMENTAL PROCEDURE

In order to determine the causes of destruction of bone cement in human body, series of numerical and empirical examinations were carried out. Numerical examinations were aimed at determination of the nature of stress in cement mass whereas empirical examinations were carried out in order to determine mechanical strength in bone cement and its application in orthopaedics. Strength testing was carried out with four types of commercial PMMA (pure cement) and PMMA composites with different fillers.

In order to develop a PMMA composite with carbon fibre, four commercial cements popular in orthopaedics, composed of the polymer in the form of the powder of liquid polymer were used: CEMEX RX (Tecres SPA), PALACOS R (Schering - Plough International Inc.), SIMPLEX P (Howmedica Limerick), PALAMED 40 (Biomet Merck).

The concept of modification of polymethyl methacrylate with carbon fibre appeared after analysis of a number of clinical cases, where loosening of the prosthesis and necessity to perform reimplantation was caused by the destruction of the layer of bone cement. Determination of the causes and course of destruction of cement coating is very difficult, thus first stage of investigations encompassed the attempts to determine the nature and level of stress in bone cement at the maximal load (walk- ing phase) using FEM tools. During numerical calculations, the bone cement was modelled as a linear-elastic element with isotropic strength properties. Thickness of the layer of cement which binds the femoral bone with the prosthesis changed according to the previous assump- tions [5, 6]. For the part close to the bone, the layer of cement was

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3÷5 mm whereas in the farther part in the area of the stem tip this value amounted to $2\div3$ mm [7]. Discrete model of bone-cement-implant arrangement was made of 84435 elements with cubicoid shape of 3D Solid type, determined on the basis of 81171 nodes, whereas PMMA model was determined on the basis of 16640 elements. For the bone cement the mechanical properties were assumed according to the results of empirical investigations - for the cement with the highest strength. Numerical tests were conducted on the basis of ADINA System 8.1 software using finite elements method [8]. Model of load for the femoral bone was assumed according to the active load model with consideration of human walk profile [9].

RESULTS AND DISCUSSION

Based on the analysis, it was found that compressive stresses occur the most often in the layer of bone cement loaded during walking phase. Maximal value of reduced stresses amounts - for the cement with the highest strength to $\sigma_{max} = 7.521$ MPa, and the nature of stress in PMMA is presented in Figure 3a, whereas the value of stresses in the area of fixing of the acetabulum in the pelvis are nearly seven times higher and reach $\sigma_{max} = 51.13$ MPa, which is presented in Figure 3b.



Fig. 3. Map of reduced stress σ [MPa] for polymethyl methacrylate Rys. 3. Mapa naprężeń zredukowanych polimetakrylanu metylu

Nature of load in prosthesis resulting from everyday activity such as walking, sitting or climbing stairs proves that the load is of dynamic (impact) nature.

The use of methods of modification of PMMA with Al_2O_3 or with bone-based materials causes reduction in mechanical strength of bone cement. In consequence, load in the modified cement mass resulted in rise in stress value. As an alternative method of modification of bone cement, an attempt was made to develop a PMMA/carbon fibre composite. Basic strength testing had to be carried out in order to determine the effect of PMMA modification with carbon fibre. Due to the profile of the load PMMA bears during use, two fundamental strength tests were made i.e. compressive strength and impact strength testing. In order to compare strength parameters in the modified cement mass to popular fillers, compression test was carried out for 10% of fillers (Fig. 4).



Fig. 4. Compressive strenght Rc [MPa] for bone cements with different fillers

The conducted investigations revealed that combination of PMMA composite with carbon fibre causes rise in compressive strength in all the bone cements. In the case of Pamaled 40, with Rc = 126 MPa, development of PMMA +10% cement fibre composite allowed for obtain- ing the value of Rc = 134 MPa. Compressive strength in Palacos R and Simplex P cements after addition of carbon fibres increased, respectively: Rc = 88 MPa to Rc = 93MPa, and Rc = 86 MPa to Rc = 91 MPa. In the case of Cemex RX, for which the compressive strength was lowest (Rc = 82 MPa), preparation of the composite with 10% fraction of carbon fibre caused, in the selected group, rise in strength to Rc = 87 MPa. The obtained results revealed that modification of PMMA with carbon fibre, unlike other fillers where mechanical parameters decreased, resulted in rise in PMMA compressive strength from 5 to 7%, depending on the cement type. Static compression testing revealed that rise in percentage fraction of carbon fibre results in an increase in PMMA strength. It was also found that optimal concentration of filler is 30%. Commercial bone cements, on the basis of

which the composite with 30% fraction of carbon fibre was developed, showed enhanced compressive strength Rc [MPa] from 20 to 24% on average, depending on cement type (Fig. 5).



Fig. 5. Compressive strength *Rc* [MPa] for bone cements and well as cements with carbon fibre

Rys. 5. Wytrzymałość na ściskanie *Rc* [MPa] wybranych cementów kostnych oraz z włóknem węglowym

Bone cement is a brittle material, which in the case of dynamic load of prosthesis on bone cement and then on the bone, might cause an appearance of microcracks which will turn into bone cement chipping. The developed composite is characterized by a rise in impact strength in cement mass (Fig. 6), which will also have a positive impact on lifetime and reliability of bone-implant connection.



Fig. 6. Impact strength in PMMA/carbon fibre composite Rys. 6. Udarność kompozytu PMMA/włókno węglowe

As was observed based on the examination by means of Charpy's method, PMMA/carbon fibre composite causes rise in impact strength by $20\div26\%$ in relation to pure bone cement, depending on the type of PMMA used. The highest rise in impact strength was observed for polymethyl methacrylate Palamed 40, whose impact strength improved from KC = 5.2 kJ/m² to KC = 6.6 kJ/m², which accounts for 126% of the value for pure cement. The lowest rise in PMMA + carbon fibre resistance to dynamic load was observed for Simplex P cement, whose KC value rose by 20% (from the value of KC = 3.0 kJ/m² to KC = 3.6 kJ/m²). In the case of Palacos R and Cemex

Rys. 4. Wytrzymałość na ściskanie *Rc* [MPa] cementu kostnego z różnymi wypełniaczami

RX, impact strength after modification rose, respectively, from $KC = 3.3 \text{ kJ/m}^2$ to $KC = 4.1 \text{ kJ/m}^2$ for Palacos R (+24%) and from $KC = 2.8 \text{ kJ/m}^2$ to $KC = 3.4 \text{ kJ/m}^2$ for Cemex RX (+22%).

The tests were performed by means of DMA 242 device by NETZSCH® with the holder for three-point free bending in the form of a beam. The investigations were carried out for the selected cement with the highest strength. The samples were subjected to vibrations with different frequency and constant amplitude within the whole heating range of 20 to 145°C, with particular consideration of the body interior temperature ranged from 35 to 42°C.

Figure 7 presents the changes in storage modulus E'and mechanical loss factor $tg \delta$ vs. temperature. The value of storage modulus E' is constant in wide range of temperatures (20 to 90°C). The maximum storage modulus E'for the sample with 10 Hz and 1 Hz displacement frequency amounted to 1600 and 1400 MPa, respectively. Dynamic mechanical properties of PMMA/carbon fibre were also evaluated in the investigations.



Fig. 7. DMTA thermogram for the material with the highest storage modulus E' and the mechanical loss factor

Rys. 7. Termogram DMTA dla materiału o najwyższym module zachowawczym E' oraz współczynniku stratności mechanicznej

The focus in the analysis of the presented results should be on the functional range within the temperatures of from 35 to 42°C, since this material will not be affected by higher temperatures while placed in human body. However, from the standpoint of behavior of this material, information about changes within the whole range of temperatures is necessary.

It is remarkable that the time of polymerization for the composite mass of PMMA/carbon fibre is analogous to the time of polymerization for pure cement. This fact is very essential since shortening of time of polymerization would reduce the time an orthopaedist have for implanting the prosthesis in the bone, whereas elongation of the time of polymerization would cause that the implanted prosthesis might move inside the bone and lose its axial arrangement in marrow cavity.

Additional advantage of PMMA/carbon fibre composite is creation of the developed surface and filing the porous structure of the bone with cement (Fig. 8a) whereas in the contact area of PMMA/prosthesis the adherence area can be observed throughout the whole surface. Therefore one can expect that PMMA 'flows' on carbon fibre, which allows for even distribution of the cement (Fig. 8b).



Fig. 8. PMMA/carbon composite fibre surface Rys. 8. Powierzchnia kompozytu PMMA/włókno węglowe

CONCLUSIONS

As can be concluded from the investigations, PMMA/ carbon fibre composites are a perfect alternative for commercial bone cements. PMMA/carbon fibre composite is characterized by improved strength parameters as compared to commercial pure cements. Carbon fibre, as a biocompatible material, is widely used in medicine while improvement in strength parameters and reduction in percentage PMMA fraction in cement coating connecting prosthesis with the bone makes it a perfect filler.

The results of the presented investigations revealed that PMMA/carbon fibre composite shows higher compressive strength Rc [MPa] as compared to pure cement (by approx. $20\div24\%$) whereas the resistance to dynamic load KC [kJ/m²] rises in relation to pure cement by ca. $20\div26\%$. Due to a complex state of load in cement during walking, improvement in these properties can limit occurrence of microcracks and minimize chipping of ce-

ment particles, which should lead to enhanced lifetime of bone cement in the body. Obtaining of a composite with improved strength parameters in relation to pure cement causes reduction in stress in cement mass. It should also be noted that application of fillers in the form of carbon fibre allowed for obtaining of a porous surface which, in consequence, allows for osseointegration of the composite while PMMA/prosthesis contact area takes considerably larger area than in the case of pure cement. The fact that the use of the filler causes reduction in the percentage of polymer and toxic monomer in the marrow cavity is also of great importance as it can lead to reduction in toxicity of the cement mass and accelerate healing.

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