MECHANICAL AND STRUCTURAL CHARACTERISTICS OF p(HEMA) HYDROGEL FOR LUMBAR DISC PROSTHESIS

Iuliana Rotaru[1], Bogdan Istrate[2], Marcelin Benchea[3], Ana Tufescu[4], Dumitru Olaru[5]

[1-5] *"Gheorghe Asachi" Technical University of Iasi, Faculty of Mechanical Engineering, Department of Mechanical Engineering, Mechatronics and Robotics, Iasi, Romania,*
e-mail: rotaruiuliana@tuiasi.ro.

Abstract: *Poly(2-hydroxyethyl methacrylate) - p(HEMA) - is a biocompatible polymer that forms a hydrogel in water. Mechanical properties, water content and oxygen permeability of p(HEMA) hydrogel can be controlled by cross-linking rate or by copolymerization; that is why p(HEMA) hydrogels have multiple biomedical applications. The objective of this paper was to analyse a biomaterial made of p(HEMA) for its use as intermediate element within intervertebral disc prosthesis on lumbar spine. The p(HEMA) biomaterial used in this study was in the form of transparent lenses. The mechanical and structural characterization of polymer based on p(HEMA) has included surface topographic analysis, determination of swelling rate and compactness, X-ray diffraction, optical and electron microscopy analysis, indentation tests and preliminary compression tests.*

Keywords: *biomaterial, hydrogel, mechanical properties, p(HEMA).*

1. Introduction

Polymers and copolymers based on 2-hydroxyethyl methacrylate (HEMA) are hydrophilic materials; after absorption of water to a state of equilibrium, they behave as elastic gel and in this form they are known as hydrogels [1]. Polymers are high molecular weight chemical substances produced by polymerization reaction from a large number of low molecular weight identical molecules called monomers. Copolymers are macromolecular compounds obtained by the mixed polymerization of two or more different monomers (copolymerization). The first and the most successful application of hydrogels based on HEMA was in the ophthalmology for hydrophilic contact lenses with excellent clinical results. Subsequently, it was successfully used to manufacture artificial lens and artificial cornea. Today HEMA hydrogels have many biomedical applications, such as bone replacement material or artificial cartilage [1,3].

In dry state, poly(2-hydroxyethyl methacrylate) - p(HEMA) – is a hard and brittle material. After swelling, it becomes soft and flexible and can be easily cut with a scalpel or scissors. Transparent hydrogels of p(HEMA) allow diffusion of liquids, suggesting the existence of a porous structure. Mechanical properties, water content and oxygen permeability of p(HEMA) hydrogel can be controlled by cross-linking rate or by copolymerization; that is why p(HEMA) biocompatible hydrogels have multiple biomedical applications [1,4].

The purpose of this paper was to characterize a biomaterial made of p(HEMA) for its use as intermediate element within intervertebral disc prosthesis on lumbar spine. The p(HEMA) biomaterial used in this study was in the form of transparent lenses. The mechanical and structural characterization of polymer based on p(HEMA) from this study has included surface topographic analysis, determination of swelling rate and compactness, X-ray diffraction, optical and electron microscopy analysis, indentation tests and preliminary compression tests.

2. Materials and methods

The biomaterial based on p(HEMA) hydrogel which was used in this study was in the form of preforms of transparent artificial lens (Corneal Industrie, France) (Fig. 1). This polymer was chosen because, in addition to being biocompatible, fully hydrated is a hydrogel used as artificial cartilage with properties similar to natural articular cartilage. Maximum degree of swelling of

these lenses is reached after a time of 48h in distilled water or physiological serum [2].

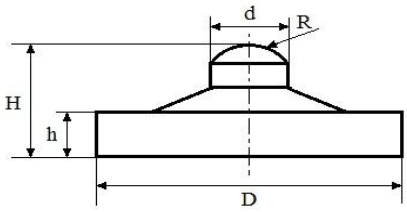


Figure. 1: *p(HEMA) transparent lens – lateral view*

Dimensions of dry and wet p(HEMA) lenses have been measured using a micrometer with 0.001 mm accuracy which can measure sizes from 0 to 25 mm. Dimensions of p(HEMA) lens are shown in Table 1. The mass of dry and wet lenses have been measured on AGN 200 analytical balance with an accuracy of 0.0001 g: 0.259 g and respectively 0.338 g.

Table 1: *The dimensions of p(HEMA) lens*

Material	Height ***H*** **(mm)**	Height ***h*** **(mm)**	Diameter ***D*** **(mm)**	Diameter ***d*** **(mm)**
Dry p(HEMA)	3.005	1.218	12.410	6.046
Wet p(HEMA)	3.315	1.310	13.691	6.672

Roughness and radius of dry and wet convex lenses have been determined using Form Talysurf Intra 50 roughness tester which is in Laboratory of Machine Elements, IMMR Department, Faculty of Mechanical Engineering, Iasi, Romania. The conical detector which has a diamond head (with radius of 2 μm and angle of 90º) has been used for these measurements.

The maximum degree of hydrogel swelling has been calculated using the formula:

$$Q_{\max} = \frac{m - m_0}{m_0} \cdot 100\,(\%). \tag{1}$$

where: m_0 - mass of dry hydrogel (g) and m - mass of swollen (hydrated, wet) hydrogel (g).

The effective volume of pores from swollen lens (volume of pores that allow passage of fluid) can be determined by determining the absorption of fluid (liquid saturation). The volume of liquid (physiological serum) absorbed V_{serum}, that is the effective volume of pores V_p from hydrated polymer, was determined with the formula:

$$V_{serum} = \frac{m_{serum}}{\rho_{serum}} = \frac{m - m_0}{\rho_{serum}}. \tag{2}$$

where: m_{serum} – mass of physiological serum and ρ_{serum} – density of physiological serum (1g/cm^3).

The total volume of hydrated lens geometry is determined as follows: lens is divided into four regular simple geometric 3D shapes (a spherical cap, a small cylinder, a truncated cone and a big cylinder, Fig. 1), the volume of each shape is determined with geometry formulas and then the total volume V_t is determined summing the four volumes which were determined previously.

The porosity is defined as the totality of pores contained in a material and is given by the ratio between the pore volume and the total volume of the material:

$$P = \frac{V_p}{V_t} \cdot 100\,(\%). \tag{3}$$

The compactness is the degree of filling of a body with solid material and is expressed as a ratio between the real volume (non-porous) and the total volume of material [5]:

$$C = \frac{V_t - V_p}{V_t} \cdot 100\,(\%). \tag{4}$$

The connection between porosity and compactness is given by the formula:

$$C = 1 - P. \tag{5}$$

Structural analysis by X-ray diffraction (XRD) was performed using X'Pert PRO MRD diffractometer from the Laboratory of Materials Study, IMMR Department, Faculty of Mechanical Engineering, Iaşi, Romania.

The structure of p(HEMA) lens has been examined by optical microscopy (OM) and scanning electron microscopy (SEM). The surface of dry and wet spherical lenses has been inspected with optical microscope IM 7000 which is located in the Laboratory of Materials Study, IMMR Department, Faculty of Mechanical Engineering, Iasi, Romania. The SEM investigation of dry lens has been performed using scanning electron microscope Quanta 200 3D that is located in the same laboratory. With SEM it can be reveal only the convex surface morphology of dry lens at a great resolution because the procedure does not allow measurement of wet materials.

Microindentation tests have been performed using the microtribometer CETR UMT-2 (Universal Materials Tester) which is in the

Laboratory of Tribology, IMMR Department, Faculty of Mechanical Engineering, Iasi, Romania. With indentation tests, the modulus of elasticity and microhardness have been obtained for dry and hydrated p(HEMA) convex lenses.

Microindentation tests have been performed using the sensor of 0.2 - 20 N with a resolution of 1 mN and a diamond conical indenter at 120º and spherical tip radius of 200 µm. A capacitive sensor was used to measure the depth of indenter penetration. The scheme of indentation test using the tribometer is shown in Fig. 2.

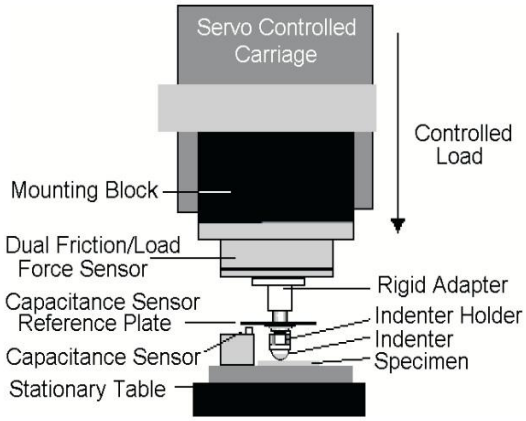


Figure 2: *Microindentation test with microtribometer CETR UMT-2 [6]*

Preliminary compression tests have been realised in order to study the rigidity and the damping capacity of dry and wet p(HEMA) hydrogel. The tests have been performed using the same microtribometer which was adapted with a circular plane indenter of 20 mm diameter (Fig. 3). Preliminary compression tests have been performed at a constant speed of 0.1 mm/s and with a maximum displacement of 0.5 mm in dry and wet p(HEMA) samples.

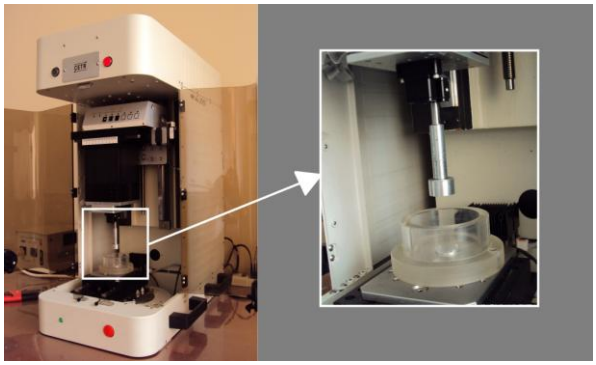


Figure 3: *Experimental setup CETR UMT-2*

3 Results and discussions

3.1 Surface topographic analysis

Several roughness measurements have been executed; the mean values of this parameter are listed in Table 3. Fig. 4 shows an example of topographic analysis for dry and hydrated convex lenses obtained with Taylor Hobson roughness tester. From Tables 1 and 3, it can be observed that not only the size and mass of the hydrogel changes due to fluid absorption, as expected, but the surface roughness of spherical cap.

Table 3: *Radius and roughness of p(HEMA) convex lens*

Material	Radius R (mm)	Roughness R_a (µm)
Dry p(HEMA)	7.967	0.0334
Wet p(HEMA)	8.775	0.0264

3.2 Swelling rate and compactness

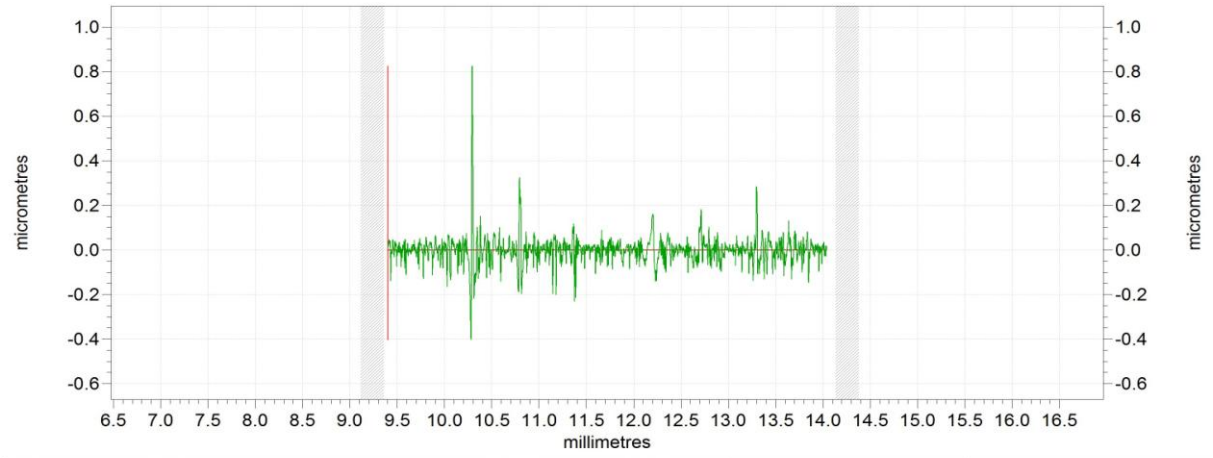
Maximum degree of p(HEMA) swelling was of 30.5% and the liquid content from hydrated lens was 0.079 g. The pore volume from wet lens was of 0.079 cm^3. Fig. 1 have presented only the dimensions which have been measured accurately (with micrometer), the others being approximated from the profile analysis using roughness tester. The total volume of hydrated lens geometrically determined was 0.272 cm^3. Also, the values of effective porosity and compactness of p(HEMA) hydrated lens were 29% and respectively 71%.

3.3 XRD analysis

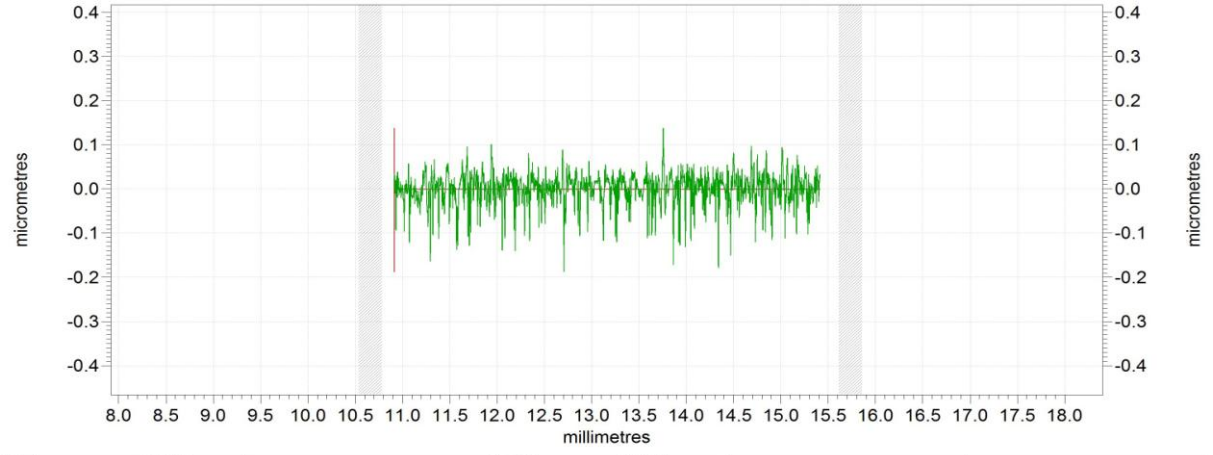
XRD analysis has showed that both dry and wet p(HEMA) lens has a amorphous structure, resulting a diffractogram with three peaks unclear contoured to 18.5º, 32º and 42º 2Theta (Fig. 5).

3.4 OM and SEM

Because of the lens transparency, OM images have shown a closed porous structure and a number of concentric circles on the inferior lens surface; these concentric circles have resulted from lens processing (Fig. 6). Also, as OM images, SEM images indicated closed pores, concentric circles and the centre point which have resulted from processing of the inferior surface (Fig. 7).



Radius	7.9675	mm			Rz(JIS)	0.0808	µm						
Ra	0.0368	µm	Rq	0.0646	µm	R3z	0.0555	µm					
Rsk	3.2207		Rku	43.5456		R3y	0.1029	µm					
Rp	0.0897	µm	Rv	0.1108	µm	Rz(DIN)	0.2005	µm					
RLo	4.6400	mm	Rt	1.2277	µm	Rmr(c) (1)	53.78	%	Height (mean)	0.000	µm		
Rlq	26.28	µm	Rdq	0.88	°	RHSC (1)	176	peaks	Height (mean)	0.000	µm		
Rda	0.56	°	Rz	0.2005	µm	Rdc (1)	1.2277	µm	mr1%	0.0	%	mr2%	100.0 %
RS	16.17	µm	RSm	140.61	µm	Rmr (1)	50.00	%	offset	0.000	µm	mr%	50.00 %
Rln	4.6400	mm	Rc	0.2645	µm	RVo (1)	0.0020	vol	offset	0.000	µm	mr%	50.00 %
Diameter	15.9349	mm				RPc (1)	469.8	pks/cm	Height (mean)	0.000	µm	bandwidth	0.00 µm



Radius	8.7752	mm			Rz(JIS)	0.1361	µm						
Ra	0.0260	µm	Rq	0.0355	µm	R3z	0.1274	µm					
Rsk	-0.9412		Rku	5.4332		R3y	0.1770	µm					
Rp	0.0767	µm	Rv	0.1340	µm	Rz(DIN)	0.2107	µm					
RLo	4.4998	mm	Rt	0.3246	µm	Rmr(c) (1)	56.10	%	Height (mean)	0.000	µm		
Rlq	18.24	µm	Rdq	0.70	°	RHSC (1)	184	peaks	Height (mean)	0.000	µm		
Rda	0.50	°	Rz	0.2107	µm	Rdc (1)	0.3246	µm	mr1%	0.0	%	mr2%	100.0 %
RS	15.08	µm	RSm	38.38	µm	Rmr (1)	50.00	%	offset	0.000	µm	mr%	50.00 %
Rln	4.5000	mm	Rc	0.1153	µm	RVo (1)	0.0015	vol	offset	0.000	µm	mr%	50.00 %
Diameter	17.5504	mm				RPc (1)	488.9	pks/cm	Height (mean)	0.000	µm	bandwidth	0.00 µm

Figure 4: *Microtopographic analysis of dry (top) and wet (bottom) p(HEMA) convex lens*

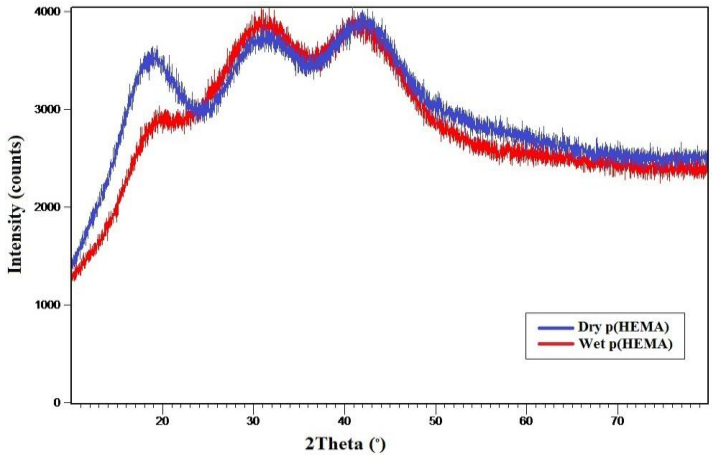


Figure 5: *Diffractogram for p(HEMA) lens*

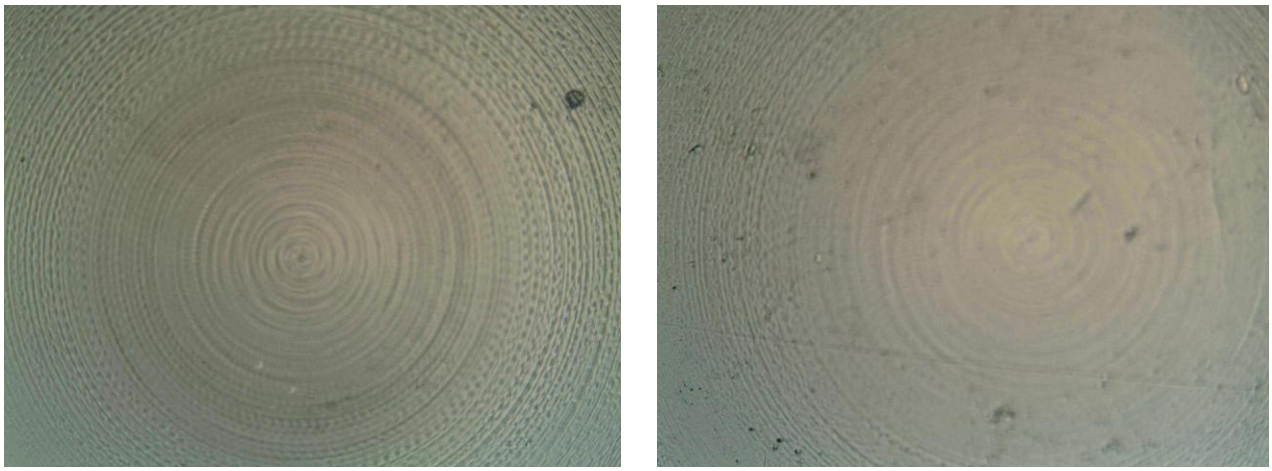
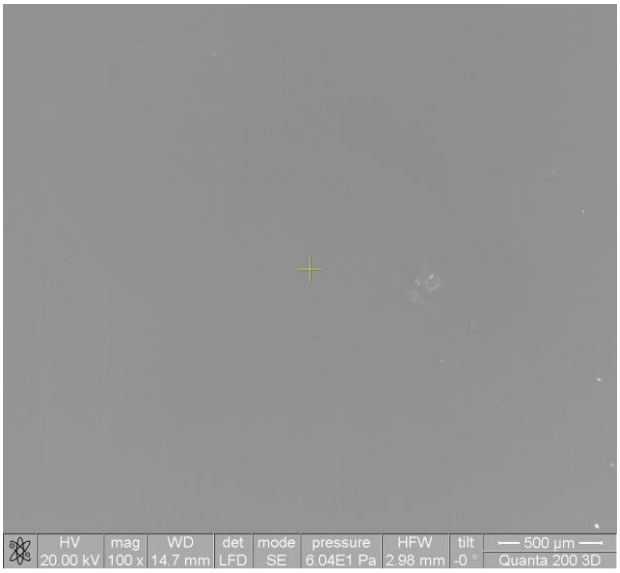
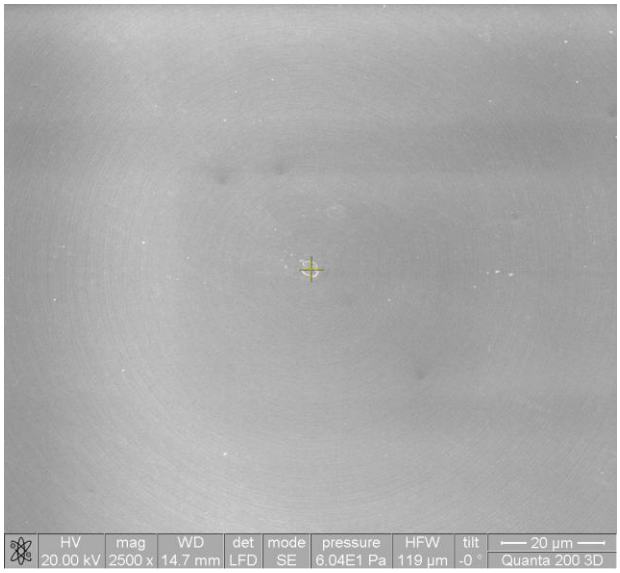


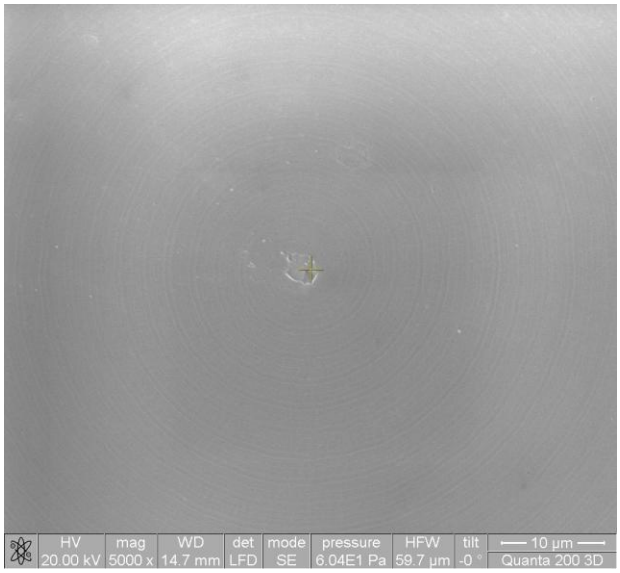
Figure 6: *The structure of dry (left) and wet (right) p(HEMA) convex lens – OM images with 20X magnitude*



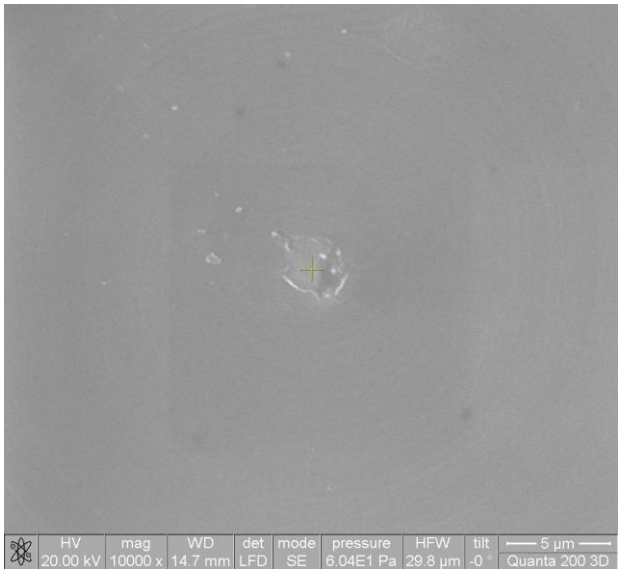
100X



2500X



5000X



10000X

Figure 7: *SEM images for dry p(HEMA) convex lens*

3.5 Microindentation

Force-displacement curves corresponding for indenter tip pressing and rising which were obtained from microindentation tests and the values of microhardness and modulus of elasticity are shown in Fig. 8 and respectively in Table 4.

Table 4: *Elasticity modulus and Rockwell hardness from microindentation tests*

Material	Elasticity modulus *E* (GPa)	Rockwell hardness *HRC* (GPa)
Dry p(HEMA)	2.18	0.205755
Wet p(HEMA)	0.004	0.001107

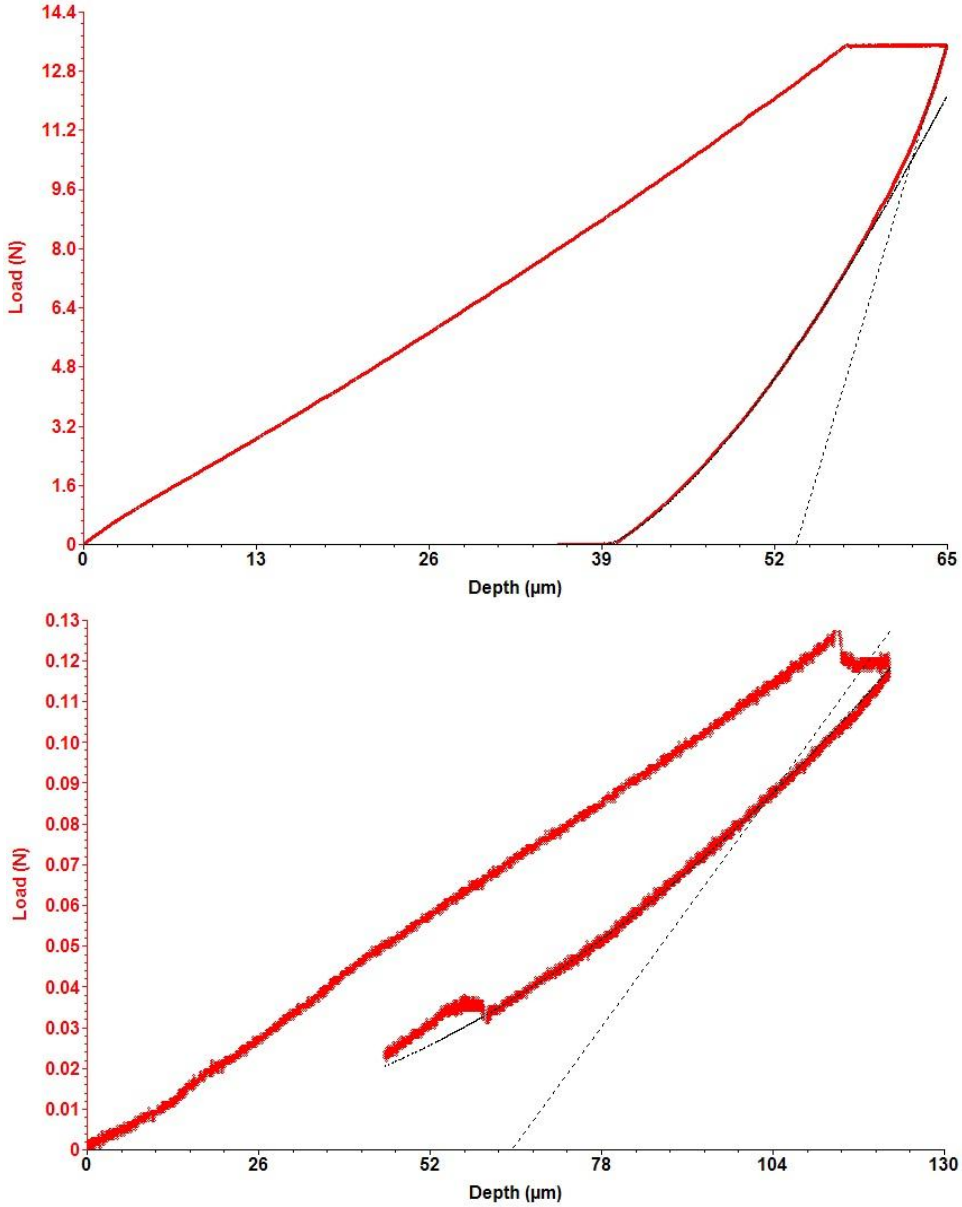


Figure 8: *The results of microindentation tests for dry (top) and wet (bottom) p(HEMA) convex lens*

Microindentation test was performed at a total load of 15 N in case of dry lens, while in case of hydrated lens the total load was 0.2 N because the hydrated p(HEMA) hydrogel is soft and flexible. For hydrated lens, when the load was higher than 0.2 N, the elasticity modulus and microhardness cannot be determined from force-displacement curve because the curve shape for the indenter rising (decompression) does not allow it (the curve cannot be approximated and thus its slope cannot be determined). While pressing the indenter tip in hydrated lens at high loads, the penetration depth initially has recorded a rapid increase with the load and then a slow increase. This indicates that the microindentation method is not suitable for soft

materials such as hydrogels; a more adequate method is nano-scale indentation.

3.6 Preliminary compression tests

The results of preliminary compression tests, which was performed at a constant speed of 0.1 mm/s and with a maximum displacement of 0.5 mm in dry and wet p(HEMA) convex samples, were presented in Fig. 9-10. As expected, the compressive load has increased with compressive depth and time.

Two third order polynomials have been fitted to the data points of load-depth curve (Fig. 9). The third order polynomials for dry and wet p(HEMA) convex lenses are the follows:

$$\left.\begin{aligned} F_{dry} &= -4.519x^3 + 11.472x^2 + 19.994x + 0.084 \\ F_{wet} &= -6.225x^3 + 11.161x^2 + 2.674x + 0.012 \end{aligned}\right\} \quad (6)$$

The load-depth curves are non-linear ascending for both dry and wet convex lenses (Fig. 9). Consequently, dry and wet p(HEMA) lenses have a variable rigidity. The rigidity K is defined as the ratio between the elementary load F and the elementary displacement (depth) x:

$$K = \frac{dF}{dx}. \quad (7)$$

The rigidity of the dry and wet p(HEMA) convex lenses, K_{dry} and K_{wet}, are determined using equations (6) and (7):

$$\left.\begin{aligned} K_{dry} &= -13.559x^2 + 22.944x + 19.994 \\ K_{wet} &= -18.676x^2 + 22.322x + 2.674 \end{aligned}\right\} \quad (8)$$

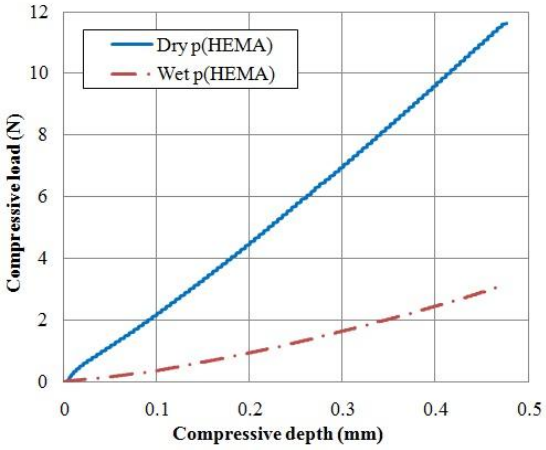


Figure 9: *Variation in compressive load with compressive depth*

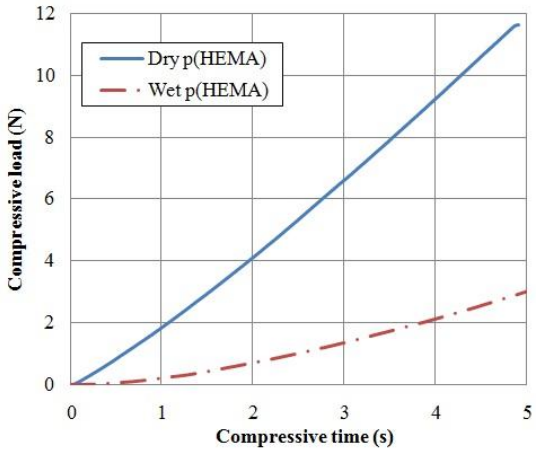


Figure 10: *Variation in compressive load with compressive time*

Using equations (6) and (8), rigidity-depth and rigidity-load curves are plotted for both dry and wet p(HEMA) convex lenses and shown in Fig. 11 and Fig. 12. It can be noted that the rigidity has increased with compressive depth and load.

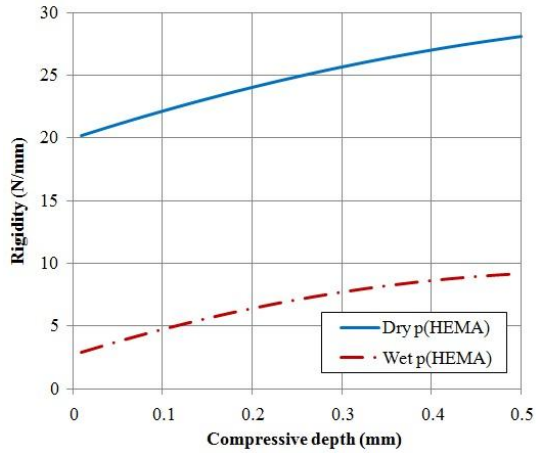


Figure 11: *Variation in rigidity with compressive depth*

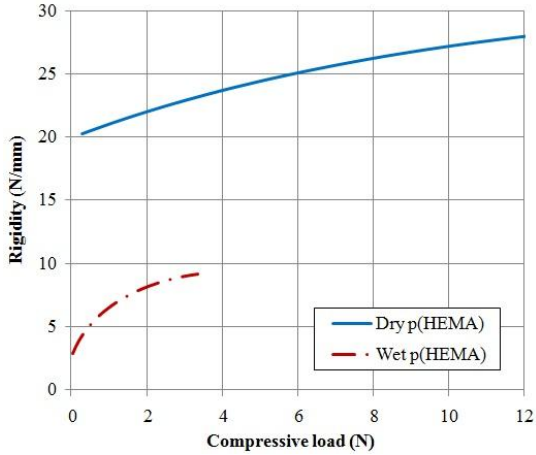


Figure 12: *Variation in rigidity with compressive load*

The damping capacity of dry and wet p(HEMA) convex lenses can be evaluated from load-time curves (Fig. 10). Thereby, at the same compressive load, for example 2 N, the time is 1.1 s for dry lens and 3.9 s for wet lens. The sample which records the greatest time at the same load is considered to have a better damping capacity than the others. Therefore, the wet p(HEMA) have a much better damping capacity than in dry state and could assure an adequate mechanical shock absorption as intermediate element within intervertebral disc prosthesis on lumbar spine.

It is well known that the mechanical and structural characteristics of hydrogels based on p(HEMA) can be controlled to become very close to the natural joint cartilage [1-4] (particularly to the natural intervertebral disc) which presents a good damping capacity; for example, the values of the elasticity modulus, oxygen permeability and water content can be controlled to be similar [1,3]. In the author's opinion these p(HEMA) hydrogels that are used for contact lenses and artificial cartilage could also be used in total lumbar disc arthroplasty as a intermediate element to assure a mechanical shock absorption.

4 Conclusions

The purpose of this paper was to analyse a p(HEMA) hydrogel for its use as intermediate element within total disc prosthesis on lumbar spine. The p(HEMA) biomaterial used in this study was in form of transparent lenses. The mechanical and structural characterization of dry and wet polymer based on p(HEMA) has included surface topographic analysis, determination of swelling rate and compactness, X-ray diffraction, optical and electron microscopy analysis, indentation tests and preliminary compression tests. The mechanical and structural characteristics have indicated that p(HEMA) hydrogels could also be used in lumbar disc arthroplasty as a mechanical shock absorption material.

Acknowledgements

This paper was realised with the support of POSDRU CUANTUMDOC "DOCTORAL STUDIES FOR EUROPEAN PERFORMANCES IN RESEARCH AND INNOVATION" ID79407 project funded by the European Social Fund and Romanian Government.

References

[1] Bostan L. E., *Materiale polimerice cu hidrofilie controlată cu aplicaţii în ingineria tisulară a acartilajului articular*, PhD thesis, Faculty of Chemical Engineering and Environmental Protection, Iasi, Romania, 2011.

[2] Corneci M. C., *Fonctionnement tribologique des articulations synoviales pathologiques: Role des interfaces phospholipidiques*, PhD thesis, INSA de Lyon, France, 2012.

[3] Tomar N., Tomar M., Gulati N. and Nagaich U., *pHEMA hydrogels: Devices for ocular drug delivery*, International Journal of Health & Allied Sciences, 1(4), 224-230, 2012.

[4] Trunfio Sfarghiu A.-M., *Modele bio-tribologique des articulations. Role mecanique et physicochimique des assemblages moleculaires du fluide synovial*, PhD thesis, INSA de Lyon, France, 2006.

[5] Jantschi L. and Bolboacă S., *Analiză chmică şi instrumentală aplicată*, Ed. AcademicDirect, ISBN 973-86211-6-X, Cluj-Napoca, Romania, 2003.

[6] http://www.cetr.com