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MECHANICAL CHARACTERIZATION OF PMMA ACRYLIC BONE CEMENTS WITH A NONLINEAR VISCOELASTIC MODEL

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Abstract. A nonlinear analytical viscoelastic model is proposed for time-dependent mechanical characterization of PMMA bone cements. A pseudo-Burger rheological model is presented and its characterization is discussed, considering instantaneous, retarded and long time viscous effects with nonlinear dependence of stress. The proposed model is fitted to available creep data of commercial PMMA bone cements. The obtained results show that the proposed model can be used to model time-dependent creep effects of PMMA bone cements for different stress levels while describing experimental data with mechanical interpretation of the material behaviour.

1 INTRODUCTION

The Polymethylmethacrylate (PMMA) Acrylic Bone Cement (ABC) is a biomaterial widely used in orthopedics, such as fixation of prosthetic implants, remodeling osteoporotic and neoplastic and vertebral fractures repair (Magnan et al., 2013). Its solid component (based mainly on PMMA) is usually mixed with a liquid, based on Methylmethacrylate (MMA). A hardened cement paste is then obtained through the polymerization reaction of the monomer (Duey et al., 2012). After the curing process, the cement becomes solid and exhibits mechanical properties suitable for implant fixation in arthroplasty and other applications (von Hertzberg-Boelch et al., 2022).

Several time-dependent mechanical properties of PMMA ABC such as viscoelastic effects including creep, relaxation and fatigue are key parameters for the applications, since bone cements can be used for very long times (Lee, 2005; Gie et al., 2004). A typical implant can be loaded by millions of cycles along its lifespan, highlighting the importance of failure due to fatigue effects (Köster et al., 2013). Moreover, the instantaneous mechanical properties of PMMA bone cements as well as delayed properties are temperature-dependent (Kuzmychov et al., 2009; Lee et al., 1978, 2002; Yetkinler and Litsky, 1998).

The investigation of time-dependent properties and mechanical behaviour of PMMA bone cements has being done by some authors. In (Köster et al., 2013; Lee et al., 1990), a general mechanical characterization of PMMA is presented. The role of time in the curing process of bone cements is investigated in (Kuzmychov et al., 2009). The effect of the mixing process of the material is investigated in (Norman et al., 1995), showing that hand-mixed and vaccun-mixed PMMA bone cements exhibit different creep behaviour.

Mechanical models and analysis can be developed using the data obtained experimentally and used to predict the behaviour of the material. An analytical model combined with numerical simulation using the Finite Element Method is presented in (Stolk et al., 2004) to evaluate creep and damage in ABC, crack propagation (Boulenouar et al., 2016) and to simulate leakage through a porous media in (Alenezi et al., 2017). A model for the temperature-dependent mechanical behaviour of PMMA under uniaxial tension is investigated in (Abdel-Wahab et al., 2017) and the development of constitutive equation for modelling PMMA material using viscoelastic models is presented in (Jo et al., 2005).

In this work, a nonlinear analytical viscoelastic model is proposed for mechanical characterization of PMMA bone cements. The developed model is fitted to two commercial PMMA ABC brands: Palacos® R (from Heraeus Medical) and Zimmer Regular (from Zimmer Biomet). The choice of these brands is due to available experimental data used to validate and calibrate the proposed model. The experimental data provided in (Norman et al., 1995) are used for formulation of the nonlinear viscoelastic model. In the latter experimental analysis, creep tests were performed at constant temperature applying different compressive stress levels on both hand and vacuum-mixed specimens. The ability of the proposed model to fit short and long-term viscoelastic behaviour of PMMA cements is presented and discussed, thus showing its relevance to modelling the nonlinear time-dependent mechanical behaviour of the material.

2 EXPERIMENTAL CREEP DATA

The experimental results presented in (Norman et al., 1995) for the viscoelastic creep of Palacos® R and Zimmer Regular acrylic bone cements are adopted in this work for the mechanical characterization of the material. Such experimental results and referred methodology are briefly presented and discussed in this section.

In (Norman et al., 1995), the components of both acrylic bone cements brands were prepared with two different mixing setups: Hand mixed and Vacuum mixed, being referred as H and V group, respectively.

After curing, cylindrical specimens measuring 25.4 mm in length and 15.5 mm in diameter were obtained. The compressive creep tests were performed using an MTS servohydraulic testing machine at constant load corresponding to 14, 30, 40 and 50 MPa stress levels for the specimens of group H and 30, 40 and 50 for group V. The tests were performed at room temperature (25^oC), which is not always the same temperature that the biomaterial would be subjected to in real applications. It is therefore expected that results derived at room temperature can lead to small discrepancies in the real mechanical behaviour (Lee, 2005).

In this work, the average values of instantaneous strain and viscoelastic creep strain at 2, 6, 12 and 24 hours reported in (Norman et al., 1995) are used as reference. The experimental tests were performed up to 24 hours or until a maximum strain of 15% is reached and are summarized in Tab. 1 and Fig. 1, showing the total strain along time for each group of specimens.

Cement Type	Applied Stress [MPa]	Creep Strain [%]				
		0h	2h	6h	12h	24h
Palacos® R (H)	50	2.30	8.60	12.20	-	-
	40	1.60	3.90	5.20	6.70	-
	30	1.10	1.75	2.30	2.80	3.60
	14	0.49	0.59	0.63	0.66	0.72
	10.5	0.40	0.48	0.51	0.53	0.56
Palacos® R (V)	50	2.10	6.60	10.20	-	-
	40	1.20	2.90	3.80	4.50	-
	30	1.00	1.79	2.10	2.30	-
Zimmer Regular (H)	50	2.10	11.70	16.10	-	-
	40	1.90	4.70	6.30	8.00	13.10
	30	1.30	2.30	2.80	3.10	-
	14	0.60	0.80	0.89	0.98	1.10
Zimmer Regular (V)	50	1.80	5.90	8.50	-	-
	40	1.50	3.10	3.70	4.20	-
	30	1.20	1.87	2.14	2.40	-

Table 1: Total strain for different time values (Norman et al., 1995).

3 VISCOELASTIC MODELLING

Considering the experimental data obtained from compressive creep tests, the purpose of the present analysis is to formulate and calibrate a mechanical description of the time-dependent behaviour of PMMA acrylic bone cements within a 1D setting. From a constitutive viewpoint and referring to Fig. 1, a Burgers rheological model might be a reasonable first option for simulating the time-dependent deformation of such materials.

3.1 Burgers Model

Burgers model is a phenomenological model classically used for representing the non-ageing viscoelastic behaviour of polymers. It is built from the association of elastic spring elements and viscous dash-pot elements (Fig. 2). In this linear rheological model, a Kelvin-Voigt component is connected in series with a Maxwell arrangement (Salençon, 2019). The total strain associated

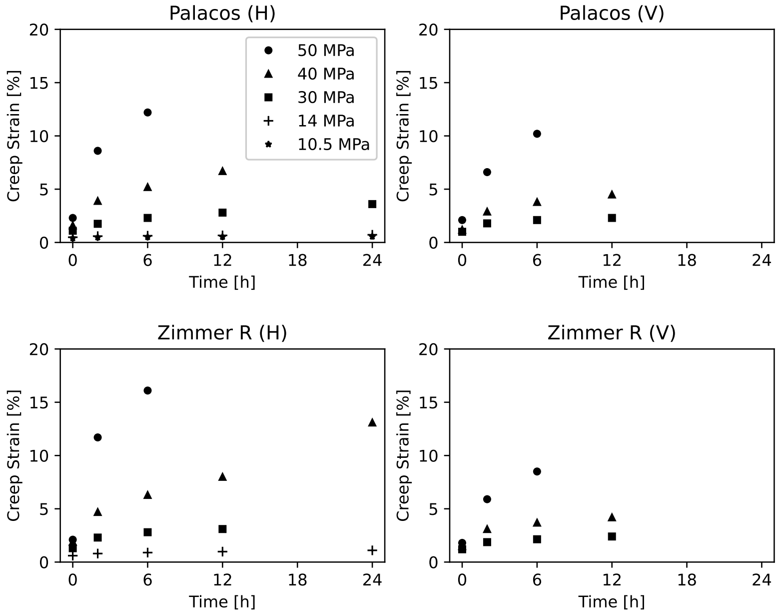

Figure 1: Experimental data.

with a prescribed stress history $t \to \sigma(t)$ reads:

$$\varepsilon(t) = \varepsilon_K(t) + \varepsilon_M(t) \quad (1)$$

where ε_K and ε_M are the strain in the Kelvin-Voigt and Maxwell element, respectively.

Referring to Fig. 2, the state equations of the springs are defined by the elastic parameters E_1 and E_2, whereas those of the dash-pots are defined by the viscosity parameters η_1 and η_2.

The strain in the Maxwell component reads:

$$\dot{\varepsilon}_M(t) = \frac{\dot{\sigma}(t)}{E_2} + \frac{\sigma(t)}{\eta_2} \quad (2)$$

and the Kelvin-Voigt part of strain obeys:

$$\eta_1 \dot{\varepsilon}_K(t) + E_1 \varepsilon_K(t) = \sigma(t) \quad (3)$$

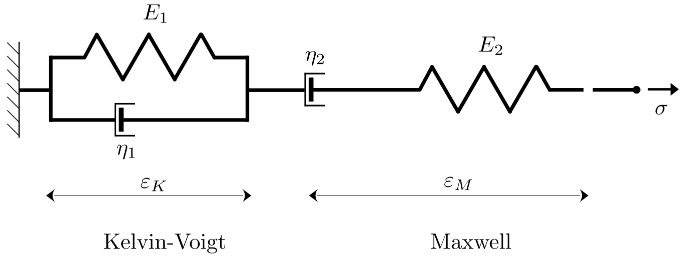

Figure 2: Burgers Model.

Given a stress history $t \to \sigma(t)$, the Maxwell and Kelvin-Voigt strain components ε_M and ε_K can be evaluated from integration of the corresponding governing differential equations Eq.

2 and Eq. 3, together with appropriate initial conditions $\varepsilon_M(t = 0^+) = \sigma(t = 0^+)/E_2$ and $\varepsilon_K(t = 0^+) = 0$. Alternatively, combining Eq. 1, Eq. 2 and Eq. 3 yields the following governing constitutive equation for the total strain:

$$\sigma + \left[\frac{\eta_2}{E_2} + \frac{\eta_1 + \eta_2}{E_1}\right]\dot{\sigma} + \frac{\eta_1\eta_2}{E_1E_2}\ddot{\sigma} = \eta_2\dot{\varepsilon} + \frac{\eta_2\eta_2}{E_1}\ddot{\varepsilon} \tag{4}$$

where the time-dependence of σ and ε is omitted in the above equation.

In the context of a uniaxial compressive creep test in which the loading of the sample is defined by a constant stress level $\sigma(t) = \sigma_0 Y(t)$, where $Y(t)$ is the Heaviside step function at the origin, the strain associated to the linear Burgers model obtaining from solving Eq. 4 is:

$$\varepsilon(t) = J(t)\sigma_0 \tag{5}$$

where $J(t)$ is the creep function of the material in such a loading:

$$J(t) = \left[\frac{1}{E_1}\left(1 - e^{-\frac{E_1}{\eta_1}t}\right) + \frac{1}{E_2} + \frac{t}{\eta_2}\right]Y(t) \tag{6}$$

As mentioned above, the Burgers model appears suitable for modelling the viscoelastic behaviour of PMMA bone cements under compressive creep loading, since it produces both elastic and viscoelastic recovery after unloading as well as permanent residual strains.

3.2 Linear Viscoelastic Modelling Applied to PMMA Bone Cements Mechanical Behaviour

As a first approach, the experimental strain creep data presented in Tab. 1 were used to calibrate the parameters (E_1, η_1, E_2 and η_2) that define the viscoelastic linear Burgers model. The deformation under creep test is fitted resorting to an appropriate least square minimization procedure.

Due to the small number of observed data, the linear response E_2 is first fitted independently and the remaining parameters are found with the nonlinear fitting procedure in order to preserve the instantaneous behaviour of the material. The *NonlinearFit* numerical procedure of Maple from Maplesoft (Maple, 2019) was used for fitting the parameters, which performs a nonlinear minimization of the squared residual of observed data (Bard, 1974). The fitting parameters are summarized in Tab. 2 and plotted in Fig. 3. The fitted correlation coefficients R^2 are all greater than 0.95.

The results presented in Fig. 3 clearly emphasize the dependence of calibrated linear Burgers model on applied stress level σ_0. This observation suggests that a nonlinear Burgers-like model in which $E_i = E_i(\sigma_0)$ and $\eta_i = \eta_i(\sigma_0)$ would be most suitable for describing the creep deformation of PMMA bone cements.

4 NONLINEAR VISCOELASTIC MODELLING

An instantaneous and time-dependent nonlinear mechanical behaviour describing deformation under constant stress is looked for. The observed PPMA ABC behaviour ranges from almost linear bounded creep at small stresses, which can be modelled using Eq. 6, to highly nonlinear unbounded creep at high stresses.

The idea developed herein is to describe creep in uniaxial compression by means of the nonlinear Burgers-like model which is completely defined by parameters $E_i(\sigma_0)$ and $\eta_i(\sigma_0)$ associated with the applied compressive stress σ_0 (Fig. 4).

Cement Type	Applied Stress [MPa]	E_1 [GPa]	E_2 [GPa]	η_1 [GPa·h]	η_2 [GPa·h]
Palacos® R (H)	50	1.111	2.174	0.333	5.556
	40	1.893	2.500	1.976	16.072
	30	3.458	2.703	8.136	44.269
	14	12.648	2.857	15.045	281.842
Palacos® R (V)	50	1.842	2.381	0.656	5.571
	40	2.044	3.333	2.917	35.754
	30	3.274	3.000	4.166	93.815
Zimmer Regular (H)	50	0.676	2.381	0.044	4.545
	40	1.464	2.105	1.711	14.252
	30	2.398	2.308	3.702	65.485
	14	5.946	2.333	8.640	125.500
Zimmer Regular (V)	50	1.784	2.778	0.425	7.694
	40	2.330	2.667	2.565	48.714
	30	4.384	2.500	4.565	69.807

Table 2: Data fitting parameters for the linear Burgers model (Eq. 6).

Instead of operating with the exact expression of $J(t,\sigma_0)$, which would be difficult to compute analytically, we resort as mentioned previously to a simplified framework. In that respect, a pseudo-Burgers model is defined by the following creep function:

$$f(t,\sigma_0) = \left[\frac{1}{E_1(\sigma_0)}\left(1 - e^{-E_1(\sigma_0)t/\eta_1(\sigma_0)}\right) + \left(\frac{1}{E_2(\sigma_0)} + \frac{t}{\eta_2(\sigma_0)}\right)\right] Y(t) \tag{7}$$

The function $f(t,\sigma)$ can therefore be viewed as an approximate expression for the real creep function: $J(t,\sigma_0) \simeq f(t,\sigma_0)$. The relevance an accuracy of such an approximation is beyond the scope of this paper and would not be discussed in the subsequent analysis.

The procedure for calibrating this simplified model consists in evaluating the stiffness and viscosity parameters $E_i(\sigma_0)$ and $\eta_i(\sigma_0)$ that provide a reasonable fit of the experimental data presented in Tab. 1.

4.1 Parameters $E_2(\sigma_0)$ and $\eta_2(\sigma_0)$ of the Pseudo-Kelvin-Voigt Element

The parameter $E_2(\sigma_0)$ is related to the instantaneous elastic mechanical behaviour of the Burgers model. As observed in experimental compression tests of PMMA bone cements (Bogdan et al., 2015; Robu et al., 2022; Kuehn et al., 2005), an almost quadratic dependency of strain is found. Given this suggestion, a parabolic shape form describing the nonlinear stress-strain relation is considered in this work (Helman and Creus, 1975):

$$\sigma = E_{20}\varepsilon(1-\beta\varepsilon) \qquad 0 \le \varepsilon \le 1/\beta \tag{8}$$

where β and E_{20} are constant parameters. A linear behaviour is retrieved from Eq. 8 considering $\beta = 0$ whereas ε can be evaluated from inverting Eq. 8 for the case $\beta \neq 0$:

$$\varepsilon = \frac{1}{2\beta}\left[1 - \sqrt{1 - \frac{4\beta\sigma}{E_{20}}}\right] \tag{9}$$

leading therefore to:

$$E_2(\sigma_0) = \frac{\sigma_0}{\varepsilon} = \frac{2\beta\sigma_0}{1 - \sqrt{1 - 4\beta\sigma_0/E_{20}}} \tag{10}$$

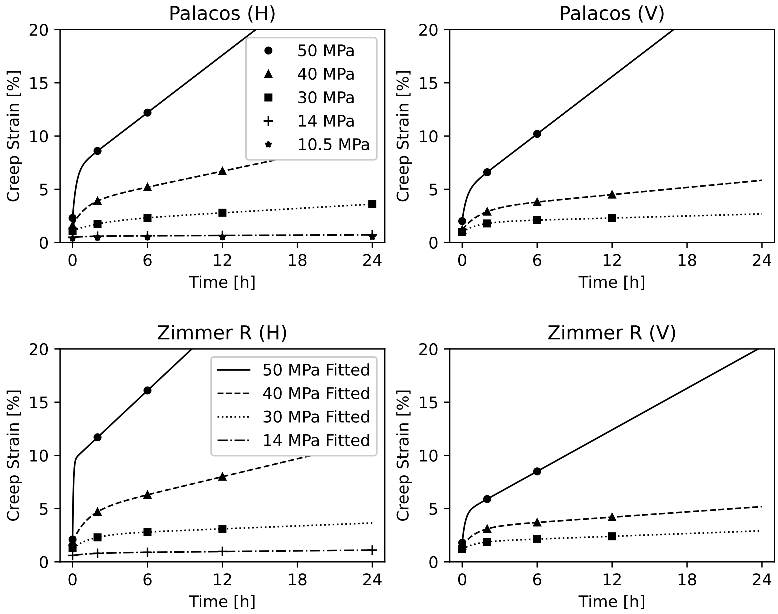

Figure 3: Fitting results obtained for linear viscoelastic Burgers model.

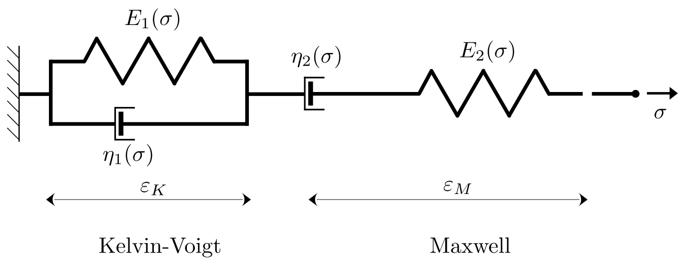

Figure 4: Nonlinear Burgers-like Model.

As regard to the viscosity parameter $\eta_2(\sigma_0)$, the following exponential-dependence should be adopted based on the experimental data:

$$\eta_2(\sigma_0) = \eta_{20} e^{-\beta_2 \sigma_0} \tag{11}$$

where the constant parameters η_{20} and β_2 shall be evaluated from the fitting procedure.

4.2 Parameters $E_1(\sigma_0)$ and $\eta_1(\sigma_0)$ of the Pseudo-Maxwell Element

Based on the experimental data presented in Tab. 2, an exponential and linear dependencies are respectively adopted for the spring stiffness E_1 and dash-pot viscosity η_1:

$$E_1(\sigma_0) = E_{10} e^{-\beta_1 \sigma_0} \tag{12}$$

$$\eta_1(\sigma_0) = \eta_{10}\sigma_0 + \gamma_1 \tag{13}$$

where parameters E_{10}, β_1, η_{10} and γ_1 are calibrated from the fitting procedure. Additionally, in order to enforce positive values for $\eta_1(\sigma_0)$, the following constrain has been considered: $\eta_1(50\ \text{MPa}) = \eta_1^*$, where η_1^* is the experimental value obtained for a stress value of 50 MPa from Tab. 2.

Note that parameter η_1 represents the asymptotic slope of the creep function of the material and the stress-relation should not result in negative values, which would not correspond to the observed experimental behaviour.

4.3 Summarized Results

The previous fitted stiffness and viscosity parameters defining the creep function in Eq. 7 associated with a pseudo-Burgers nonlinear model are combined to describe the creep behaviour of PMMA bone cements:

- Palacos® R (H):

$$f(t,\sigma_0) = \frac{1-\sqrt{1-16.6688\sigma_0}}{26.0058\sigma_0} + t\frac{e^{108.8\sigma_0}}{1243.8} + \frac{1-e^{-t\frac{30.023e^{-67.81\sigma_0}}{-394.611\sigma_0+19.964}}}{30.023e^{-67.81\sigma_0}} \tag{14}$$

- Palacos® R (V):

$$f(t,\sigma_0) = \frac{1-\sqrt{1-19.24375\sigma_0}}{39.05038\sigma_0} + t\frac{e^{141.2\sigma_0}}{7525.6} + \frac{1-e^{-t\frac{7.2956e^{-28.75\sigma_0}}{-185.620\sigma_0+9.937}}}{7.295e^{-28.75\sigma_0}} \tag{15}$$

- Zimmer Regular (H):

$$f(t,\sigma_0) = \frac{1}{2.2681} + t\frac{e^{94.22\sigma_0}}{634.38} + \frac{1-e^{-t\frac{14.054e^{-59.14\sigma_0}}{-222.3196\sigma_0+11.16}}}{14.054e^{-59.14\sigma_0}} \tag{16}$$

- Zimmer Regular (V):

$$f(t,\sigma_0) = \frac{1}{2.684} + t\frac{e^{110.3\sigma_0}}{2443.7} + \frac{1-e^{-t\frac{15.89e^{-44.95\sigma_0}}{-208.4\sigma_0+10.845}}}{15.89e^{-44.95\sigma_0}} \tag{17}$$

In the above expressions, $f(t,\sigma_0)$ is evaluated in $[1/\text{GPa}]$, σ_0 in [GPa] and t in [h]. The creep functions evaluated with the proposed model is shown in Fig. 5 together with experimental data. The continuous lines are referred to the obtained fitted results while the symbols to the experimental data.

5 DISCUSSION AND CONCLUSIONS

The proposed nonlinear viscoelastic model based on a pseudo-Burgers rheological model fits the available time-dependent mechanical behaviour of PMMA bone cements as indicated with the comparison with experimental data shown in Fig. 5. The selected rheological model considers instantaneous, retarded and long time viscous effects which gives the fundamental modelling assumption for the characterization of the material. Moreover, the nonlinear dependence of stress is discussed and developed for each component of the Burgers model. As a result, the developed nonlinear model can be used to predict time-dependent creep functions of PMMA bone cements for different stress levels. The obtained results are presented as equations and graphical information.

The fitted results predict the experimental data better for small values of applied stress. For the selected group of bone cement brands, the model applied to vacuum-mixed cases fits better to the experimental data compared to the hand-mixed cases. Since the material porosity is

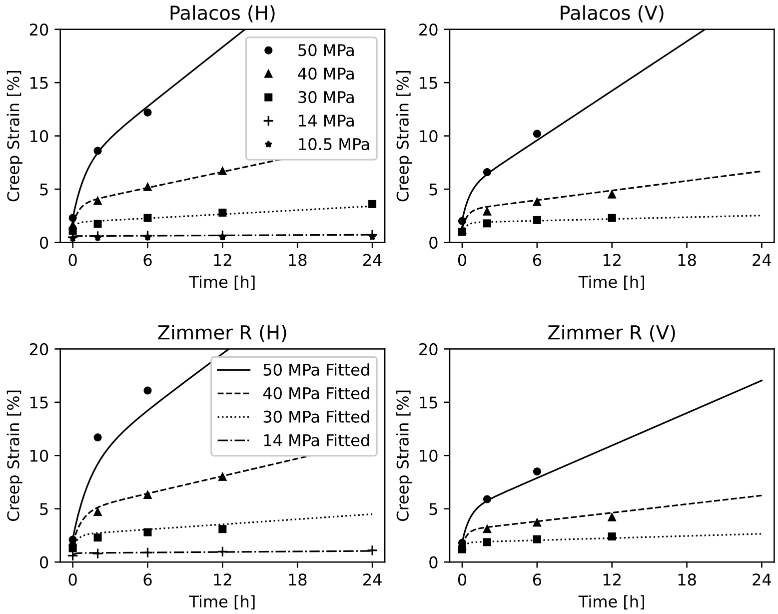

Figure 5: Experimental data and fitted nonlinear model.

reduced with the vaccum-mixed method, an increase of stiffness is also expected. This effect could reduce the nonlinear effect of the material for the investigated stress levels.

Although the small number of available experimental data of variety of applied stresses, the proposed model satisfactorily describes the experimental data with mechanical interpretation of the material behaviour. A larger number of experimental data with more multiplicity of applied stresses should increase the precision of the model.

Finally, the investigation of mechanical behaviour of the PMMA bone cements should be further refined considering *in vivo* conditions and temperature-dependent effects as well as aging effects. Assessment of fatigue properties is also a fundamental issue to be foreseen in the future.

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