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VISCOELASTICITY OF PMMA STUDIED BY NANOINDENTATION

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Abstract: *The paper focuses on microscale viscoelastic properties of Poly-methyl methacrylate (PMMA) measured by a nanoindenter. PMMA serves as a reference homogeneous material suitable for evaluating of theoretical and experimental aspects of the nanoindentation process and computational methods. Viscoelastic constants are calculated with two analytical approaches and compared. Sensitivity to key experimental parameters including maximum load and loading time is assessed.*

Keywords: PMMA, creep, viscoelasticity, nanoindentation.

1. Introduction

Characterization of time-dependent properties, especially creep properties, is of significant importance for structural materials. Creep plays an important role in the analysis of many building composites, including concrete subjected to permanent loads or prestress. The microscale level of materials can be mechanically accessed by nanoindentation (Oyen and Cook (2009)). This technique is capable of measuring material response to indentation of a precise tip pressed to the material. Characterization of composite materials, e.g. cement hydrates (Vandamme and Ulm (2013)), is a difficult task due to the intrinsic heterogeneity of the material and the presence of various pores and defects appearing on different microstructural levels of the composite. Very often, nanoindentation must include a statistical evaluation of the results (Němeček et al. (2023)). Many attempts have been performed to characterize elastic or inelastic properties of composites with nanoindentation. Still, viscoelastic properties are the least studied due to many uncertainties coming from either material heterogeneity or experimental variability in nanoindentation, e.g., various loading protocols or different tips Němeček et al. (2024). The methodology is not standardized yet. High stresses developed under sharp tips also bring a substantial question whether the measured creep stays in the linear regime. Linearity is usually assumed in most studies but not confirmed. This paper concentrates on disclosing some theoretical aspects and influential factors affecting the evaluation of viscoelastic properties from nanoindentation measurements utilizing PMMA which represents a homomogeneous material exhibiting significant creep behavior analogous to other material components like, e.g., C-S-H gels in hydrated cement.

2. Theory and methods

Microscale mechanical properties can be studied with the aid of nanoindentation using sharp or blunt indenters pressed into the sample surface. A general formula for the time-dependent depth of a rigid indenter penetration into a linearly viscoelastic solid is given by (Lee and Radok (1960))

$$h^m(t) = \kappa \int_0^t J(t-\xi)\frac{dP(\xi)}{d\xi}d\xi \tag{1}$$

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where $J(t)$ is the creep compliance function, m and κ are geometric constants (e.g. $m = 2$ and $\kappa = \frac{\pi}{2\tan\alpha}$ for cone or pyramid with α being the equivalent cone angle; $\alpha = 70.3^\circ$ for Berkovich and $\alpha = 42.3^\circ$ for cube corner tip), P is the load and h is the penetration depth (Oyen and Cook (2009)). Assuming a step load, the formula simplifies to

$$h^m(t) = \kappa P_{max} J(t) \tag{2}$$

where P_{max} is the maximum load defined through Heaviside function $H(t)$ as $P(t) = P_{max}H(t)$. The creep function $J(t)$ can take various forms. Assuming a generalized Kelvin model one can write

$$J(t) = \frac{1}{E_0} + \sum_{i=1}^{n} \frac{1}{E_i}(1 - e^{-\frac{t}{\tau_i}}) \tag{3}$$

where $E_{r,0}$ is the instantaneous elastic modulus and E_i, τ_i are elastic moduli and retardation times of respective Kelvin units.

2.1. Accounting for plastic deformation

Several deformation components can be generated in the material by nanoindenter. For PMMA, the loading part is composed of not only elastic and viscous deformations but also non-negligible plastic strains that are a consequence of high stresses under the tip. The holding part contains a viscoelastic response, while the unloading part is assumed to be purely elastic for a fast unloading rate. The amount of individual deformation types depends strongly on the acuity of the indentation tip. While flat or spherical indenters tend to produce only elastic or visco-elastic deformations, sharper indenters cause further plastic or even fracture deformations. Since nanoindenters are often equipped with sharp tips (Berkovich or cube corner tip), the evaluation of viscoelastic properties must take into account the development of plastic strains that evolve mainly over the loading period. Ignoring the strains and related add-on deflections to Eq. 2, respectively, result in overestimation of creep compliance values as confirmed both experimentally and numerically (Vandamme et al. (2012)). To account for plastic strains developed in the loading period, Němeček et al. (2023) used a simple correction by adding an additional parameter, h_0, directly to Eq. 2 such that

$$(h(t) - h_0)^m = \kappa P_{max} J(t). \tag{4}$$

This formula leads to a successful fitting of the short-term experimental data. One of the advantages of this simple method is that it allows to estimate the amount of plastic deformation appearing in the loading phase depending on the type of an indenter.

Vandamme et al. (2012) analyzed the time-dependent response of a viscoelastoplastic material indented with a conical rigid tip. Their results indicate that the rate of change of the contact creep compliance is independent of any plastic deformation occurring during the loading phase. Based on this finding, a relationship between contact compliance and the testing parameters could be established (Vandamme and Ulm (2013)) as

$$J(t) = \frac{1}{E_{r,0}} + \frac{2a_c\Delta h(t)}{P_{max}} = \frac{1}{E_{r,0}} + \frac{ln(1 + t/\tau)}{C} \tag{5}$$

where $\Delta h(t)$ is the increment of indenter's penetration depth during holding phase, $a_c = \sqrt{\frac{A_c}{\pi}}$ is the radius of contact. Vandamme and Ulm (2013) further proposed a logarithmic function for fitting $\Delta h(t)$ from experiments leading to the second expression in Eq. 5, where C is the creep modulus and τ is the characteristic time.

3. Experiments and methods

Nanoindentation was performed on PMMA samples in the form of $40 \times 40 \times 4$ mm^3 blocks in ambient conditions (air, 21 ^{o}C, 20-40% RH). Based on previous studies (Němeček et al. (2024)), it was found that a very sharp cube corner tip overestimates creep compliance compared to less acute Berkovich or spherical tips. Thus, Berkovich indenter was used in this study. The loading protocol in the low load domain was prescribed as follows: linear loading for 5 s; 40 s holding at maximum force of 1 mN; linear unloading for 5 s using Hysitron TI-980 indenter. The loading protocol in the high load domain was prescribed with linear loading for 5 s; 60 s holding at maximum force (10 or 100 mN) and linear unloading for 5 s using CSM Nanohardness tester. To monitor reversible creep deformation (back-creep) an additional segment monitoring the deformation for the next 100 s after unloading to less than 5% of the maximum load was added. Then, the analytical approaches described in Section 2. were applied. This included non-linear least square regression of (i) the depth-time data of the holding period by Eqs. 3 and 4 using two Kelvin units and (ii) the depth increment-time data in the holding period using the logarithmic fit (Eq. 5). Then, creep functions $J(t)$ were constructed for each of the model.

4. Results and discussion

Experimental depth-time data acquired by the nanoindenter are illustrated in Fig. 1a where one can observe increased deformations over holding segment later used for fitting of viscoelastic constants and also reversible deformation after unloading in the 10 mN case. When fitting data from the holding period, both models provided very good agreements with the depth-time data, as illustrated in Figs. 1b and 1c. Parameters of the creep compliance functions for the different model variants are summarized in Tab. 1. Only small deviations are obtained between the parameters and $J(t)$ functions across the maximum load interval using the Kelvin chain model (Fig. 2a). This supports the idea of linearity of creep even for the high stresses generated by Berkovich tip. The amount of plastic deformation estimated by the model, h_0, scales with the load.

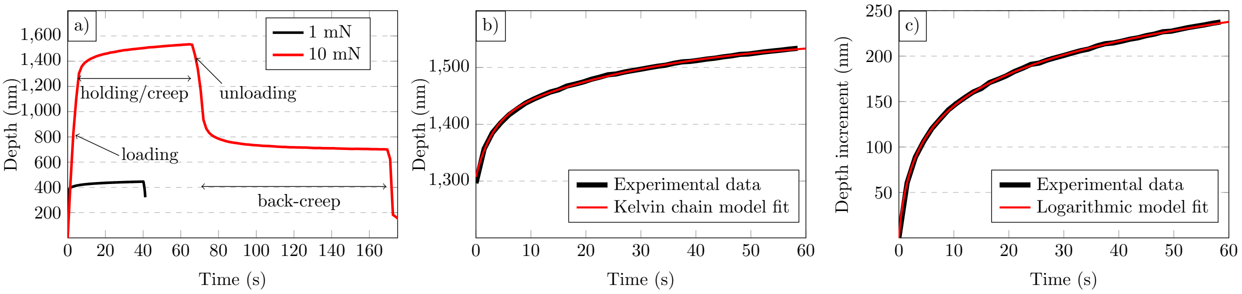

Fig. 1: (a) Example of nanoindentation depth-time curves, (b) fitting depth data with Kelvin chain model, (c) fitting of depth increment with the logarithmic model.

Tab. 1: Parameters of creep compliance functions.

Kelvin chain model						**Logarithmic model**			
						param set (1)		param set (2)	
P_{max} (mN)	E_1 (GPa)	τ_1 (s)	E_2 (GPa)	τ_2 (s)	h_0 (nm)	C (GPa)	τ (s)	C (GPa)	τ (s)
1	30.2	2.85	16.0	21.7	40.4	24.0	0.58	–	–
10	27.2	2.98	15.3	33.3	235.5	25.4	0.71	30.8	0.71
100	26.4	2.58	15.0	28.6	653	26.5	0.57	32.4	0.57

Different situation appears when using the logarithmic model. Significant variations were received. Results from the logarithmic model do not confirm perfect linearity for all loads. Functions $J(t)$ for cases of 10 mN and 100 mN lie below the 1 mN curve (Fig. 2b). The model requires the knowledge of the contact radius a_c, which can be derived from the contact depth h_c and related contact area A_c. For small loads (1 mN), the change in the contact radius over the holding period is not critical and does not affect the evaluation of viscoelastic constants. However, for higher loads and longer holding periods, the a_c varies significantly over the holding phase. This change is illustrated again in Fig. 2b which shows calculated $J(t)$ functions for 10 mN and 100 mN using the contact radius at the end (1) and at the beginning of the holding phase (2), respectively. A notable shift of $J(t)$ towards lower values was encountered. This sensitivity is impractical for routine measurements at different load levels. Also, significant deviations between $J(t)$ functions evaluated with the logarithmic model compared to Kelvin chain model were found. These deviations decrease as the load increases, as can be seen again in Fig. 2b.

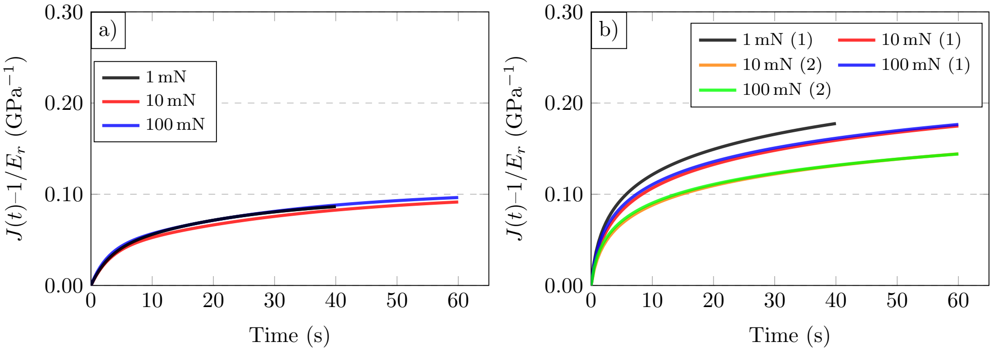

Fig. 2: Creep compliance functions for (a) Kelvin chain model, (b) logarithmic model.

The comparison of total deformations acquired during the holding and back-creep segments showed that the back-creep deformation was larger compared to the deformation in the holding segment for 10 mN and 100 mN cases (approx. +30%). This suggests that part of the viscous deformation developed also in the loading segment. Thus, none of the methods could provide accurate results due to finite ramping time. A higher loading rate could overcome this shortcoming, which is however difficult to achieve experimentally.

5. Conclusions

Nanoindentation using a Berkovich tip was employed to derive viscoelastic properties of PMMA, which serves as a reference material for other material components exhibiting analogous creep. Accurately estimating creep compliance functions from nanoindentation data poses a significant challenge due to various experimental and theoretical uncertainties. Both models studied account for the elimination of plastic deformations from the experimental data in a different way. The results from Němeček et al. (2023) appear to provide consistent outcomes over a large load span (1–100 mN), preserving the linearity of creep. Unfortunately, the Vandamme et al. (2012) model is sensitive to changes in the contact radius during the holding period and tends to yield results that depend on the applied load. Moreover, it appears that not all viscoelastic deformations could be identified from the holding period leading to the conclusion that none of the models provided error-free results. A more in-depth study is needed to determine a reliable methodology for small-scale creep assessment for PMMA as well as for other materials.

Acknowledgment

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References

Lee, E. and Radok, J. (1960) The contact problem for viscoelastic bodies. *Journal of Applied Mechanics*, 27, 3, pp. 438–444.

Němeček, J., Němečková, J., and Němeček, J. (2024) Micro-scale creep of C-S-H quantified by nanoindentation and different evaluation methods. In *Concreep12 conference, June 5–7, 2024, Delft University of Technology, Delft, The Netherlands*.

Němeček, J., Trávníček, P., Keppert, M., Halodová, P., Rosnecký, V., and Němeček, J. (2023) Nanomechanical analysis of gamma-irradiated cement paste exposed to different humidities. *Construction and Building Materials*, 393, pp. 131969.

Oyen, M. L. and Cook, R. F. (2009) A practical guide for analysis of nanoindentation data. *Journal of the Mechanical Behavior of Biomedical Materials*, 2, 4, pp. 396–407.

Vandamme, M., Tweedie, C., Constantinides, G., Ulm, F.-J., and Van Vliet, K. (2012) Quantifying plasticity-independent creep compliance and relaxation of viscoelastoplastic materials under contact loading. *Journal of Materials Research*, 27.

Vandamme, M. and Ulm, F.-J. (2013) Nanoindentation investigation of creep properties of calcium silicate hydrates. *Cement and Concrete Research*, 52, pp. 38–52.