European Journal of Mechanics / A Solids 68 (2018) 1–8

Contents lists available at ScienceDirect

European Journal of Mechanics / A Solids

journal homepage: www.elsevier.com/locate/ejmsol

Creep and recovery analysis of polymeric materials during indentation tests

Joseph Lejeune[a,b], Vincent Le Houérou[a], Thibaud Chatel[a], Hervé Pelletier[a,*], Christian Gauthier[a], Rolf Mülhaupt[b]

[a] Université de Strasbourg, Institut Charles Sadron, UPR 22 CNRS, 23 rue du Loess, BP 84047, F-67034 Strasbourg Cedex 2, France
[b] Freiburg Institute for Advanced Studies (FRIAS), Albert-Ludwigs-Universität Freiburg, Albertstraße 19, D-79104 Freiburg i.Br., Germany

ARTICLE INFO

Keywords:
Amorphous polymers
Indentation
Creep
Viscoelastic recovery

ABSTRACT

The present study describes comparative results obtained experimentally on two polymeric surfaces during contacts with a rigid spherical indenter. An experimental setup specifically dedicated to transparent materials has been used to analyze the creep phenomenon over long holding time segments (10^5 s) and to study the recovery process once contact has been removed. As a function of imposed contact time, a normalized value of representative strain is defined to quantify creep and recovery steps. Experimental results indicate that the recovery of the deformed surface after contact is mainly dependent on the initial contact time. Finite Elements Modeling will be used to discuss further the time-depending behaviors of the materials.

1. Introduction

Viscous properties of polymeric materials and their experimental determination have become an important area of research in the field of materials science. The surge of interest stems from the increasing use of polymers in many manufactured products covering a wide range of industrial applications. Indeed, polymers and related composites (polymeric matrix with fillers or additives (Mkaddem et al., 2013)) have been widely used in aerospace (Soussia et al., 2013), automotive (Bertrand-Lambotte et al., 2002; Schüssele et al., 2012), microelectronic (Biniek et al., 2013) food packaging (Silvestre et al., 2011), and biomedical applications (Pozza et al., 2012), due to their adequate strength, lightness, easy processing and low cost (Vielhauer et al., 2013). However, even if their elastic modulus values (1–10 GPa) are low in comparison with metallic or ceramic materials, polymers have interesting values of yield stress and strain (Dittmann et al., 2013). However, their mechanical properties are well known to be sensitive to temperature and strain rate. At room temperature, they usually exhibit viscoelastic and viscoplastic behaviors: during constant load tests (creep test) or constant imposed displacement (relaxation test), evolutions of strain and stress respectively occur (Struick, 1978; Martinez-Vega et al., 2002; Kolařík and Pegoretti, 2008).

Viscous properties are classically determined experimentally using tensile/compressive tests (Struick, 1978). Standard creep test methods consist in subjecting a bulk test specimen to a constant tensile or compressive load and measuring the corresponding deformation of the specimen as a function of time. Such tests are neither adapted to the manufacturing process of polymeric materials nor to the development of their future applications. Actually, the large number of relatively large-sized samples required is not adequate with service products (miniaturized devices, obtained by micromouldings). Such macroscopic standard creep tests do not take into account for the heterogeneity present in the material, especially in the near surface region, due to the strong interaction with the processing tools. Moreover, polymeric materials are more and more used as films or coating for decoration and protection purposes and are then subjected to mechanical contacts that generate surface damage. It is well known the surface and bulk properties of polymers may differ, as proven notably by molecular dynamics simulations (Frenkel and Smit, 1996).

Contact mechanical testing approaches have become quite common in the determination of viscoelastic and viscoplastic properties of surfaces (Fu and Fischer-Cripps, 2005; Gwynne et al., 2010). Compared to conventional creep experiments, indentation creep tests have several advantages. Using contact techniques (microindentation or nanoindentation), only small amounts of materials are needed for automated testing with a minimal sample preparation. Moreover, elastic or viscoelastic mechanical properties can be spatially mapped over the polymer surface, especially to highlight heterogeneities into the manufactured material (Scrinzi et al., 2011). Nevertheless, experimental determination of creep properties using depth-sensing indentation technique is not a simple task. A suitable method for measuring very long indentation creep (a day or more) in viscoelastic materials is still of great interest, especially due to non-linear viscoelastic behavior observed even at shallow indentation depth. Moreover, the indenter's tip

---

[*] Corresponding author.
E-mail address: herve.pelletier@insa-strasbourg.fr (H. Pelletier).

http://dx.doi.org/10.1016/j.euromechsol.2017.10.003
Received 7 March 2017; Received in revised form 31 August 2017; Accepted 14 October 2017

geometry has a rough effect on the experimental results as a function of the experimental conditions (maximum load, loading rate, holding time) (Fischer-Cripps, 2004; Menčík et al., 2011).

A number of investigations have been undertaken to measure the time-dependent mechanical responses of polymeric materials, using microindentation or nanoindentation techniques (Vandamme et al., 2012; Chen et al., 2012). Both for sharp (conical or pyramidal) (Menčík et al., 2011) or spherical (Oyen, 2005; Lee and Radok, 1960) indenters, studies have used load-displacement curves deduced from depth-sensing indentation. These previous studies propose models to extract mechanical properties for time-dependent materials (creep function, creep compliance, modulus values for instantaneous or long-time responses), using the evolution of the total penetration depth, recorded during step-loading conditions or ramp-hold creep experiments (Oyen, 2005). As initially done for elastic-plastic contact and time-independent response, authors tend to decompose the measured total penetration depth as a sum of elastic, viscoelastic and plastic displacements, and then propose constitutive responses (Oyen, 2007). Such proposed models reproduce with more or less accuracy load-displacement curves deduced from single or multiple ramp-hold creep tests for short holding time (less than 1000 s) (Olesiak et al., 2010).

The main problem of these approaches is related to the determination of the true contact depth, instead of the measured total penetration depth, especially at low normal applied load, in order to account for sinking or piling up effects, during loading/holding/unloading segments. That explains why we have developed a specific experimental set-up that allows in situ observation of the true contact area between the tip and the tested surface, during the holding segment (Chatel et al., 2011). In this previous study, we have presented original experimental results obtained on PMMA, showing the evolution of the true contact radius as a function of time, both during creep and recovery after withdrawal of the spherical tip. We have shown the influence of temperature and of the initial normal applied load on the average strain imposed during the contact (creep phase) and stored in the residual imprint (recovery phase). In the present study, we compare the time-dependent mechanical responses of two bulk polymers (PMMA and CR39) for long creep and recovery times (10^5 s). We demonstrate the influence of both the rheological properties of the tested materials and of the holding time during creep phase, especially on the mechanical response during recovery phase after complete withdrawal of the tip. The discussion benefits from additional numerical modeling used as reference of typical constitutive responses.

2. Materials and methods

2.1. Materials

In this study, the mechanical behavior during indentation creep tests of two amorphous polymeric materials is investigated at room temperature: a thermoplastic polymer (polymethyl methacrylate) PMMA, and a thermoset polymer called CR39 (poly allyl diglycol carbonate). These materials were respectively provided by Arkema and Essilor. They are transparent and used in the ophthalmic lens industry among others, where contact degradation is of great importance.

2.2. Experimental setup

Material visco-elasticity characterization was done by bulk relaxation in compression experiments. These relaxation tests were conducted under deformation in the range 0.75% and 20%.

Creep and recovery phenomena, studied using indentation tests, both during holding time at constant load and after rapid withdrawal of the spherical tip, were directly monitored using a homemade experimental device (Gauthier and Schirrer, 2000). This original experimental system has been mainly used to study scratch responses of polymeric materials (Chatel et al., 2011, 2013; Gauthier and Schirrer, 2000). Since then, a first analysis of creep phenomena on PMMA surface has been proposed (Chatel et al., 2011). This home-made scratch and indentation system is an efficient tool to understand tribological phenomena of polymeric surfaces, such as tested materials in the present study. Indeed, the contact size of about 100 μm can be observed in situ during scratch and indentation tests (Chatel et al., 2011). As a consequence the effective penetration depth and all geometric data can be directly determined with accuracy, without any help of a specific analytical model. However, in comparison with standard indentation test systems, the load-displacement curves cannot be assessed.

Microindentation tests, using a glass spherical indenter with a tip radius R of 400 μm, were carried out at room temperature. The applied normal load, F_N, was chosen to perform creep tests with initial imposed average strain close to 0.2, according to Tabor's ratio a_0/R (Tabor, 1970), with a_0, the initial value of the contact radius, just after the loading process with a constant loading rate. The applied normal load was kept at the constant value for creep duration of 10^5 s. During experiments, a dead weight is applied thanks to the experimental indentation system, so no drift phenomena occur. During holding time, the normal load is recorded as a function of creep time, and ten pictures showing the contact between the spherical tip and the deformed surface are recorded for each tested time decade, allowing the determination of contact radius $a_c(t)$ and its evolution over the holding time during creep (Fig. 1). Due to contact issues (during the early stage of the dead weight loading, the applied normal force may exhibit a slight overshoot and the camera focus often needs to be readjusted), the first contact observation is only reliable after 0.5 s.

After the creep phase of 10^5 s, the spherical indenter is rapidly removed from the deformed surface and a glass plate is placed over the residual imprint. As described in Ref. (Chatel et al., 2011), the glass slide generates some interference fringes that provide an accurate determination of the curvature radius of the residual imprint, $R(t)$. As performed during creep phase, the residual contact radius $a_R(t)$ can be estimated, and coupling $R(t)$ and $a_R(t)$, we are able to determine the residual depth $h_R(t)$, as a function of the recovery time (Fig. 2).

2.3. Data analysis

During creep phase, using in situ observation, the average contact pressure imposed during microindentation tests at constant normal applied load can be estimated as a function of creep time:

$$p_m(t) = \frac{F_N(t)}{\pi a_c^2(t)} \tag{1}$$

as defined initially by Tabor (1970) and confirmed by finite element modeling (Pelletier et al., 2009), the average strain, imposed during indentation with a spherical indenter can be expressed as follows:

$$\varepsilon = k\frac{a}{R} \tag{2}$$

with a, the true contact radius, R, the tip radius and k, a constant close to 0.2 for amorphous polymeric materials (Pelletier et al., 2009). To follow the evolution of average strain imposed during creep phase, due to the variation of the contact radius, $a_c(t)$, the following expression, describing the normalized strain as a function of time, has been introduced (Chatel et al., 2011):

$$\frac{a_c(t)/R}{a_c(t_0)/R} = \frac{a_c(t)}{a_c(t_0)} \tag{3}$$

with $t_0 = 0.5\,s$, which corresponds to the first available contact observation, after the loading process. Similarly, to quantify mechanically the viscoelastic recovery phenomena after indentation, a normalized strain during recovery can be defined as follows:

$$\frac{a_R(t)/R(t)}{a_R(t_0)/R} \tag{4}$$