The influence of strain hardening of polymers on the piling-up phenomenon in scratch tests: Experiments and numerical modelling

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Abstract

The aim of this study was to relate the scratching behaviour of polymers to their mechanical properties. A thermosetting resin (CR39) and a thermoplastic polymer (PMMA) were studied using a microscratch tester allowing in situ observation of the contact area. These two polymers exhibit different elastic and viscoplastic properties, the main difference being the large ability of CR39 to strain harden, whereas PMMA softens. A spherical indenter was used to vary the level of deformation imposed on the samples. The response was initially elastic, then viscoelastic and finally mainly viscoplastic with increasing penetration of the indenter into the material. The two polymers displayed the same response for small levels of deformation, while at larger strains PMMA showed more pronounced plastic behaviour. The origin of this difference in behaviour was investigated by means of a three dimensional finite element analysis. The rheology of PMMA and CR39 was simplified and modelled by assuming linear elastic behaviour and a viscoplastic law taking into account their strain hardening capacity at high strains. Strain hardening was found to be a key factor to correctly model the material flow around the indenter. The response of the polymers was governed by the ratio between the plastic and elastic strains involved in the deformation in the contact region. In first approximation, the representative strain was imposed mainly by the geometry of the indenter, while the elastic deformation was controlled by the mechanical properties of the material, a larger strain hardening leading to a greater elastic deformation and a lower plastic strain thus a better scratch resistance of the specimen.

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1. Introduction

Transparent polymers are widely used in many industrial applications such as ophthalmology and the automobile industry and are often preferred to glass because of their lower density and lower brittleness. However, their relatively low hardness makes their surface susceptible to damage. Thus, scratches alter their optical, aesthetic and mechanical properties and hard varnishes are deposited on the surface to avoid the occurrence of such alterations. The industrial and academic communities are both interested in developing new materials having a better scratch resistance, which passes through a better understanding of the mechanics of the scratch test (Fig. 1).

According to Briscoe et al. [1], the deformation modes of polymers during a scratch test may be classified in three main categories: quasi-elastic behaviour, ductile ploughing and severe damage. In the first regime, there is no significant residual groove due to the large viscoelastic recovery at the rear of the indenter. In the second mode, the material flows plastically around the indenter and this ductile ploughing is...
accompanied by piling-up at the sides of the groove. The last regime is reached for severe conditions of deformation where cracks develop at the edge or within the groove [2]. In some cases, machining or chip formation may also be observed. The behaviour of the material and the appearance of one of these three deformation modes depend on several factors including the friction [3], temperature [4], indenter speed [5] and level of deformation imposed on the specimen [6,7]. The latter parameter is controlled by the shape of the indenter and scratching mode maps relating the strain the temperature, normal load ... to the kind of deformation and damage modes have been established [1]. In the case of scratching with spheres, Gauthier et al. [8] have shown that the level of deformation \( \varepsilon \) is governed by the relation proposed by Tabor for indentation of metals [9]:

\[
\varepsilon = \frac{0.2 a}{R} \tag{1}
\]

where \( R \) is the radius of the scratching tip and \( a \), the radius of the contact area (Fig. 1). The deformations under the indenter tip are not homogeneous and this relation gives an average value of the deformation. As this equation was established for scratch tests on materials having a very small elastic strain at yielding:

\[
\varepsilon_{ey} = \frac{\sigma_y}{E} \tag{2}
\]

where \( E \) and \( \sigma_y \) are, respectively, Young’s modulus and the yield stress, the coefficient 0.2 does not hold for polymers which have large elastic strains [10]. Nevertheless, for spherical indenters, the deformation remains proportional to the ratio \( a/R \), which is termed the representative strain in the following discussion.

Over the last few years, efforts have been made to gain a fundamental understanding of the scratch test on polymers in order to relate the deformation modes to the mechanical properties of the materials [8,11–13]. Using a finite element analysis, Bucaille et al. [12] were able to link the contact pressure, contact geometry and the elastic recovery at the rear of the indenter to the elastoplastic properties of the material. Despite the fact that the rheological model employed in this study is a very rough description of the real rheology of polymers, as it introduced only \( E \) and \( \sigma_y \), it gives valuable information concerning the material flow around the indenter. Bucaille et al. [14] recently introduced a more complex rheological model and showed that the strain hardening of polymers and the ratio between the elastic and plastic strains are key points to explain the scratch resistance. In this context, the present work deals with the transition between the quasi-elastic and ductile ploughing regimes. Microscratching tests were carried out on two polymers at small and large strains. Pile formation and the extent of elastic recovery were observed in situ and related to the material behaviour measured under compression. These tests were then modelled using three dimensional finite element modelling. The aim of the numerical simulation was to show, by using a simplified modelling of the polymer rheology, that there is a strong link between the strain hardening of polymer and the material flow around the indenter during scratching.

![Fig. 1. Principle of the scratch test and in situ observation device. The main geometrical parameters are defined on the schema.](image-url)
2. Experimental procedures

2.1. Experimental set-ups

Two technical approaches previously developed were employed to investigate the bulk and surface behaviour of polymeric materials: a compressive test and a scratch test. The mechanism of the compressive test is based on the moving cross head of an Instron 4502 tensile machine and the whole apparatus is enclosed in an Instron environmental chamber. Compressive tests over a wide range of strain rates ($10^{-4}$ to $10^{-1}$ s$^{-1}$), within a temperature range covering the α and β relaxation peaks of common polymers (−70 to +120°C) and measuring the longitudinal and radial strain, are the main characteristics of this system. The longitudinal strain is limited to 20% during tests.

The experimental device for the scratch test, called the 'microvisioscratch', comprises a commercial servomechanism bearing a small, temperature controlled transparent box which contains only the sample and the scratching tip [5]. Control of the moving tip and recording of the normal and tangential load, $W$ and $F_t$, respectively, scratching speed $V_{\text{tip}}$ and temperature $T$ are computer driven. A built-in microscope allows in situ observation and measurement of the groove left on the surface. Scratching over a wide range of speeds (1–10$^4$/m/s) and within the same temperature range (−70 to +120°C) are the main innovative features of the system. The normal load $W$ applied to the moving tip can be selected from 0.05 to 5 N. This normal load was adjusted by the crushing of a spring having a low stiffness. In the present experiments, performed at room temperature, the speed of the tip was kept constant at 0.1 mm/s while the normal load was varied step-wise within a single groove in as many steps as required to explore the entire range of strain sensitivity. The moving tip was a cone-shaped diamond having an apex angle of 60° and a tip radius $R=116$ μm. According to the relation given by Briscoe et al. [1], the mean strain rate:

$$\dot{\varepsilon} = \frac{de}{dt} \approx \frac{V_{\text{tip}}}{2a}$$

lies in the range 0.5–2 s$^{-1}$ for these tests. Fig. 1 shows a schematic diagram of the scratch apparatus and Fig. 2 an in situ photograph taken during the scratching process for three typical types of behaviour: elastic sliding (upper), viscoelastic sliding (centre) and viscoplastic scratching (lower). The main geometrical parameters of the contact and the groove are defined on these figures.

2.2. Materials

Two materials were used in this study: a commercial grade of cast polymethylmethacrylate (PMMA) and an amorphous polymer called CR39 (diethylene glycol bis(allylic carbonate)). CR39 is a thermoset resin cast for 20 h according to a specific temperature cycle (20–100°C). Young’s modulus $E$ of CR39 is typically 2 GPa and that of PMMA typically 3 GPa at 20°C and 1 Hz. Cylindrical samples 12 mm long and 5 mm in diameter were used for the compression test while the scratch test sample was a plate a few millimetres thick.

3. Experimental results

3.1. Compression

In the elastic domain of CR39 or PMMA under compression at small strains, the increase of the stress with the strain is almost linear and can be approximated by the Young’s modulus, as seen in Fig. 3a. The unloading exhibits non-linear elastic behaviour and the elastic part of the whole deformation is much greater than that deduced from a simple linear elastic unloading. This is especially striking in the case of CR39, for which after loading up to a total strain of 0.18, the residual plastic strain after unloading is 0.10 whereas the elastic strain of this material at yielding $\varepsilon_y$ during loading may be estimated to be 0.03. Two explanations for this behaviour should be noted: (i) CR39 is a thermoset resin having a large anelastic unloading and (ii) its behaviour depends strongly on the strain hardening of the compressive strain/stress response. PMMA does not display this type of behaviour although the elastic domain during loading (defined by the intersection of the linear elastic curve and the yield stress level) is of the same order of magnitude as for CR39. In Fig. 3b, the compressive true stress $\sigma$ normalised...
Fig. 3. Compressive true stress $\sigma_{\text{comp}}$ vs. true strain (a) and compressive true stress normalised to the yield stress $\sigma_{\text{comp}}/\sigma_y$ vs. true strain (b) for CR39 and PMMA. $\varepsilon_0/dt=10^{-4}$ s$^{-1}$, $T=21^\circ$C.

to the yield stress $\sigma_{\text{comp}}/\sigma_y$ is plotted against the true strain. The strain hardening of CR39 during loading is easily compared to the softening of PMMA at low strain. Of course, at high strain these two materials display strain hardening as described in the numerical simulations.

3.2. Sliding and scratching

3.2.1. In situ observations

One should first recall here the difference between scratching and sliding behaviour. The nature of the strain inside the half spherical volume under the contact area determines the nature of the process involved and in all cases, the bulk material displays elastic behaviour (Fig. 2).

- In the case of elastic sliding, as showed on polymer at very low contact strain, there is no groove left on the surface and the contact is a full disk: the rear angle $\omega$ is $\pi/2$ and the rear length is comparable to the frontal length.

- During viscoelastic sliding, the groove initially left on the surface relaxes within a time comparable to the contact time and the rear angle $\omega$ is significantly less than $\pi/2$ (Fig. 2b). At the transition between viscoelastic sliding and viscoplastic scratching, a viscoelastic-plastic contact exists for which the material in the contact area is subject to both viscoelastic and plastic strains. In this case, there is an elastic unloading and partial recovery of the groove after passage of the tip.

- During viscoplastic scratching, the material under the contact surface is mainly subject to plastic strain. The elastic recovery depends on the ratio between the yield stress and the Young’s modulus and is weak but visible under the in situ microscope: the rear angle $\omega$ (Fig. 2c) is typically $0.3$ rad for polymers.

The same experimental procedure was adopted for both materials. After starting the passage of the tip, a normal load was applied to it and increased stepwise. At each loading step and throughout the scratching process, in situ photographs were taken to save information on the shape of the true contact area. Fig. 4a–c show representative photographs for the two specimens. At a normal load of a few tenths of a Newton, the tip slides over the surface of the polymeric material and no residual groove is observed on either PMMA or CR39. The
ratio between the contact radius and the radius of the tip \((aR)\) is typically 0.24. A viscoelastic contact is observed as the ratio \(aR\) tends to 0.3. The beginning of the viscoelastic-plastic regime is then observed as \(aR\) tends to 0.34: the groove left on the surface has a horizontal imprint (a) and two lateral sloping faces (b) and does not present lateral plastic pads. The edges of the groove do not remain parallel and as the lifetime of the groove increases the slope of the lateral faces decreases (b) and with it the depth of the groove. As the viscoelastic-plastic contact strain increases, the lateral plastic pads generated by the elastic unloading increase (c) and a frontal push pad appears (d). As the ratio \(aR\) becomes greater than 0.42, PMMA flows viscoplastically around the indenter and the frontal and lateral pads merge to form a continuous cord (e). A higher contact strain does not further modify the shape of the contact area. Conversely, on CR39 (Fig. 4b) the frontal push pad and lateral pads of the groove left on the surface are never continuous. At a normal load of about 3 N, the residual groove on PMMA does not recover significantly, whereas on CR39 there is a significant recovery (Fig. 4c). At a given ratio \(aR\), the normal load on the tip during contact is always greater on PMMA than on CR39; it means that at the strain rates of the scratch test, the mean stress in the range of the strains involved are always higher for PMMA than for CR39 as in compression tests performed at much lower strain rates (Fig. 3a).

PMMA and CR39 are brittle materials at room temperature under shear and tensile stresses. However, in the case of the experimental parameters during scratching, the grooves left on the surface do not show crazing or cracking and are ductile. Other experiments show that with lower radius tip or under higher normal load crazing in PMMA and cracking in CR39 may appear [15].

### 3.2.2. Geometry of the contact and apparent friction coefficient

The geometry of the true contact area, which is a full disc at low strains, is modified as the contact radius increases. In addition, plastic flow around the tip can generate a plastic pad pushed in front of it if the yield criterion is attained in the contact area. Since the material is viscoelastic and viscoplastic and sensitive to temperature, at constant normal load, an increase in tip speed or a decrease in temperature decreases the contact radius and also the mean strain. So, the same results may be obtained at different temperatures and sliding speeds by adjusting the normal load to vary the strain and strain rate.

The recovery of the groove and the symmetry of the contact area can be estimated by means of the rear angle \(\omega\) varying from 0 to \(\pi/2\) (Fig. 2). Fig. 5 depicts the evolution of the rear angle with the “mean strain” \(aR\) for PMMA and CR39. The contact is symmetrical (\(\omega = \pi/2\)) up until a representative strain \(aR\) of about 0.2 for both materials. This indicates that the range of the elastic contact does not depend only on the absolute value of Young’s modulus, but rather on the ratio between the yield stress and Young’s modulus \(\epsilon_y\), which represents the elastic strain at yielding; this ratio is equal to 0.03 for both polymers, as seen in Fig. 3b. When \(aR\) lies in the range 0.2–0.42, the rear angle decreases in the same manner for PMMA and CR39. At \(aR = 0.3\), plastic deformation occurs under the tip and thereafter a permanent groove remains on the surface and plastic deformations in the strained volume increase as \(aR\) increases. At higher normal loads, the shape of the contact area depends on the material. The rear contact angle \(\omega\) continues to decrease for PMMA as previously described [8]. In the case of CR39, at a critical ratio \(aR\) of about 0.45, the rear angle passes through a minimum of 0.5 rad and then increases slowly for higher values of \(aR\). Although the contact is mainly viscoplastic at this critical strain, the increase in \(\omega\) suggests that the elastic strain during the unloading increases. The evolution of the size and shape of the push pad with the strain presents similarities to the evolution of the rear angle. While the contact remains viscoelastic or viscoelastic–plastic, there is no push pad, but as the contact becomes viscoplastic \((aR > 0.4)\), a frontal push pad appears. The normalised push pad is defined as the ratio between the thickness of the pad and the radius of the contact and is shown in Fig. 6 as a function of the strain. On PMMA, the normalised push pad increases more quickly than on CR39. Fig. 7 shows the evolution of the apparent friction coefficient:

\[
\mu_{app} = \frac{F}{W} \tag{4}
\]

as a function of the ratio \(aR\) for the two polymeric materials. This apparent friction coefficient is the ratio between the tangential force and the normal load applied to a moving scratching tip. It includes a so-called “true local” friction coefficient which is the friction shear stress to normal stress ratio at the interface between the tip and the surface being scratched and a “geometrical” friction coefficient, which is the plough effect due to the wave front created ahead of the moving tip and depends on the shape of the tip [16]. This coefficient can be divided to a first order into a ploughing
component and an adhesive component. At a very low ratio $a/R$, where the contact is elastic and there is no ploughing part, it is possible to estimate the adhesive part. A value in the range 0.08–0.12 was determined for viscoelastic sliding on PMMA or CR39. While $a/R$ lies between 0.2 and 0.4, the rear angle decreases and the ploughing part of the apparent friction coefficient increases in the same manner for both materials. As $a/R$ exceeds 0.4, like the normalised push pad (Fig. 6), the apparent friction coefficient increases more rapidly on PMMA than on CR39. This is due to the difference in ploughing component which increases more quickly on PMMA than on CR39 as we have previously seen.

The geometrical sphere–cone transition of the indenter used in experiment is for $a/R = 0.87$. This transition was not reached during scratching experiments. However, above this transition, the deformation continues to increase although the material surface is in contact with the conical part of the indenter. This effect has already been used both in indentation [17] and in scratch [18] to explore deformation mechanisms of polymers. This shows that scratching does not only involve deformation of the surface but also a bulk response of the material.

4. Numerical modelling

4.1. Finite element model

Scratching of polymers was modelled using the Forge3® implicit code which has an automatic remeshing procedure. The finite element (FE) mesh is a right-angled parallelepiped and Fig. 8 shows the half of the mesh corresponding to the region $z > 0$, the plane $x = 0$ being a symmetry plane. The remaining degrees of freedom of the FE domain were fixed in the other directions by the planes $y = 0$ and $z = 0$ in which nodes were allowed to move. The size of the domain was chosen so that boundary effects did not influence the results. Its dimensions conformed to the rules given by Bucaille [19], i.e., a width of 10 times the contact radius, a height of six times the penetration depth and a length of at least seven times the contact radius, so as to reach a steady regime (constant forces and groove geometry independent of the scratch length). The rigid indenter was modelled as a sphere of 100 $\mu$m radius, which was moved along the y-axis at constant penetration depths $h = 2, 4, 10$ and 20 $\mu$m for PMMA and $h = 2, 5, 10$ and 20 $\mu$m for CR39. The scratching speed was kept constant at $V_{tip} = 1 \mu$m/s. The elements of the domain were three-dimensional meshes with four-node tetrahedra. Far from the indenter, these elements had a typical length of about 30 $\mu$m. Near the indenter, the mesh was refined so that at least 20 nodes were in contact with the sphere at the edge in the plane $x = 0$. At a penetration depth of 2 $\mu$m, the elements were 3 $\mu$m long. As an example, simulation of a scratch test on PMMA required 11,000 nodes and 45,000 elements, about 12 h of
CPU time and a remeshing procedure every 20 increments. At each time increment, the forces and geometrical parameters defined in Figs. 1 and 2 were computed by a post processing procedure giving the average values in the steady state regime.

4.2. Rheological model

PMMA and CR39 were considered to be homogeneous. The aim of the numerical modelling was to show that there is a strong influence of the elastic and the viscoplastic parts on the scratching behaviour. We did not focus on a complex modelling of polymer rheology but rather on a simplified rheological law which explains the phenomenon observed experimentally. The three parameters \( K, h_g \) and \( h_p \) were determined by inverse analysis of indentation tests [17,19].

The friction coefficient \( \mu \) was obtained from measurements of the apparent friction coefficient during scratching [10]. \( \nu \) was measured under compression [19]. \( E \) was obtained in indentation tests [20] and \( m, K \) and \( h_p \) were deduced by inverse analysis of indentation tests [17,19].

The friction at the interface between the indenter and the material in scratch tests was modelled with a Coulomb’s friction coefficient \( \mu \). Its value was determined from measurements of the apparent friction coefficient \( \mu_a \) for sliding of a sphere on PMMA or CR39 [19] (Table 1). As shown by Lafaye et al. [22], the adhesive part of the apparent friction coefficient depends on the level of strain imposed on the polymer at the contact surface and increases as the material under the contact area deforms plastically. Consequently, it is difficult to give an accurate value of the local friction coefficient \( \mu \); our value is an approximate one and was assumed to be independent of the strain and strain rate.

Although, elasticity of polymers is most often non linear viscoelasticity and so depends on strain and strain rate, the elastic behaviour was modelled by a linear incremental law, defined by Young’s modulus \( E \) and Poisson’s ratio \( \nu \), both taken as constant. \( E \) was determined in indentation tests from the slope at the beginning of the unloading curve using the method described by Hochstetter et al. [20] (Table 1). The Poisson’s ratios \( \nu \) of PMMA and CR39 were obtained in compression tests [19]. The yield condition is given by von Mises’ yield criterion while the flow stress may generally be described by a G’sell–Jonas law [21]:

\[
\sigma = K_\mu \frac{\varepsilon_{vp}}{\varepsilon_{vpe}} (1 - e^{-\varepsilon_{vp}/\varepsilon_{vpe}})\varepsilon_{m}^{\varepsilon_{vpe}}
\]

where \( \varepsilon_{vp} \) and \( \varepsilon_{vpe} \) are, respectively, the generalised viscoplastic strain rate and strain; \( K_\mu \) the consistency; \( \varepsilon_m \) a thermal coefficient, \( h_g \) the strain hardening coefficient and \( m \), the sensitivity to the strain rate. In the formalism of G’sell and Jonas, the term \( (1 - e^{-\varepsilon_{vp}/\varepsilon_{vpe}}) \) describes the viscoelastic behaviour under loading, but does not model the elastic unloading part of the deformation, and this term was not considered in the present work. In our simulation, the elastic recovery is directly related to the ratio of the flow stress \( \sigma \) to the Young’s modulus \( E \). Thermal effects are neglected and \( \varepsilon_{vpe} \) is equal to 0. Eq. (6) then becomes:

\[
\sigma = K_\mu \varepsilon_{vp}^{\varepsilon_{vpe}} \text{ which implies } \varepsilon_{m} = K_\mu \varepsilon_{vp}^{\varepsilon_{vpe}}
\]

The three parameters \( K, h_g \) and \( m \) were determined by an inverse method adapted to large deformations and based on the interpretation of the force–penetration curves in indentation tests with two indenter shapes [17] (Table 1 and Fig. 9). In this description, the strain hardening effort was simply \( e^{\varepsilon_{vpe}} \) and does not depend on the value of the strain rate.

The friction at the interface between the indenter and the material in scratch tests was modelled with a Coulomb’s friction coefficient \( \mu \). Its value was determined from measurements of the apparent friction coefficient \( \mu_a \) for sliding of a sphere on PMMA or CR39 [19] (Table 1). As shown by Lafaye et al. [22], the adhesive part of the apparent friction coefficient depends on the level of strain imposed on the polymer at the contact surface and increases as the material under the contact area deforms plastically. Consequently, it is difficult to give an accurate value of the local friction coefficient \( \mu \); our value is an approximate one and was assumed to be independent of the strain and strain rate.

4.3. Finite element results and first comparisons with experiments

Fig. 10 shows that for small penetration depths, the material sinks down in front of the indenter and as the strain imposed on the material increases, pads are formed at the front and sides of the groove. A push pad appears at larger strains for CR39 than for PMMA, while for a given representative strain \( \varepsilon_R \), the thickness of the push pad \( \varepsilon_p \) is less for CR39. The frontal pad formed on PMMA is sharper than the pad formed on CR39. Geometrical analysis provides easily the relation between the contact radius \( a \), the radius of the tip

<table>
<thead>
<tr>
<th>Material</th>
<th>PMMA</th>
<th>CR39</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \mu )</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>( \nu )</td>
<td>0.1</td>
<td>0.078</td>
</tr>
<tr>
<td>( E (\text{MPa}) )</td>
<td>204</td>
<td>145</td>
</tr>
<tr>
<td>( h_g )</td>
<td>0.5</td>
<td>4.5</td>
</tr>
<tr>
<td>( d ) (GPa)</td>
<td>4.2</td>
<td>2.1</td>
</tr>
<tr>
<td>( \nu )</td>
<td>0.35</td>
<td>0.4</td>
</tr>
</tbody>
</table>

The friction coefficient \( \mu \) was obtained from measurements of the apparent friction coefficient during scratching [10]. \( \nu \) was measured under compression [19]. \( E \) was obtained in indentation tests [20] and \( m, K \) and \( h_p \) were deduced by inverse analysis of indentation tests [17,19].

Fig. 9. Rheological behaviour of PMMA and CR39 used to model 3D scratching, \( d \varepsilon/dt = 10^{-3} \text{ s}^{-1} \). The values of the rheological parameters were determined by inverse analysis of indentation tests (Table 1).

![Fig. 9. Rheological behaviour of PMMA and CR39 used to model 3D scratching, \( d \varepsilon/dt = 10^{-3} \text{ s}^{-1} \). The values of the rheological parameters were determined by inverse analysis of indentation tests (Table 1).](image-url)
Fig. 10. Scratch profile obtained by simulation in the plane \( x = 0 \) as a function of the penetration depth of the indenter for (a) PMMA and (b) CR39.

This relation provides the value of the contact height \( h_c \) for a contact with a radius \( a \). It is drawn graphically on Fig. 11, on which we have also reported the calculated values of the reduced contact radius versus the values of the reduced penetration depth for the two materials. As can be seen in Fig. 11, the contact between the frontal part of the indenter and the CR39 surface always lies a little below the nominal surface \((h_c < h)\), even at the highest penetrations where a pad is observed. On the contrary, it lies above the nominal surface \((h_c > h)\) for the PMMA for \( a/R > 0.4 \). On CR39 the elastic deflection at the front of the contact is greater than on PMMA and we can notice that the difference in contact geometry becomes marked for \( a/R > 0.4 \) about as observed previously in experiments (Fig. 6).

With regard to the rear contact angle, we observe some discrepancies with experiments (Fig. 5): even for the smallest representative strains \((a/R \sim 0.17)\), the rear contact angle computed by simulation is not larger than 0.8 rad for CR39 or 0.5 rad for PMMA (Fig. 12). It decreases steadily as \( a/R \) increases, but it is always greater for PMMA than for CR39. It means that the elastic recovery is greater for CR39 whatever the representative strain, as it can be seen on Fig. 10, and displays a constant relative diminution as \( a/R \) increases. We can notice, however, that for \( a/R \sim 0.4 \), the calculations provide the good order of magnitude of the rear contact angle \((0.2-0.4 \text{ rad})\) if we take into account the experimental error on this quantity which is difficult to measure accurately. The calculated values of the apparent friction coefficient (Fig. 13) are higher than the experimental ones (Fig. 7), but their evolution with \( a/R \) for the two materials present a very great similarity with experimental evolution: \( \mu_0 \) increases in the same manner for the two polymers until a representative strain of 0.4, after which it increases more slowly for CR39 (Fig. 13). As in experiments, the higher values for PMMA are mainly due to the smaller rear contact angle which induces an increase in the ploughing component of the apparent friction coefficient.

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**Fig. 11.** Evolution of \((a/R)^2\) vs. \(h/R\) for PMMA and CR39 according to numerical results.

**Fig. 12.** Evolution of the rear contact angle vs. \(a/R\) for PMMA and CR39 according to numerical results.
of elongation\[21,23,24\]. Bucaille et al. \[14,17\] considered related to the extension of macromolecules in the direction of resin, the macromolecules are cross-linked and form a stiff network, which leads to a strong increase in the flow stress and a large value of the strain hardening exponent \(h_p\) (Table 1). Consequently, the stresses in CR39 are initially lower than those in PMMA, as observed under compression, but become much higher for larger strains, for instance about 75\% higher at \(\varepsilon=0.4\).

A similar remark can be made concerning the values of the friction coefficient: these scratch tests suggest, if we consider the values for \(aR=0.2\) (Fig. 7), a value \(\mu_\text{PMMA}.\) has previously observed that such difference in friction coefficient do not induce significant difference in contact geometry, our main conclusions remain valid.

In scratching experiments, a full “apparent” elastic contact \((\varepsilon_\text{app}=\pi/2)\) was observed up to \(aR=0.2\), whereas in simulation the rear contact angle did not exceed \(\pi/4\) (Figs. 5 and 12).

Several phenomena are responsible for this lack of elastic recovery at the rear of the indenter in simulation:

- Firstly, assuming a frictionless contact during indentation, it is well known that the end of the elastic regime is attained as the contact radius satisfies the relation \[\frac{a}{R} = 2.6\left(1 - \varepsilon_\text{app}^2\right)\] (9)

In the sliding regime of scratching experiments, the end of the apparent elastic contact (full circular disc) corresponds to \(a/R=0.2\) (Fig. 5). Hence, for CR39, for example, with \(E=2.1\) GPa and \(v=0.4\), this gives \(\varepsilon_\text{app}=192\) MPa, which is higher than the value derived from Eq. (7) and Table 1 (\(\varepsilon_\text{app}=145\) MPa for \(E=\pi/4\)). Furthermore, in simulation the indenter speed is two decades lower than in scratching experiments. According to Eq. (7), the end of the apparent elastic contact is reached for \(a/R=0.07\) (PMMA) or 0.1 (CR39) and these values of \(a/R\) are effectively smaller than those chosen for simulation (Fig. 12).

So, the numerical results are in agreement with the relation (9).

- Secondly, the viscoelastic domains of CR39 and PMMA under compression at small strains were modelled by means of Young’s modulus (Fig. 3). However, the unloading exhibits non linear viscoelastic behaviour and the effective elasticity contribution to the total deformation is much greater than that predicted by the simple equation \(s/E\). Gauthier et al. \[8\] have also shown that the elastic recovery of the groove depends on the contact strain. These viscoelastic phenomena were not taken into account in simulations and strains beyond the yield were assumed to be viscoplastic. The present rheological model represents the material behaviour fairly well in front of the indenter, so long as the material remains under compression. On the contrary, at the rear contact area, the stresses are relaxed and the elastic recovery is, therefore, underestimated.
Finally, the numerical simulations do not take into account the different strain hardening under compression as opposed to tension, the latter being much higher [23,24]. This modifies the predicted flow of the material around the indenter. Since the material behind the indenter is mainly under tension and it has been shown that a larger strain hardening amplifies the elastic effects, it is a second reason for which the simulations thus underestimate the elastic recovery.

These points demonstrate that it is difficult to directly correlate the behaviour of the polymers under compression with their behaviour during indentation or scratching. Certain interesting features are nevertheless observed, for which numerical modelling provides some explanations. One of these concerns the influence of strain hardening.

Simulations of a scratch test on CR39 without strain hardening ($h_g = 0$) were carried out for two penetration depths, 2 and 20 μm. At $h = 2$ μm, no changes in the apparent friction coefficient or scratch profile were observed, whereas for $h = 20$ μm, the material flow was completely modified and the push pad degenerated into a chip (Fig. 14). No such damage was detected under the same conditions in scratching experiments. This shows that the formation of a push pad and the material flow at the front of the contact depend strongly on the rheology of the polymer, in this case CR39. In the sliding contact regime, the elastic strain at yield $\varepsilon_{ey}$ controls totally the material behaviour under the indenter, while under scratching conditions strain hardening is an additional fundamental parameter.

The material under the tip is under high hydrostatic pressure and simulations indicate that tensile stresses act on the rear edge of the contact area and on the bottom of the groove. Fig. 14 shows that as the strain hardening coefficient increases, the elastic recovery at the bottom and sides of the groove becomes larger. Since CR39 exhibits significant strain hardening under compression, one may assume that its strain hardening is still higher under tension, as observed for ductile polymers [23]. This effect is not expected to be so great for PMMA, as its strain hardening coefficient is quite weak under compression. This explains why the evolution of the rear contact angle in experiments is different in the viscoplastic regime for PMMA and CR39 (Fig. 4).

### 5.2. Elastic and plastic deformations

Different deformation regimes were observed on PMMA and CR39 with increasing penetration of the spherical indenter into the material. At representative strains below 0.4, the two polymers displayed similar behaviour in scratching experiments, although their elastic and plastic properties are different. On the other hand, numerical results have shown that for two materials having different Young’s moduli but similar plastic properties at small strains ($\sigma R < 0.2$), the elastic recovery and scratching behaviour are different. It would, in fact, seem clear that the behaviour of a material in a scratch test depends on both its elastic and its plastic properties. During scratching, the elastic recovery is directly related to the ratio of the flow stress $\sigma$ to the Young’s modulus $E$ of the specimen [12] and this elastic strain represents the elastic deformation in the contact area (Fig. 9):

$$\varepsilon_{ey} = \frac{\sigma}{E}$$  \hspace{1cm} (10)

As the yield stress and Young’s modulus of PMMA are both about twice those of CR39, $\varepsilon_{ey}$ is the same for the two polymers and their behaviour during scratching is similar in the viscoelastic-plastic regime. Owing to the different values of $h_g$ of PMMA and CR39, the evolution of the flow stress is different. The elastic strain $\varepsilon_{ey}$ and viscoplastic strain $\varepsilon_{vp}$, defined $\varepsilon_{vp} = \varepsilon - \varepsilon_{ey}$, do not remain constant as the penetration of the sphere into the material increases. In Fig. 15, the elastic strain is compared to the viscoplastic strain. This curve is very interesting as it can be used to establish some correlation between compression and scratch tests. The viscoelastic behaviour observed in scratching experiments holds for $\sigma R < 0.2$ and a true strain of 0.03 until the curves $\varepsilon_{vp}/\varepsilon_{ey}$ for PMMA and CR39 deduced from the compression tests are perfectly superimposed (Fig. 15a). The viscoplastic contact behaviour of the two polymers becomes different as the ratio $\sigma R$ exceeds 0.45 (Figs. 5 and 6) and the curves $\varepsilon_{vp}/\varepsilon_{ey}$ deviate at the true strain $\varepsilon = 0.12$ (Fig. 15a). According to these two sets of values, the ratio of the true representative strain in scratch test to $\sigma R$ lies in the range 0.15–0.27, which corresponds approximately to the coefficient 0.2 given by Tabor [9] for indentation on metals Eq. (1). The difference observed between the two materials in simulation is directly linked to the difference in the evolution of $\varepsilon_{vp}/\varepsilon_{ey}$ (Fig. 15b).

Since $\varepsilon_{vp}/\varepsilon_{ey}$ is always larger for PMMA than for CR39, this material exhibits a more pronounced plastic behaviour, i.e., forms higher push pads and has a lower rear contact angle (Figs. 10–12).
Although the ratios \( \alpha/R \) are the same for penetration depths of 5 and 4 \( \mu \text{m} \) on CR39 and PMMA, respectively, the plastic strains involved in the deformation are completely different (Figs. 16 and 17). It is particularly important to recall here that even if one often refers to the representative strain as above, the strains are not homogeneous near the indenter, the plastic strain being maximal below the tip and decreasing with increasing distance from the indenter. The plastic strain increases as the penetration depth increases and for \( h = 20 \mu \text{m} \) the maximum values are about 1.75 and 0.63 for PMMA and CR39, respectively. This means there is a ratio of three between the plastic strains in the two materials, for a given value of \( \alpha/R \). It is not surprising to observe such a difference since the ratio \( \varepsilon_{vp}/\varepsilon_e \) is two to three times higher for PMMA than for CR39 (Fig. 15b). Thus, the plastic strain depends not only on \( \alpha/R \) but also on the strain hardening and on the rheology of the material, i.e., the ratio of Young’s modulus to the yield stress.

6. Conclusion

Experimental and numerical analyses of the scratch test with a spherical tip were carried out at various penetration depths on two polymeric materials, a thermosetting resin (CR39) and a thermoplastic polymer (PMMA). According to compression testings, the main difference between these two materials is the large ability of CR39 to strain harden, whereas PMMA softens at low strain. As the sphere penetrated more deeply into the material during scratching, i.e., as the representative strain increased, three deformation modes were observed: viscoelastic sliding, a viscoelastic–plastic contact and viscoplastic scratching. Since the elastic strain at yield \( \varepsilon_y \), the ratio of the yield stress to the Young’s modulus, is comparable for the two polymers, they displayed similar behaviour in scratching experiments in the first two regimes. As the representative strain increases, the deformation in the contact region becomes mainly viscoplastic and the behaviour of the polymers could be clearly differentiated: PMMA formed more pronounced push pads, while even at
the largest penetration depths an instantaneous recovery of CR39 at the rear of the indenter was still visible.

Numerical modelling of scratching on PMMA and CR39 was performed using three dimensional finite element software. Even if the numerical simulation is not able to recover precisely the experimental results, we observed a clear similarity between the numerical results and the experimental ones, especially for the evolution of the apparent friction coefficient and the rear contact angle with the representative strain. In addition, numerical simulation demonstrates clearly that the flow at high penetration depth and so high strain is strongly dependent on the strain hardening of the polymer: its elimination induces the formation of a chip which is not strongly dependent on the strain hardening of the polymer. Hence, the greater the ability of a polymer to strain harden, the better its scratch resistance.

References