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Research article

Influence of curing thermal history on cross-linking degree of a polydimethylsiloxane: Swelling and mechanical analyses

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Abstract. In this work, the change of the elastic properties induced by a change in cross-linking conditions of polydimethylsiloxane is investigated by measuring its shear modulus by dynamic mechanical analysis and correlating it to that predicted from swelling measurements. Polymer cross-linking is performed at different curing temperatures reached with ramps at different heating rates. From both mechanical and swelling measurements, the molecular weight between cross-links, M_C, is determined, and its dependency on the applied thermal history is analyzed. The main results are: (i) the elastic modulus of the cured material is not significantly affected by the heating rate adopted, while (ii) the curing temperature has a significant influence on the polydimethylsiloxane mechanical properties. In addition, (iii) M_C evaluation from swelling measurements is in good agreement with that estimated from mechanical measurements when appropriate theories are considered. This last result suggests that swelling experiments can be considered as a reliable tool to predict the elastic modulus of the polydimethylsiloxane studied. The quantitative information reported in this paper, also obtainable by the suggested method if other thermal curing histories are applied, is extremely useful for the proper design of devices based on polydimethylsiloxanes.

Keywords: *rheology, mechanical properties, Sylgard 184, swelling analysis, crosslinking degree*

1. Introduction

Polydimethylsiloxanes (PDMSs) are being used in a wide range of applications for their outstanding characteristics such as good physiological inertness, good biocompatibility, high temperature and humidity stability, as well as high deformability [1]. Due to its mechanical and physical behavior, PDMS is used in several applications such as medical sensors, electronic devices like micro-electro-mechanical systems (MEMS), and in smart soft composites [2–7]. PDMSs are chemically cross-linked elastomers with alternating silicon and oxygen atoms in the backbone chain and side methyl groups. It is known that the cross-linking kinetics [8–10] and the structure of the formed network and thus the relevant mechanical properties [11, 12] are significantly affected by the thermal history applied during the cross-linking

process. The use of PDMS materials such as Sylgard 184, the one considered in this study, in products such as medical devices, soft morphing structures, soft robots and actuators for flexible electronics is widely reported in the literature [13–18]. The mechanical performance of these devices strongly depends on the behavior of the constituent materials. For example, Colombo *et al.* [14], have shown how the curing time affects the mechanical properties of the PDMS used for mock arteries production. To correctly reproduce the behavior of the artery, a proper curing history was necessary. Therefore, knowing how the PDMS mechanical properties depend on the curing history is very important for proper material and device design. Some results about the dependency of the elastic modulus of the PDMS considered on the thermal history applied during the cross-linking

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process have already been reported in the literature [2, 11, 12, 14]. Johnston *et al.* [11] compared the mechanical properties of the PDMS cured under different thermal histories but limited them to the curing conditions suggested by the material supplier [19]. Placet and Delobelle [12] investigated its dynamic-mechanical response and Colombo *et al.* [14] examined the effect of the curing time on its mechanical properties at just one fixed temperature. To the authors' knowledge, no systematic study on the curing conditions' effect on the mechanical properties of the resulting material has been reported up to now.

In a previous paper [20], the authors performed a systematic characterization of the rheological behavior of the same PDMS carried out in dynamic conditions during the cross-linking process performed under different thermal histories and this study allowed us to describe the network forming kinetic and compare it with existing literature [8–10, 21]. The focus of the present work is on the evaluation of how the adopted thermal history affects the PDMS network structure and, consequently its mechanical properties. To this aim, a systematic study of the dynamic mechanical properties of the cured PDMS has been carried out, considering the effect of both the temperature at which the curing is performed and the heating rate adopted to reach this temperature.

Swelling measurements were also performed on the differently cured PDMS to estimate the values of the mean molecular weight between cross-links, M_C, for the different networks produced. The results obtained are comparable to the M_C values determined from the shear modulus of the cross-linked PDMS following the theory of rubber elasticity.

2. Experimental
2.1. Material
The PDMS employed in this work is the commercial Sylgard 184 provided by Dow Corning, Michigan, United States. It is a two-part liquid component system consisting of an elastomer base (component A) and a curing agent (component B). Both components are liquid and transparent. Typically, the elastomer base contains linear PDMS pre-polymers with two vinyl end groups that react with the component B multifunctional crosslinker leading to a three-dimensional cross-linked network. The exact composition of Sylgard 184 is proprietary, anyway, the materials safety data sheet states that the elastomer base (component A) contains mainly *dimethylvinylsiloxy-terminated* (CASRN:

68083-19-2), *dimethyl siloxane* and *dimethylvinylated* and *trimethylated silica* (CASRN: 68988-89-6) and the curing agent (component B) contains mainly *siloxanes* and *silicones*, *dimethyl*, *methylhydrogen* (CASRN: 68037-59-2), *dimethylvinylsiloxy-terminated* (CASRN: 68083-19-2), *dimethyl siloxane*, *dimethylvinylated* and *trimethylated silica* (CASRN: 68988-89-6) and *methyl-vinylcyclosiloxane* (CASRN: 2554-06-5).

The two components were mixed at room temperature in 10:1 (A:B) mass ratio with a low-speed helix mixer (200 rpm speed, helix diameter 20 mm, beaker diameter 30 mm) for 10 min, as suggested by the supplier [19]. Then the mixture was degassed in a vacuum bell jar for 20 min (low vacuum $P = 9 \cdot 10^4$ Pa) and kept at 4 °C to avoid premature cross-linking before testing. Rheological experiments were performed within 5 h after mixing. It will be shown in Section 4 that no variation in the mixture properties occurs during this period.

2.2. Rheometry
Dynamic mechanical analyses in shear mode were performed with an Anton Paar MCR 502 rheometer in plate-plate configuration (plate diameter 25 mm), to measure the complex shear modulus evolution under different thermal histories. These could be applied by means of the Peltier plate and hood system H-PTD 200 which equipped the rheometer.

The uncured PDMS mixture was first poured on the rheometer lower plate, then the upper plate was moved toward the lower one and positioned at a distance of 1 mm. The material in excess was trimmed before starting the test. All rheological measurements were carried out at a constant frequency of 1 Hz, whereas the strain amplitude was set to 5% for values of the complex modulus, G^*, up to $4 \cdot 10^5$ Pa and to 0.5% for larger values. The linear behavior of the material was verified for both the experimental conditions applied. All the measurements were performed in air and at atmospheric pressure. A normal force of 1 N was applied during the tests to avoid sample slip between the plates. All the tests were carried out at least three times to verify the repeatability of the measurement.

The applied thermal histories consist of four steps, listed and detailed below:
1. *isothermal stabilization step:* the sample is kept at 25 °C for 1 min to stabilize the material temperature;

2. *heating step:* the sample is heated at a constant rate up to a defined temperature, T_C. Heating rates from 1 to 8 °C/min were employed. This range of heating rates was chosen based on the previously published kinetics study [20]. Heating rates higher than 8 °C/min do not allow homogeneous heating of the sample. On the other hand, 1 °C/min is a rate low enough that cross-linking can occur almost entirely during sample heating;

3. *isothermal step:* the sample is kept at a constant temperature, referred to as curing temperature T_C, for a curing time, t_C, of 4 h (curing temperatures in the range 65–150 °C were employed);

4. *cooling step:* the sample is cooled to 25 °C at the rate of 8 °C/min and kept at this temperature for 10 min to stabilize its temperature.

Table 1 summarizes the values of the heating rates adopted and the curing temperatures investigated; Figure 1 shows the different thermal histories applied. The thermal histories for which the heating rate, $\dot{T}$ is 8 °C/min and T_C ranges from 65 to 150 °C,

Table 1. Heating rate and curing temperature values of the thermal histories applied for the PDMS curing during the rheological tests.

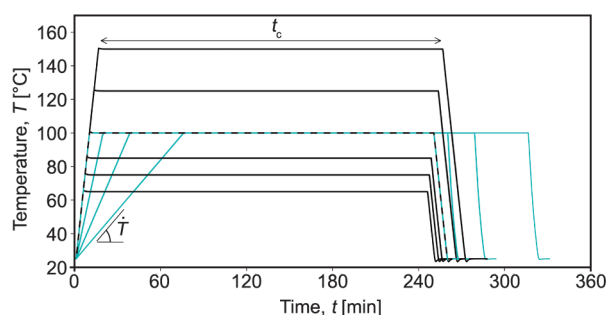| Thermal history code	Heating rate, $\dot{T}$ [°C/min]	Curing temperature, T_C [°C]
T100_R1	1	100
T100_R2	2	100
T100_R4	4	100
T100_R8	8	100
T125_R8	8	125
T150_R8	8	150
T65_R8	8	65
T75_R8	8	75
T85_R8	8	85

Figure 1. Temperature *vs.* time of all thermal histories applied for the PDMS curing during the rheological tests. In black thermal histories for which the heating rate, $\dot{T}$ is 8 °C/min, in light blue thermal histories for which $T_C = 100$ °C and $\dot{T}$ ranges from 1 to 8 °C/min.

which were performed to investigate the effect of the curing temperature on the properties of the PDMS are reported in black. The thermal histories for which $T_C = 100$ °C and $\dot{T}$ ranges from 1 to 8 °C/min, performed to study the effect of the heating rate on the PDMS properties, are reported in light blue.

2.3. Swelling measurements

The cylindrical samples (thickness = 1 mm, diameter = 25 mm) obtained at the end of all the rheological experiments performed with a heating rate $\dot{T} = 8$ °C/min were cut in smaller specimens (about one-third of the cylinder volume) and were used for swelling measurements. All swelling measurements have been performed at room temperature, 25 °C. The initial mass of each specimen, m_0, was measured before immersing it in a large excess of toluene for 24 h to reach the equilibrium state. Then the swollen specimen mass, m', was measured, after lightly pressing the specimen between two pieces of filter paper. The specimens were then left to dry at room temperature and atmospheric pressure for 12 h and the mass of the dried specimen, m, was measured again. The drying time of 12 h allowed the complete release of the toluene dissolved in PDMS, as indicated by the fact that no further mass loss was observed even after subsequent drying of the sample in a vacuum. From these measurements, the polymer extractable fraction, w_{ext} and the volume fraction of polymer in the swollen network, v_2 can be obtained as Equations (1) and (2):

$$w_{ext} = \frac{m_0 - m}{m_0} \tag{1}$$

$$v_2 = \frac{\frac{m}{\rho}}{\frac{m' - m}{\rho_s} + \frac{m}{\rho}} \tag{2}$$

where m_0, m' and m are the initial, swollen and dry mass of the specimen as described above, ρ_s is the solvent density (0.865 g/cm³ for toluene at room temperature [22]) and ρ is the density of the dry polymer after the extraction of the free chains. This last property was derived from the polymer density measured before extraction, ρ_0, according to Equation (3):

$$\rho = \rho_0 \cdot \left(1 - w_{ext_m}\right) \tag{3}$$

where w_{ext_m} is the average value of the measured extractable fractions, w_{ext}.

The polymer density ρ_0 was measured at 25 °C with a pycnometer and resulted to be 1.070±0.015 g/cm³.

3. Theoretical analysis

According to the theory of rubber elasticity, the rubber shear modulus is correlated with the mean molecular weight between cross-links, M_C. Considering the 'phantom network' assumption [23] the following relation can be proposed (Equation (4)):

$$G = \frac{\rho_0 \cdot R}{2M_C} \cdot T \qquad (4)$$

where G is the shear modulus of the rubber, ρ_0 is the polymer density, R is the gas constant (8.314 J/(mol·K)) and T is the absolute temperature. The mean molecular weight between cross-links M_C, can also be correlated to the swelling of ideal rubbers, through on the Flory-Huggins theory of polymer solutions [24]. According to different hypotheses, different correlations were proposed [25–27]. Equations (5), (6) and (7) describe the equilibrium swelling with the functionality of the cross-link equal to 4, according to Flory and Rehner [25], Rennar and Oppermann [26] and Robert *et al.* [27], respectively:

$$-\frac{\rho}{M_C} = \frac{\ln(1 - v_2) + v_2 + \chi \cdot v_2^2}{V_s \cdot v_2^{1/3}} \qquad (5)$$

$$-\frac{\rho}{M_C} = \frac{\ln(1 - v_2) + v_2 + \chi \cdot v_2^2}{V_s \cdot \frac{v_2^{1/3}}{2}} \qquad (6)$$

$$-\frac{\rho}{M_C} = \frac{\ln(1 - v_2) + v_2 + \chi \cdot v_2^2}{V_S \cdot \left(v_2^{2/3} - \frac{v_2}{2}\right)} \qquad (7)$$

where ρ is the dry polymer density, M_C is the mean molecular weight between cross-links, V_S is the molar volume of the solvent (for toluene 106.52 cm³/mol [22]) and v_2 is the volume fraction of the polymer in the swollen network. χ is the solvent-polymer interaction parameter: it depends on both the solvent and the polymer and may vary somewhat with concentration and with the extent of cross-linking [28]. The determination of the toluene-PDMS interaction parameter is not trivial and different values can be found in the literature [29]. According to Robert *et al.* [27] the toluene-PDMS interaction parameter can be calculated as $\chi = \chi_0 + \beta \cdot v_2$ where $\chi_0 = 0.44$ and $\beta = 0.36$. Differently, Bueche [28] considered a constant toluene-PDMS interaction parameter $\chi = 0.465$. For the sake of clearness in the present work, these two

proposals for the solvent-polymer interaction parameter will be referred to as χ_R (Robert) and χ_B (Bueche), respectively. In our analysis, for the determination of M_C from swelling experiments χ_R was adopted when Equation (7) was used while either χ_R and χ_B were adopted when Equation (5) and (6) were used. M_C values obtained from swelling tests were then compared to those obtained from rheological analysis.

4. Results and discussion
4.1. Rheometry

As explained in Section 2.2, different thermal histories have been carried out to cure the PDMS (Figure 1) and the change in time of its complex shear modulus G^* was monitored.

After mixing the elastomer base and the curing agent, the mixture was kept at 4 °C and the rheological experiments started within 5 h. Figure 2 shows that no systematic and significant variation in the properties of the mixture occurs during this period, suggesting that the material is stable if stored at low temperatures.

The rheological analysis performed allows to monitor the evolution in time of the complex shear modulus during the whole curing history. All the details about the rheological curves are shown and explained in [20]. In this work we limited our attention to G^* at the end of the applied thermal history, investigating the effect of the thermal history. The loss factor, $\tan\delta$, measured at 25 °C for the cured material,
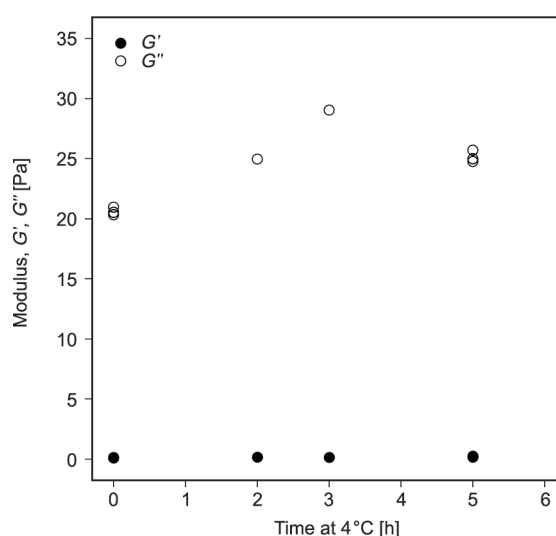

Figure 2 Storage modulus, G' and loss modulus, G'' of the uncured PDMS just mixed as a function of the time in which the mixture is kept at 4 °C before applying the desired thermal history.

is small (tan δ < 0.096), thus the dissipative component of the complex modulus is very limited compared to the elastic one. The PDMS displays therefore a behavior very close to that of an ideally elastic material, and its complex modulus almost coincides with its elastic modulus.

G values measured at the end of each thermal history (G is assumed equal to G^*, as explained before) are reported in Figure 3 for the different thermal histories, as a function of (Figure 3a) the heating rate for the curing temperature $T_C = 100\,°C$ and of (Figure 3b) the curing temperature for the heating rate $\dot{T} = 8\,°C/min$.

As shown in Figure 3a, the heating rate has no evident effect on the properties of the cured PDMS.

On the other hand, Figure 3b shows that the curing temperature has a significant influence on the PDMS mechanical response: G at 25 °C increases from 0.65±0.06 to 1.25±0.03 MPa when the curing temperature is increased from 65 to 150 °C.

During the cross-linking process, elastomer base and curing agent react to form an infinite network. Figure 3b shows that varying the temperature at which this process takes place, the mechanical properties change, and this indicates that the network that is formed has been modified. Usually, the amount of curing agent is in excess of the elastomer base because in addition to the main hydrosilylation reactions, other secondary reactions take place. This can change the way in which the molecules of the two components react, affecting the structure of the resulting network and thus its mechanical properties, as explained in Section 3. Higher temperature leads

to the formation of a stiffer PDMS, indicating the formation of more cross-links, probably because fewer secondary reactions take place at a higher temperature.

Literature [11, 12, 14] reports that the structure of the formed network and the consequent mechanical properties are significantly affected by the thermal history applied during the polymer network formation. In our analysis, the influence of the thermal history has been split into two contributions: the effect of the heating rate and the effect of the curing temperature, and we have shown that the major contribution to the mechanical properties change is given by the curing temperature at which the cross-linking takes place, independently of the heating rate applied to reach this temperature in the first stages of the cross-linking.

4.2. Swelling

The values of the initial mass of each specimen m_0, the swollen specimen mass, m', and the mass of the dried specimens, m, are reported in Table 2. As can be seen, the mass of the swollen specimens almost doubles the initial mass of the specimens, thanks to the good interaction between polymer and solvent. Instead, the mass of the specimens dried after swelling is almost equivalent – slightly lower – to the initial mass. This small difference can be attributed to the removal of an excess unreacted component of the mixture.

Polymer extractable fraction, w_{ext} and volume fraction of polymer in the swollen network, v_2 have been calculated for all the PDMS cured at different curing temperatures according to Equation (1) and (2),
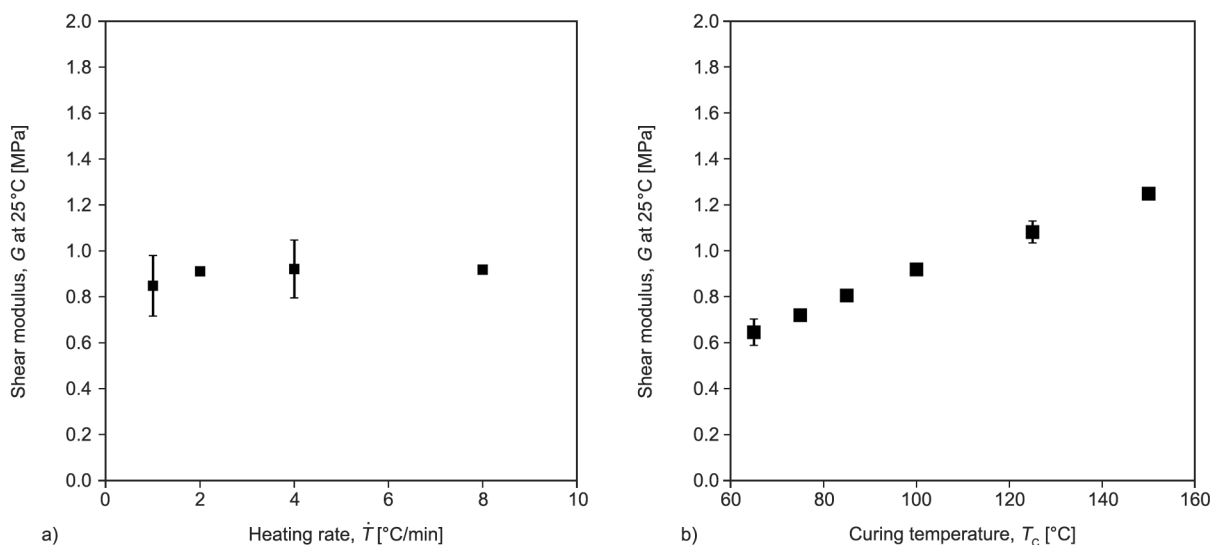

Figure 3. Shear modulus, G measured at 25 °C, at the end of the cooling step *vs.* (a) heating rate, $\dot{T}$ and (b) curing temperature T_C applied in the curing process.

Table 2. Initial mass of the specimens, m_0, swollen specimens mass, m', and mass of the dried specimens, m, measured during swelling tests.

Sample name	Thermal history code	m_0 [g]	m' [g]	m [g]
a1	T65_R8	0.1016	0.2050	0.0967
a2	T65_R8	0.1225	0.2402	0.1173
a3	T65_R8	0.0912	0.1876	0.0872
b1	T75_R8	0.0847	0.1688	0.0807
b2	T75_R8	0.0885	0.1754	0.0841
b3	T75_R8	0.0893	0.1737	0.0849
c1	T85_R8	0.0661	0.1300	0.0631
c2	T85_R8	0.1009	0.1955	0.0958
c3	T85_R8	0.0688	0.1335	0.0655
d1	T100_R8	0.0984	0.1869	0.0936
d2	T100_R8	0.0971	0.1826	0.0925
d3	T100_R8	0.1119	0.2108	0.1064
e1	T125_R8	0.1329	0.2473	0.1267
e2	T125_R8	0.1026	0.1865	0.0980
e3	T125_R8	0.0788	0.1458	0.0753
f1	T150_R8	0.1152	0.2060	0.1099
f2	T150_R8	0.0698	0.1240	0.0668
f3	T150_R8	0.1094	0.1971	0.1045

respectively, and are reported in Figure 4 *versus* the curing temperature.

The extractable fraction (Figure 4a) seems not to be affected by the curing temperature and varies between 4 and 5%, with a mean value of 4.7±0.2%.
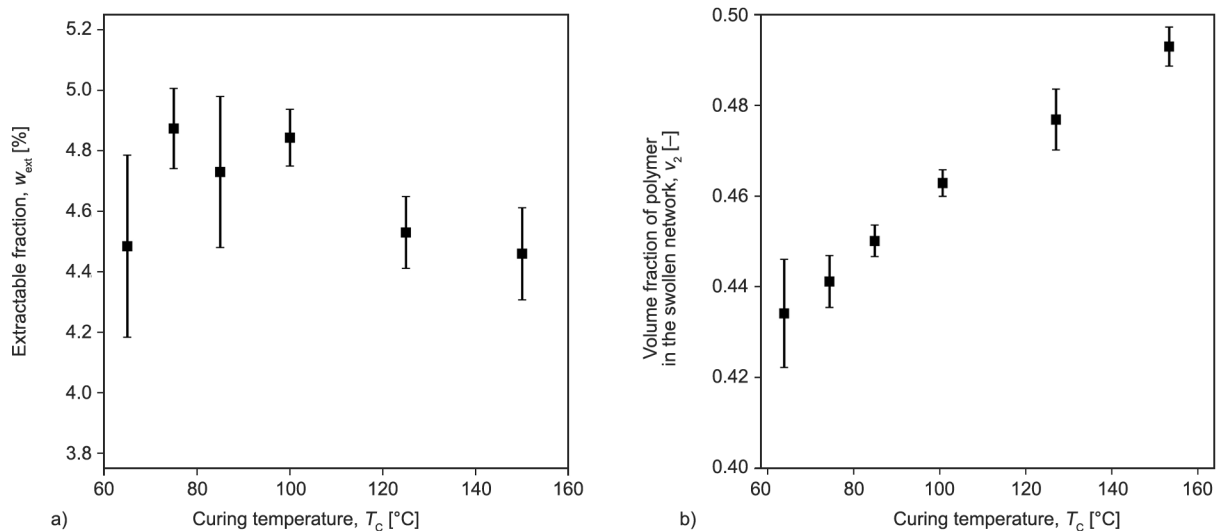
As expected, the volume fraction of polymer in the swollen network v_2 increases with the curing temperature (Figure 4b). Higher temperatures have been shown to lead to the formation of stiffer PDMS (higher G, as shown in Figure 3b), suggesting that the PDMS formed has higher density of cross-links,

corresponding to lower molecular weight between cross-links. When a cross-linked polymer is immersed in a good solvent, the network absorbs the solvent until an equilibrium is reached. In particular, the higher the density of cross-links, the lower the amount of solvent absorbed and, consequently, the quantity of polymer in the swollen sample (v_2) is larger.

4.3. Mean molecular weight between cross-links

The mean molecular weight between cross-links, M_C was estimated, according to Equation (4), from the experimental value G at 25 °C of the PDMS cured at different curing temperatures. As shown in Figure 5 (black crosses), it varies from 2066±189 g/mol for $T_C = 65$ °C to 1062±22 g/mol for $T_C = 150$ °C. As expected [24], the higher G, the lower the corresponding M_C.

The mean molecular weight between cross-links obtained from swelling measurements evaluated according to Equations (5), (6) and (7) as reported in Section 3 are also shown in Figure 5 (colored markers). The values of M_C obtained from the swelling data depend strongly on both the equation and the solvent-polymer interaction parameter, χ adopted. The differences among the results arise from the different assumptions on which models are based [29, 30]. It is beyond the purpose of this article to discuss each model assumption validity for the material under study. We limit our analysis to the assessment of the more appropriate theory based on the consistency of estimates from swelling experiments and dynamic mechanical analyses.

Figure 4. (a) Polymer extractable fraction, w_{ext} and (b) volume fraction of polymer in the swollen network, v_2 *vs*. curing temperature, T_C.
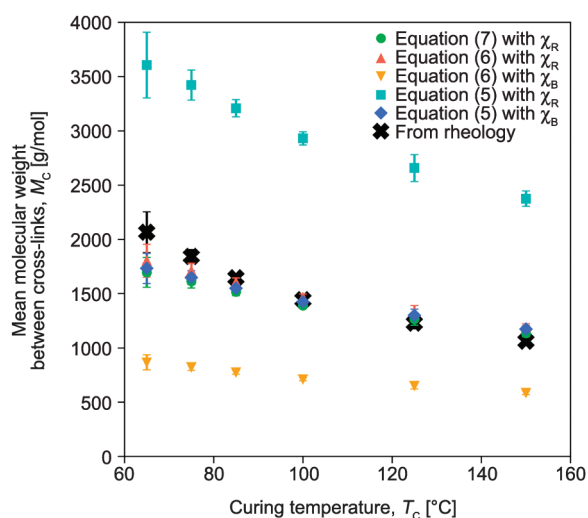
Figure 5. Mean molecular weight between cross-links *vs.* curing temperature. Black crosses represent data from rheological analysis and colored markers are data from swelling analysis. For the latter data, values obtained according to Equation (5) from Flory and Rehner, Equation (6) from Rennar and Oppermann and Equation (7) from Robert are reported.

As can be seen in Figure 5, the theory from Robert *et al*. [27] (● green dots), the theory from Rennar and Oppermann [26] with the toluene-PDMS interaction parameter χ_R from Robert *et al*. [27] (▲ red up triangles) and the theory from Flory and Rehner [25] with χ_B from Bueche [28] (◆ blue diamonds) lead to very similar results. On the other hand, M_C from Flory and Rehner theory [25] with χ_R from Robert *et al*. [27] (■ light blue squares) are significantly higher than those from the theories just mentioned, while the results of the analysis based on the theory from Rennar and Oppermann [26] with the toluene-PDMS interaction parameter χ_B from Bueche [28] (▼ orange down triangles) are significantly lower. Furthermore, M_C calculated from the elastic modulus (✖ black crosses in Figure 5) are in good agreement with M_C obtained with the first three mentioned swelling models (● green dots, ▲ red up triangles and ◆ blue diamonds in Figure 5).

This result clearly indicates that the theory from Robert *et al*. [27], the theory from Rennar and Oppermann [26], with χ_R and the theory from Flory and Rehner with χ_B can be adopted to estimate M_C out of swelling experiments on the PDMS studied.

5. Conclusions

In this work, the change of the network structure and mechanical properties of PDMS Sylgard 184 after

cross-linking was investigated in a systematic way by varying the heating rate and the curing temperature. To this aim, two approaches were considered: the measurement of the complex shear modulus by dynamic mechanical analyses during the whole curing process and swelling measurements on the cured elastomers.

The shear modulus of the cured material is not significantly affected by the heating rate adopted. On the other hand, the curing temperature has a significant influence on the PDMS mechanical properties. From both rheological and swelling measurements, the molecular weight between cross-links, M_C, has been estimated and its dependency on the applied thermal history has been investigated. The shear modulus of the cross-linked PDMS has been related to M_C according to the theory of rubber elasticity. Furthermore, several correlations between M_C and the volume fraction of the polymer in the swollen network have been considered, following theories based on the one initially proposed by Flory. A good agreement between the two approaches has been found and this confirms that changes in the network structure are the main cause of the changes in the mechanical properties.

On the basis of the good agreement obtained, M_C estimates based on simple swelling experiments and the appropriate theories mentioned above can be considered a reliable tool to predict the shear modulus of the PDMS considered. This might simplify the material characterization after any curing cycle since both a dynamometer and relatively large specimens with a proper shape are required to perform a mechanical test, while only a precision weighing scale balance is necessary for swelling measurements, and a small sample of any shape is suitable for this kind of measurements.

With our investigation of the effects of heating rate and curing temperature on the structure of the network and on the mechanical properties of the PDMS studied, we hope to provide quantitative information for researchers designing applications employing the PDMS studied cured under certain conditions as a part of more complex structures.

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