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# Post-curing and structural relaxation of epoxy networks during early stages of aging for civil engineering applications

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## Abstract

In this work, epoxy networks used for civil engineering constructions are aged at ambient temperature, and the impact of the early stages of aging on mechanical properties is analyzed. It is observed that Young's modulus and the yield stress increase with the aging time whereas the strain at break and the toughness decrease. The structural causes for these variations are investigated by swelling experiments, infrared spectroscopy, thermogravimetric analysis, differential scanning calorimetry, and dynamic mechanical analysis. By combining experimental results, it is deduced that the aging process occurs in two stages. During the first 7 days of aging, the change in mechanical properties should essentially be attributed to the postcuring, which results in additional crosslinking reactions. When increasing the aging time to 21 days, the crosslinking density reaches a plateau, but a second stage emerges, in which the mechanical properties continue evolving in the same way, with variations of the mechanical parameters being of comparable amplitude. This second stage is attributed to the evolution of the thermodynamic state of the glass, leading to a densification of the epoxy network through minimization of the free volume, and favoring the creation of physical interactions between polymer segments. Therefore, structural relaxation is shown to have a significant impact on the mechanical properties.

## KEYWORDS

dynamic mechanical analysis, physical aging, tensile tests

## INTRODUCTION

The addition of epoxy resin helps protecting concrete structures from water infiltration. This procedure increases the durability of civil engineering constructions which can last up to 100 years [1]. During service, the epoxy networks are confronted to several aging mechanisms, which may affect their properties. The commonly identified mechanisms impacting epoxy networks are in general the hydrolytic degradation [2], the photodegradation [3, 4], the thermal oxidative degradation [5, 6], the loss of additive, the post-curing [7],

and the structural relaxation [8], the latter being the object of this study.

The structural relaxation occurs in any glass stored below its glass transition temperature. It consists of the minimization of its thermodynamic quantities, such as enthalpy and specific volume, through conformational rearrangements, to approach equilibrium [9-21]. In glassy polymers, this phenomenon can affect the dimensional stability during storage. It may also induce changes of mechanical [22] and barrier properties [23], as examples. It is sometimes neglected since it is a long-time process, which makes

its study very challenging; however, it becomes a concern for any application targeting durability.

In the case of epoxy networks used in civil engineering, the study of structural relaxation even deserves a higher interest. Indeed, to ease the process, the epoxy mixture is generally applied then cured under ambient conditions. Therefore, these specific epoxy systems generally exhibit a glass transition temperature, which is higher than the temperature of service to ensure good mechanical and barrier properties, but sufficiently low to facilitate the total curing. They typically serve at temperatures situated about 20-30 K below the glass transition, at which the structural relaxation proceeds sufficiently fast to consider it has a real impact on the material properties [24].

Determining how these properties evolve under structural relaxation is still a matter of investigations. One can expect an increase of the barrier, which would be interesting to enhance the protective role of epoxy toward concrete. On the other hand, different parameters influence the mechanical properties evolution. An increase of Young's modulus and yield stress could be seen as improvements while a decrease of the strain at break may be highly detrimental for the concrete properties. If the epoxy loses its ductility (disappearance of the plastic domain), some cracking can occur which would likely lead to the loss of the epoxy barrier.

Investigating the variation of enthalpy toward equilibrium is the most common way to probe the structural relaxation. This is typically managed from calorimetric measurements. The analysis is performed on a small size sample, and under neutral atmosphere, to prevent the interfering of other degradation mechanisms. Nevertheless, it is still interesting to confront the material to a media that mimics environmental conditions. In the present study, the case of humid environment is not explored. Besides, the networks are annealed in an environment insulated from UV rays. The epoxy networks are aged at ambient temperature, under air, in a dry environment. Obviously, under these conditions, other mechanisms cited above, for example, thermal oxidation, may also influence the behavior of the epoxy networks. Therefore, it is mandatory to limit the observation to a time window during which the structural relaxation is expected to be the main aging mechanism. We assume that during the early stages of aging, neither additive loss nor oxidation should affect the mechanical properties. On the other hand, postcuring could have a major impact.

The curing of an epoxy resin involves the conversion of reactive species in a crosslinking reaction. In many cases, the oxirane cycle opens when reacting with the amine functions of linear or cyclic diamine hardeners, which initiates the formation of a tridimensional network. Due to the increase of the viscosity associated with the network formation, the chain segment mobility is hindered. The gel percentage may be less than 100%. In that case, some species remain reactive into the material days after the formation of the network. These reactive species can be converted when the material is heated above its glass transition [25-28]. This is related to the increase of molecular mobility, easing the chain motion, and favoring interactions between uncured parts. Since the structural

relaxation of the polymer in the glassy state also occurs through conformational rearrangements of the chain segments [13]. It can also give rise to supplementary crosslinking reactions. To investigate the influence of the structural relaxation on mechanical properties, one must ascertain that postcuring does not occur during the period of study.

Thus, in this study, the evolution of the epoxy network crosslinking density is assessed from swelling experiments, differential scanning calorimetry (DSC), and dynamic mechanical analysis (DMA). Thermogravimetric analysis (TGA) and Fourier transform infrared spectroscopy (FTIR) are respectively used to check that the epoxy networks do not undergo additive loss nor oxidation during the aging. Finally, the evolution of the mechanical properties during structural relaxation is followed thanks to tensile and Charpy shock tests.

## EXPERIMENTAL SECTION

The materials used in this work are industrial systems. The epoxy networks were obtained by mixing a prepolymer of bisphenol A diglycidyl ether (BADGE) with a hardener mixture containing triethylene tetramine mixed with a cycloaliphatic diamine hardener, produced by SPPM (Société Parisienne des Produits et Matériaux). The proportions of resin versus hardener are, respectively, 100:58 in weight and 1.5:1 in volume. Benzylic alcohol was also added as a plasticizer (about 6% mass) to improve the processing and ease the curing, which was performed in ambient conditions because they are representative of civil engineering ones. After homogenization, the mixture was poured into both rectangular and dog bone section silicon molds for Charpy impact tests and tensile tests, respectively. The applicability of silicon molds for the curing of epoxy resins has been extensively studied recently [29]. The samples were obtained after 5 days of polymerization (curing) since a minimum amount of time is required to get acceptable rigidity and dimensional stability. Then, they were removed from the silicon molds to be put in a furnace under air, at 25°C, protected from UV light. During the annealing, they were held and wedged by stainless steel sheets of 2 mm thickness. In the following part, the term "unaged material" refers to a sample that has been cured during 5 days but not annealed in a furnace while aging times of 7, 14, and 21 days correspond to the annealing duration.

The crosslinking density of the epoxy networks was qualitatively investigated from gravimetric measurements consecutive to an immersion in acetone for various durations. The progressive swelling of the network was followed by calculating the degree of swelling OS [30] from

$$OS = \frac{t.m}{m_0} = \frac{m_t - m_0}{m_0}, \quad (1)$$

where  $m_t$  is the sample weight for a given  $t$  immersion time,  $m_0$  is the sample weight before immersion, and  $t.m$  is the variation of weight consecutive to the immersion.

TGA experiments were performed from 30°C to 600°C in a Thermal Analysis® Discovery apparatus under nitrogen atmosphere.

Samples annealed during 21 days were put in platinum pans compatible with high temperature. The heating rate was 10 K/min.

The nitrogen flow was 25 ml/min. The rate of the nitrogen balance purge flow was also equal to 25 ml/min. The TGA apparatus is calibrated thanks to mass standards, and by using the Curie point of nickel as temperature reference. Before the analyses, a baseline was performed with an empty furnace.

Modulated temperature differential scanning calorimetry (MT-DSC) measurements were done on 5-7 mg samples in a Q2000 apparatus (TA Instruments®) under a 50 ml/min flow of nitrogen. Three successive scans were performed from 0°C to 120°C (100°C for the unaged sample). A heat-only modulation procedure was used, with a heating rate of 2°C/min, a period of 60 s, and an amplitude of ±0.318°C. The calibration in temperature and energy was normally carried out using standard values of indium, and the specific heat capacities for each sample were measured using sapphire as a reference. MT-DSC allows separating the glass transition signature, which is obtained in the reversing signal from the kinetic events, which appear in the nonreversing signal.

Dynamic mechanical analyses were performed with a Q800 apparatus (TA Instruments®) under air on 17 x 10 x 4 mm<sup>3</sup> rectangular shape samples, using the single cantilever mode. The epoxy networks were scanned from 30°C to 115°C at a heating rate of 5°C/min. The oscillation frequency was 1 Hz, and the amplitude of deformation was kept constant at a value of 15 µm.

The infrared spectroscopic details were recorded on an apparatus of the brand Thermo Fischer® Nicolet i510 in transmission mode in the 425-4000 cm<sup>-1</sup> range, with a resolution of 4 cm<sup>-1</sup>. A background correction was performed before the experiment. One hundred fifty individual scans were co-added. Measurements were performed on three samples of 4 mm thickness for each aging condition, to verify the repeatability of the obtained results. Analysis of IR spectra was done using the Omnic apparatus software.

Tensile tests were performed at 0.1 mm/min in a Zwick® mechanical testing machine at ambient temperature, on a set of five dog bone shape samples for each aging duration. The sample dimensions were 25 mm length, 4 mm width, and 3 mm thickness. The data treatment was done using the testXpert II software.

A total of three samples were used for Charpy impact test for toughness measurements. The tests were performed on a CEAST apparatus, with a 2 J hammer, a pendulum speed defined by the purchaser being 2.8 m s<sup>-1</sup>. The samples were 80 mm length, 10 mm large, and 4 mm thick unnotched specimens.

## RESULTS AND DISCUSSION

### Identification of the aging mechanisms

The first part of the discussion aims at determining the aging mechanisms occurring during the annealing of the epoxy networks. As previously

**TABLE 1** Plasticizer loss features in virgin and 21 days annealed samples, and significant absorbance ratios, from preliminary experiments of TGA and FTIR, respectively.

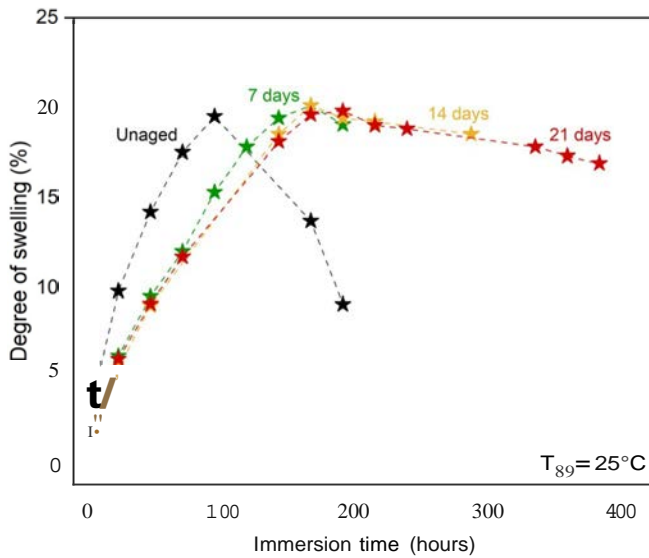
TGA	Virgin	Annealed		
First mass loss onset (°C)	80 ± 2	80 ± 2		
Residual mass at 250°C (wt. %)	95 ± 1	95 ± 1		
FTIR	Unaged	7 days	14 days	21 days
A (1745 cm <sup>-1</sup> /1606 cm <sup>-1</sup> )	0.283	0.118	0.095	0.116
A (914 cm <sup>-1</sup> /1606 cm <sup>-1</sup> )	0.1483	0.1225	0.0822	0.1612

Abbreviations: FTIR, Fourier transform infrared spectroscopy; TGA, thermogravimetric analysis.

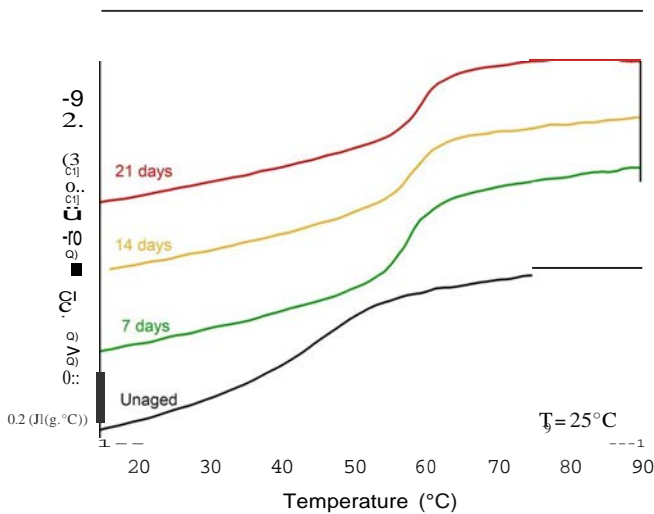
mentioned, the epoxy networks are protected from hydrolytic degradation and photo-degradation. To assess that plasticizer did not migrate from the epoxy network during the annealing, preliminary TGA experiments were performed, which main results are summarized in Table 1 (the results are more extensively discussed in supporting information). Classically, the plasticizer loss is recorded at about 200°C, while the thermal degradation of the epoxy network starts 100°C higher. Both virgin and annealed networks exhibit similar residual plasticizer contents. Thus, the annealing does not induce any plasticizer migration. Oxidation can be evidenced by investigating from FTIR the appearance or growth of peaks, attributed to amide functions, in the domain situated between 1620 and 1800 cm<sup>-1</sup> [31]. In Table 1, are reported the ratios between the absorbance A of the peak recorded at 1745 cm<sup>-1</sup>, being the only apparent peak clearly discernible in this domain, and the peak at 1606 cm<sup>-1</sup>, considered as invariant (these FTIR spectra regions are shown in Supporting Information). A (1745 cm<sup>-1</sup>/1606 cm<sup>-1</sup>) does not increase with the aging time. Thus, the hypothesis of oxidation induced by annealing is also excluded. The conversion of the oxirane cycle, characterizing postcuring, is investigated from the ratio between the amplitudes of the peak at 914 cm<sup>-1</sup> and the peak at 1606 cm<sup>-1</sup> [6]. By increasing the aging time up to 14 days, the ratio decreases, suggesting additional crosslinking. On the other hand, the data recorded for the 21 days aged networks do not follow this trend. Similar variations were observed by investigating the creation of hydroxyl groups from the opening of the oxirane cycle (results not shown there). Thus, complementary results are required.

Figure 1 reports gravimetric results following the immersion in acetone for various aging durations. For the unaged material, the sample mass quickly increases, then reaches a maximum corresponding approximately to an immersion of 100 h, and finally decreases. The gain of weight shows that the epoxy successfully trapped the solvent. It is interesting to notice that a mass loss occurs afterward. We assume that this second step is associated with an extraction by the solvent of the free chains, that is, the uncured parts.

By increasing the aging time to 7 days, the gain of weight is delayed. Finally, the curves corresponding to 14 and 21 days perfectly superimpose. Besides, for all the aged materials, the sample mass reaches a plateau. These results suggest an increase of the crosslinking density, firstly, because one can expect that the solvent diffusion is slowed down



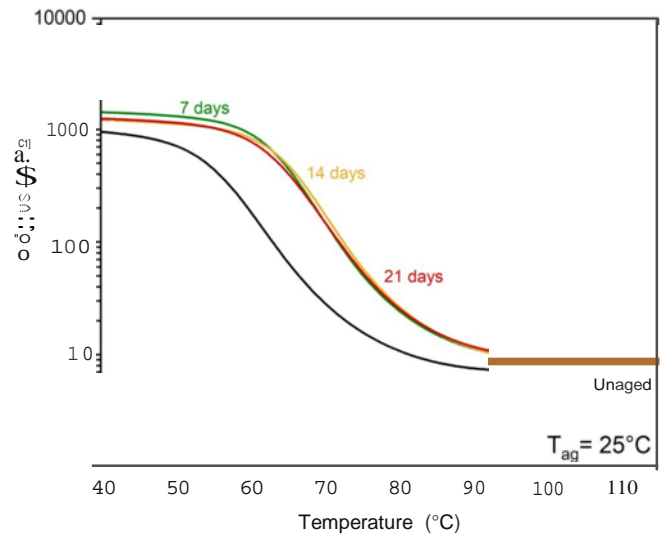
**FIGURE 1** Degree of swelling as a function of the immersion time for unaged epoxy network and epoxy networks having been aged during 7, 14, and 21 days at  $T_{ag} = 25^\circ\text{C}$ .



**FIGURE 2** Modulated temperature differential scanning calorimetry reversing heat capacity signal in the glass transition region for unaged epoxy network and epoxy networks having been aged during 7, 14, and 21 days at  $T_{ag} = 25^\circ\text{C}$ .

into a dense network [28,32], but also because the existence of a plateau indicates that no free chain remains in the aged epoxy.

According to Figure 2, which shows the MT-DSC results, the glass transition of the aged epoxy networks shifts towards higher temperatures in comparison with an unaged material. This result is consistent with our assumption that post-curing proceeds during the first days of aging. Likely, the perfecting of the network also induces its structural homogenization at the local scale. Thus, the distribution of the relaxation temperatures becomes narrower. From 7 to 21 days of aging, the curves look similar, probably because all aged materials exhibit similar crosslinking densities.



**FIGURE 3** Dynamic mechanical analysis storage modulus in the glass transition region for unaged epoxy network and epoxy networks having been aged during 7, 14, and 21 days at  $T_{ag} = 25^\circ\text{C}$ .

The storage modulus of the epoxies has been determined in both glassy and rubbery states, thanks to DMA measurements (Figure 3). The modulus in the rubbery state used to be particularly dependent on the crosslinking density [32], since it is the mechanical response of the system above the glass transition. This means that it is essentially governed by the covalent architecture, taking the lead over the contribution of the interchain connectivity which plays, in contrast, a major role in the glassy state. In comparison with the unaged material, the annealed epoxy networks exhibit higher modulus below and above the glass transition. It is worth mentioning that a shift of the glass transition signature towards higher temperatures is also recorded, as it has been from calorimetric measurements. Finally, the modulus of the unaged material increases with temperature in the rubbery state. This may indicate thermally activated postcuring. This behavior is not observed for the annealed materials.

By combining data resulting from swelling tests, as well as MT-DSC and DMA measurements, it seems reasonable to conclude that the unaged epoxy system undergoes postcuring during the first days of aging. On the other hand, all the data recorded from the same techniques appear similar between annealed samples. For this reason, we assume that the crosslinking density evolution with aging time quickly reaches a plateau. From 14 to 21 days of aging, the only aging mechanism influencing the epoxy properties seems to be the structural relaxation.

### Impact of the structural relaxation on the mechanical properties

Stress versus strain curves obtained from the tensile tests are presented in Figure 4. In consistency with the other results, the aged materials exhibit a higher Young's modulus, a higher yield stress, a



lower toughness, and a lower strain at break than the unaged material, the variations of these mechanical parameters with aging time being given in Figure S. With the increase of the crosslinking density, the epoxy network becomes more rigid and loses ductility while its strength increases.

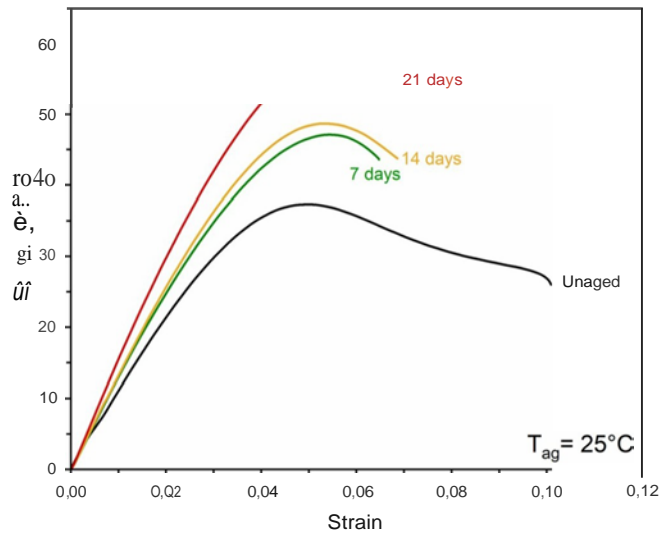
From 14 to 21 days of annealing, a slight increase of rigidity is observed through the increase of Young's modulus, but more intriguing is the increase of the yield stress (Figure 4). As previously discussed, no result obtained from swelling tests, neither MT-DSC or DMA measurements, points toward a continuous increase of the crosslinking density with aging time. For this reason, we assume that this change of mechanical properties is related to the structural

relaxation occurring during physical aging, as it is known that it proceeds by a reduction of the free volume and a densification of the glassy material [33].

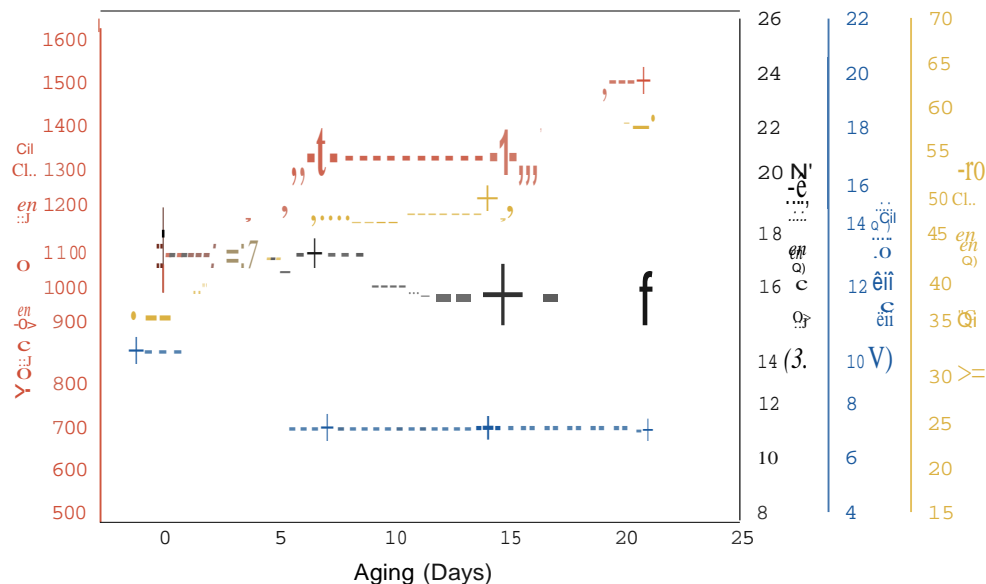
To summarize, the obtained results suggest that the achievement of the postcuring marks the beginning of a second stage of aging during which the mechanical properties of the epoxy network continue evolving, leading notably to a strong increase of the material strength. In structural terms, this second stage differs from the postcuring. The structural relaxation does not involve the creation of new covalent bonds through crosslinking. It lies in the creation of new physical interactions between polymer segments as the system minimizes its free volume.

## CONCLUSIONS

Since epoxy resins used for civil engineering constructions are simultaneously confronted with various degradation factors, like oxidation, hydrolysis, or structural relaxation, as examples, it is useful to get a better understanding of the impact of each one, before trying to investigate the impact of their concomitant actions. In this study, we observe that the first days of the epoxy network in its glassy state are characterized by a modification of its chemical structure during postcuring, through crosslinking, while the polymer starts its way to equilibrium through structural relaxation. Thus, Young's modulus increases, as well as the yield stress, whereas the strain at break and the toughness decrease. The evolution of the mechanical properties is however not restricted to the only changes of the crosslinking density. The polymer still proceeds to macromolecular rearrangements when the post-curing is over. It reorganizes into more ordered conformations, which exhibit higher rigidity and resistance. These results highlight how the structural relaxation occurring during physical aging can strongly affect the mechanical properties of polymers.



**FIGURE 4** Stress-strain curves obtained from tensile tests performed on unaged epoxy network and epoxy networks having been aged during 7, 14, and 21 days at  $T_{ag} = 25^\circ\text{C}$ .



**FIGURE 5** Young's modulus, yield stress, toughness, and strain at break of the epoxy network as a function of the aging duration.

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## CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

## DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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