



### **Science Arts & Métiers (SAM)**

is an open access repository that collects the work of Arts et Métiers Institute of Technology researchers and makes it freely available over the web where possible.

This is an author-deposited version published in: <https://sam.ensam.eu>  
Handle ID: <http://hdl.handle.net/10985/22138>

#### **To cite this version :**

Abir BEN ABDALLAH, Achraf KALLEL, Tarek HASSINE, Fehmi GAMAOUN, Abbas TCHARKHTCHI - Modeling of viscoelastic behavior of a shape memory polymer blend - Journal of Applied Polymer Science - Vol. 139, n°13, p.51859 - 2021

# Modeling of viscoelastic behavior of a shape memory polymer blend

Abir Ben Abdallah<sup>1,2,3</sup>  | Achraf Kallel<sup>4</sup>  | Tarek Hassine<sup>3</sup> |  
Fehmi Gamaoun<sup>5</sup> | Abbas Tcharkhtchi<sup>2</sup>

<sup>1</sup>R&D Departement, Aptiskills, Levallois Perret, France

<sup>2</sup>Laboratoire PIMM, Arts et Métiers ParisTech, CNRS, CNAM, HESAM Université, Paris, France

<sup>3</sup>Laboratoire Mécanique de Sousse, Université de Sousse, ENISO, Sousse, Tunisia

<sup>4</sup>Research Center, Léonard de Vinci Pôle Universitaire, Paris, France

<sup>5</sup>Department of Mechanical Engineering, College of Engineering, King Khalid University, Abha, Saudi Arabia

## Correspondence

Abir Ben Abdallah, R&D Departement, Aptiskills, Levallois Perret, France.  
Email: abir.benabdallah91@gmail.com

## Abstract

Shape memory effect (SME) of polymers is a property that concerns both, macroscopic and microscopic changes. The variation of internal polymer properties such, as molecular weight ( $M_w$ ), rigidity, and viscoelasticity could alter its SME. In this study, a bi-parabolic model with six parameters is used to describe the viscoelastic behavior of a shape memory polymer (SMP) blend (40% poly(caprolactone), PCL/60% Styrene–Butadiene–Styrene) with different PCL  $M_w$ . These parameters are determined using the Cole–Cole method. Modeling curves ( $E'' = f(E')$ ) will be then compared to experimental data from dynamical mechanical analysis (DMA) tests. It is shown that the bi-parabolic model predicts well the behavior of the SMP mixture for different  $M_w$  of PCL. Afterwards, the evolution of the model parameters with the  $M_w$  of PCL is investigated. It is revealed that, when  $M_w$  of PCL drops, the relaxation modulus  $E_0$  increases. This result proves that the rigidity of the SMP blend rises with  $M_w$  declines.

## KEYWORDS

blends, mechanical properties, structure–property relationships, theory and modeling

## 1 | INTRODUCTION

Polymers are characterized by their viscoelasticity. A viscoelastic material represents simultaneously the behavior of an elastic solid with a modulus  $E$  (or  $G$ ) and of a viscous Newtonian liquid with a viscosity  $\eta$ .<sup>1</sup> The blend used in this study is a viscoelastic thermosensitive shape memory polymer (SMP). This polymer has the ability to deform to a temporary shape and to recover its initial shape when exposed to thermal stimulus.<sup>2</sup> In fact, the use of SMP expands in recent years. Due to their high capacity of deformation, fixing and recovery, and to the biocompatibility of some SMP, they are widely used in biomedical and aeronautical fields.<sup>3,4</sup> Although, the shape recovery of SMP is a macroscopic property, their shape memory effect (SME) and mechanism depend on microscopic characteristics such as

morphology, rigidity, internal and viscoelastic properties, and especially molecular weight ( $M_w$ ).<sup>5</sup> It has been shown that the SME might be associated to the irreversible microstructure variation.<sup>5,6</sup> Consequently, even the SMP recovers its SME, it will not regain its original characteristics. Thus, the thermo-mechanical response and the shape memory mechanism depend on these viscoelastic internal properties. Then, it is crucial to model the viscoelastic behavior of SMP to determine the relation between the SME and the viscoelastic properties. In addition, thermosensitive SMP exhibit thermomechanical behavior when they are submitted to a stress or a strain. Their deformation could be large and non-linear. Predicting SMP behavior with applying various approaches is necessary to better understand the shape memory mechanism and to determine the different parameters that influence the SME. This could permit to SMP to

meet various and specific industrial applications. Constitutive modeling can be used to predict the thermomechanical behavior of thermal-induced SMP.<sup>7</sup> Constitutive models of thermosensitive SMP can be classified into two categories: phenomenological models based on the phase transition approach and rheological models based on viscoelastic approach with rate-temperature-dependent properties parameters.<sup>7</sup> Since the rheological models take into consideration the polymer's macroscopic physical phenomena, they are widely used for the characterization of their viscoelastic behavior. These models should be able to describe the phenomenon of creep and relaxation.<sup>8</sup> In our study, rheological modeling is employed. Thermo-viscoelastic constitutive models are composed of classical dashpot and spring elements.<sup>9</sup> The choice of an appropriate model that predicts the viscoelastic behavior of different materials depends on their nature. In the case of polymers, chemical structure, morphology, and physical state are predominant parameters.<sup>1</sup> Thus, thermoplastics, elastomers and thermosets cannot be represented by the same model. They are composed of a combination of dampers (Newtonian viscous elements) and springs (Hookean elastic elements). Among these models, the Zener model, the Maxwell model, and the Kelvin-Voigt model can be cited.<sup>8</sup> The model of Generalized Maxwell is the most used to characterize the behavior of SMP.<sup>10</sup> However, some recent studies<sup>11</sup> have reported that the bi-parabolic model can also be employed. This model is applied to describe the viscoelastic properties of large range of polymeric materials.<sup>12</sup> In this study, the bi-parabolic model will be used to describe the viscoelastic behavior of the SMP blend.

The parabolic element is an analog model whose behavior combines a spring and a damper (dashpot). The representation of the parabolic element by a generalized Maxwell (or Kelvin-Voigt) model requires infinity of springs and dampers.<sup>13</sup> The used bi-parabolic model consists in adding to the set of bi-parabolic elements a parallel spring with a very low rigidity. This model represents the viscoelastic behavior of the polymer through the determination of six parameters. These six constants could be obtained by an optimization process from the complex modulus results. Then to validate the model, the results of thermomechanical tests applying Cole–Cole method are used.

## 2 | NUMERICAL MODEL FOR THE VISCOELASTIC BEHAVIOR

### 2.1 | Bi-parabolic model

For this study, a shape memory polymer blend of (40% poly(caprolactone) PCL/60% Styrene–Butadiene–Styrene, SBS) is used. To predict the viscoelastic behavior of the

(PCL/SBS) mixture, a bi-parabolic model is used. Figure 1 shows rheological representation of this model.

As reported in study of Toussaint,<sup>1</sup>  $E_1$  presents the difference between the relaxation modulus  $E_0$ , which characterizes low frequencies and high temperatures (LF/HT), and the instantaneous modulus  $E_\infty$ , which characterizes high frequencies and low temperatures (HF/LT):

$$E_1 = E_\infty - E_0. \quad (1)$$

As presented in Figure 1, the bi-parabolic model present two branches. The stress of the polymer will then be presented as an addition of two values:

$$\sigma = \sigma_I + \sigma_{II}. \quad (2)$$

The first branch represents a pure elastic behavior:

$$\sigma_I = \varepsilon E_0. \quad (3)$$

The second one represents a viscoelastic behavior:

$$\sigma_{II} = \varepsilon_{21} E_1 = \frac{\eta_k}{\tau_k} (i\omega\tau_k)^k \varepsilon_{22} = \frac{\eta_{k'}}{\tau_{k'}} (i\omega\tau_{k'})^{k'} \varepsilon_{23}. \quad (4)$$

Two viscosity values are used in the bi-parabolic model.  $\eta_k$ : viscosity at high temperature.  $\eta_{k'}$ : viscosity at low temperature.  $k$ : parameter associated to relaxation modulus (LF/HT).  $k'$ : parameter associated to instantaneous modulus (HF/LT),  $0 < k' < k < .$

According to Reference [14], the relaxation times are calculated as follow:

$$\tau_k = \frac{\eta_k}{E_1} \text{ \& } \tau_{k'} = \frac{\eta_{k'}}{E_1}. \quad (5)$$

According to the principal of time superposition,<sup>1</sup> the relaxation times are proportional:

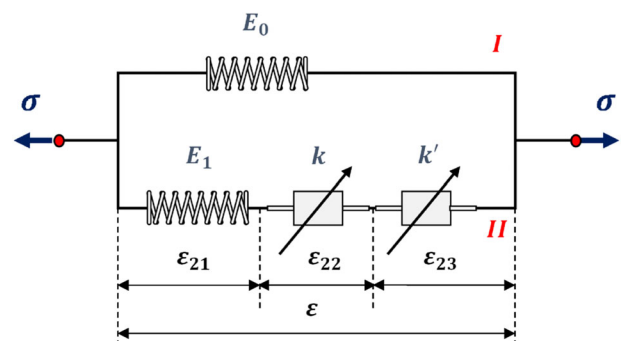


FIGURE 1 Representation of bi-parabolic model [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

$$\tau_{k'} = \delta \tau_k. \quad (6)$$

Using Equation 4 and Equation 5, the total strain in the viscoelastic branch is calculated by:

$$\varepsilon = \varepsilon_{21} + \varepsilon_{22} + \varepsilon_{23} = \frac{\sigma_{II}}{E_1} \left( 1 + (i\omega\tau_k)^{-k} + Q(i\omega\tau_k)^{-k'} \right). \quad (7)$$

With

$$Q = \delta^{-k'}. \quad (8)$$

Using Equations 2, 3, 7 and 8 the total stress is calculated by the following equation:

$$\sigma = \sigma_I + \sigma_{II} = \varepsilon \left( E_0 + \frac{E_1}{1 + (i\omega\tau_k)^{-k} + Q(i\omega\tau_k)^{-k'}} \right). \quad (9)$$

Then the bi-parabolic model is presented as follow:

$$E^* = \frac{\sigma}{\varepsilon} = E_0 + \frac{E_\infty - E_0}{1 + (i\omega\tau_k)^{-k} + Q(i\omega\tau_k)^{-k'}} = E' + iE''. \quad (10)$$

The complex modulus  $E^*$  consists of a real part ( $E'$ ) and an imaginary part ( $E''$ ).

$E'$  presents the storage modulus, characteristic of the potential energy (reversible energy):

$$E' = E_0 + (E_\infty - E_0) \frac{1 + \cos\left(\frac{K\pi}{2}\right)(\omega\tau)^{-K} + Q\cos\left(\frac{K'\pi}{2}\right)(\omega\tau)^{-K'}}{D}. \quad (11)$$

$E''$  is the loss modulus, characteristic of the dissipated energy:

$$E'' = (E_\infty - E_0) \frac{\sin\left(\frac{K\pi}{2}\right)(\omega\tau)^{-K} + Q\sin\left(\frac{K'\pi}{2}\right)(\omega\tau)^{-K'}}{D}, \quad (12)$$

where

$$D = \left[ 1 + \cos\left(\frac{K\pi}{2}\right)(\omega\tau)^{-K} + Q\cos\left(\frac{K'\pi}{2}\right)(\omega\tau)^{-K'} \right]^2 + \left[ \sin\left(\frac{K\pi}{2}\right)(\omega\tau)^{-K} + Q\sin\left(\frac{K'\pi}{2}\right)(\omega\tau)^{-K'} \right]^2, \quad (13)$$

$i$ : complex number ( $i^2 = -1$ );  $\omega$ : angular frequency =  $2\pi f$ ; and  $f$  is the frequency.

The correlation between the values of  $k$  and  $k'$  and the characteristics of the polymer could be found at the level of its macromolecular structure. In fact, Diaz<sup>15</sup> reported that  $k$  and  $k'$  correspond to the molecular movements of polymers. Other research<sup>1</sup> related  $k'$  to the entropic elasticity character of the polymer, which corresponds to the restoring forces applied by the segments of the macromolecular chains of the polymer. They related  $k$  to the property of viscous flow which is associated to the displacement of the center of gravity of kinetic elements.  $E_0$  characterizes the elasticity of the material and  $E_\infty$  its viscous behavior. The parameters of the bi-parabolic model are determined using the Cole-Cole method.

## 2.2 | Cole-Cole method

The Cole-Cole diagram of the bi-parabolic model is represented by Figure 2. In this method,  $E''$  (imaginary part of the complex modulus  $E^*$ ) is plotted versus  $E'$  (real part of  $E^*$ ).

From the Cole-Cole diagram, the model parameters are determined as follows.

1.  $E_0$  and  $E_\infty$  are determined graphically by linear extrapolation from the rubbery and the glassy state. Intercepts of the Cole-Cole curve with  $x$ -axis give  $E_0$  and  $E_\infty$ .
2.  $k$  and  $k'$  are proportional to  $\alpha$  and  $\beta$ .  $\alpha$  and  $\beta$  are the angles made by the plot at the low- and high-frequency intercepts, respectively.

$$k = \frac{\alpha}{\frac{\pi}{2}}, \quad (14)$$

$$k' = \frac{\beta}{\frac{\pi}{2}}. \quad (15)$$

3. The relaxation time  $\tau$  is determined from  $f_0$ , at which  $E''$  is maximum. It equal to:

$$\tau = \frac{1}{2\pi f_0}. \quad (16)$$

Cole-Cole curves can be experimentally determined from dynamical mechanical analysis (DMA) tests.

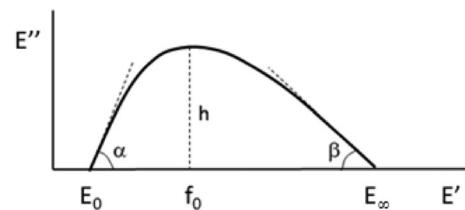


FIGURE 2 Cole-Cole diagram<sup>16</sup>

### 3 | RESULTS AND DISCUSSIONS

#### 3.1 | Materials and methods

In this work, a SMP of (60%) SBS and (40%) PCL is used. In previous study,<sup>17</sup> it is revealed via shape memory tests that this immiscible blend shows a 100% Shape memory effect. The SBS used in this study is provided by Sinopec Group. It is an amorphous elastomer whose main chain is composed of a flexible polybutadiene group in the middle and two rigid polystyrene groups on both sides. SBS has a glass transition temperature of  $-76^{\circ}\text{C}$ .<sup>18</sup> The PCL used in this work is supplied by Perstrop Limited UK. This polymer is semicrystalline biodegradable thermoplastic. It has a relatively low melting temperature of  $59.7^{\circ}\text{C}$ , a transition temperature of  $-40.1^{\circ}\text{C}$  and a crystallinity degree of 50.5%.<sup>18</sup> A mixture of 60% of SBS and 40% of PCL is primarily extruded through a twin-screw extruder machine (Brabender, Duidburg Germany). For all experimental tests, the extruded pellets are injected into an injection molding machine to get final samples with a gauge size of  $25 \times 4 \times 2 \text{ mm}^3$ . Table 1 illustrates the properties of the SMP blend (PCL/SBS).

In order to obtain the experimental values of the storage and the loss modulus, (PCL/SBS) samples are submitted to DMA tests using DMA Q800 TA Instruments type machine. These experiments are performed with a constant frequency (1 Hz) and a constant strain rate (0.1%); a temperature range between  $-120$  and  $100^{\circ}\text{C}$ , and a heating ramp of  $5^{\circ}\text{C}/\text{min}$ .

#### 3.2 | Experimental results

DMA tests are performed to obtain the experimental values of the storage and the loss modulus. The storage and the loss modulus are necessary to establish Cole-

Cole curve for the SMP. DMA results are shown in Figures 3 and Figure 4.

We can remark that  $E'$  and  $E''$  spectrums display four distinct regions corresponding to the physical state variation of the blend:

Zone 1:  $T < -100^{\circ}\text{C}$  In this region, the amorphous phase of SBS and PCL, is in the glassy state. The modulus represents very high value ( $> 3500 \text{ MPa}$ ) and the blend is very rigid.

Zone 2:  $-100^{\circ}\text{C} < T < -65^{\circ}\text{C}$  It corresponds to the glass transition of the SBS phase. At the end of this zone, SBS becomes rubbery with very low rigidity. However, PCL remains in glassy state.

Zone 3:  $-65^{\circ}\text{C} < T < 0^{\circ}\text{C}$  This region corresponds to the glass transition of the PCL phase.

Zone 4:  $0^{\circ}\text{C} < T < 50^{\circ}\text{C}$  In this zone, the amorphous phase of the two polymers (PCL and SBS) is in the rubbery state with very low rigidity. However, the crystalline phase of PCL remains in the crystalline state. It is this crystalline phase that will ensure the strength of the blend.

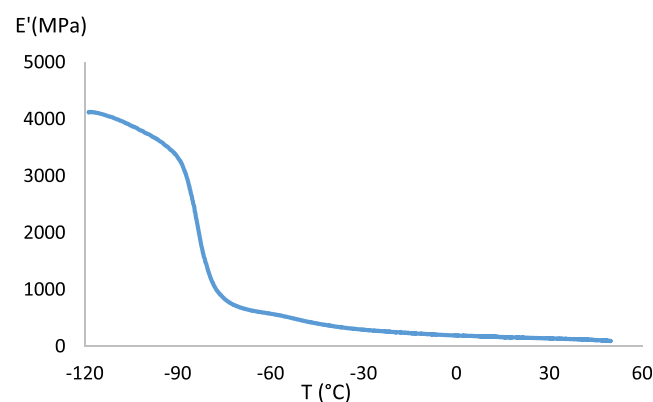


FIGURE 3  $E' = f(T)$  spectrum of (PCL/SBS) blend [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

Symbol	Name	Unit	Value
$M_{w1}$ (PCL)	Weight average molecular weight	(g/mol)	71,22
$M_{n1}$ (PCL)	Number average molecular weight	(g/mol)	42,14
$X_C$	Crystallinity degree	(%)	51,4
$T_{\alpha 1}$	Glass transition temperature	( $^{\circ}\text{C}$ )	$-80,7$
$T_{\alpha 2}$	Glass transition temperature	( $^{\circ}\text{C}$ )	$-39,7$
$T_C$	Crystallization temperature	( $^{\circ}\text{C}$ )	34,1
$T_m$	Melting temperature	( $^{\circ}\text{C}$ )	58,9
$\epsilon_r$	Deformation at rupture	(%)	$292 \pm 9$
$E$	Young modulus	MPa	$116 \pm 3,5$
$R_r$	Recovery rate	(%)	100

TABLE 1 Properties of the shape memory polymer mixture (PCL/SBS)<sup>17</sup>

Experimental curve of  $E'' = f(E')$  is then plotted and presented in Figure 5.

According to Figure 5, we distinguish the same four previous zones which are associated to the state variation of SBS (amorphous phase) and PCL (amorphous phase and crystalline phase):

Zone 1: Zone of high rigidity In this region, the elastic modulus  $E'$  of the blend represents an elevated value ( $> 3500$  MPa).

Zone 4: Zone of low rigidity In this region,  $E'$  represents low value.

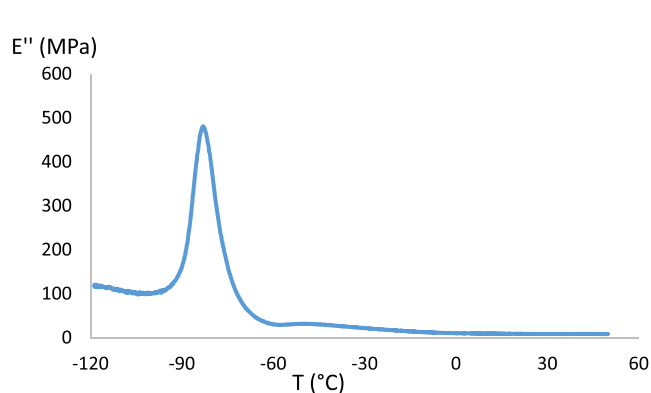
Zone 3: Zone of moderate variation of the elastic modulus  $E'$  and the viscous modulus  $E''$ . This region is associated to the glass transition of the PCL phase. Figure 6 represents a zoom of Cole–Cole diagram in zone 3. According to this Figure,  $E'$  increases from 200 to 500 MPa.

Zone 2: Zone of significant variation of  $E'$  and  $E''$ . A zoom in the Cole–Cole diagram in this region is illustrated in Figure 7. In this zone,  $E'$  rises from 600 to 3500 MPa and  $E''$  displays a maximum at 480 MPa.

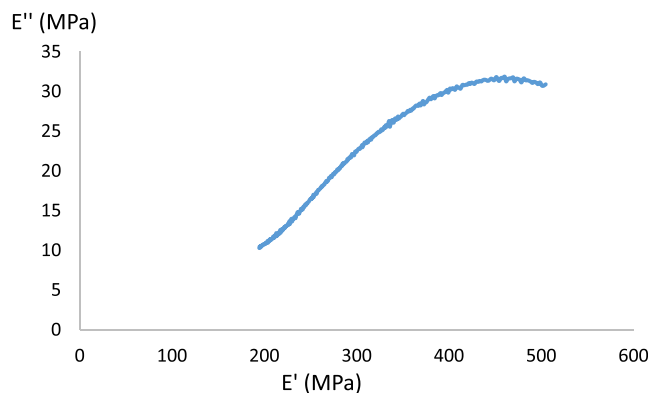
As the elastic modulus  $E'$  and the viscous modulus  $E''$  depict a significant variation in zone 2, the viscoelastic behavior of the SMP blend (PCL/SBS) will be modeled only in this zone. In order to investigate the evolution of the bi-parabolic model parameters, mixtures with different  $M_w$  of PCL are used. Table 2 summarizes the used materials in this study.

The mixture with  $M_w = 71,220$  g/mol is called reference material. This blend represents a total shape memory effect (TSME).<sup>19</sup> Ben Abdallah et al reported in previous studies<sup>5,17</sup> that the mechanical and physico-chemical properties change with the decrease in  $M_w$  of PCL. This alteration of the blend properties causes the diminution of its recovery rate  $R_r$ . For example, the (PCL/SBS) mixture with  $M_w = 71,220$  g/mol represents better SME ( $R_r = 100\%$ ). Then, by decreasing the  $M_w$  of PCL, the shape memory effect of the blend declines. This polymer initially with TSME, becomes a material with partial shape memory effect (PSME).<sup>17</sup>

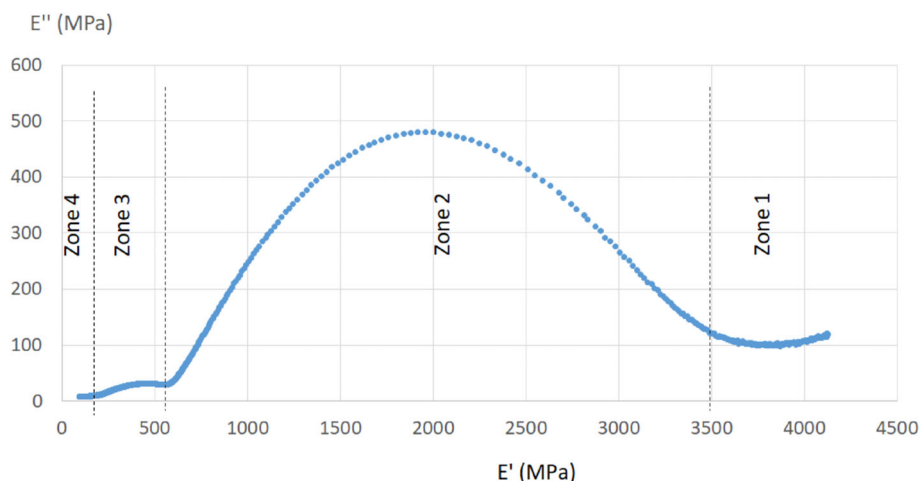
The bi-parabolic model is then used to predict the viscoelastic behavior of the SMP blends (PCL/SBS) with different  $M_w$  of PCL in zone 2.



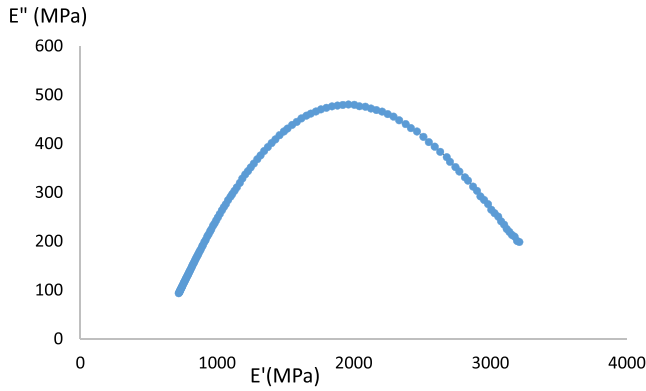
**FIGURE 4**  $E'' = f(T)$  spectrum of (PCL/SBS) blend [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



**FIGURE 6** Cole–Cole diagram of (PCL/SBS) blend in zone 3 [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



**FIGURE 5** Experimental Cole–Cole diagram of (PCL/SBS) blend [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



**FIGURE 7** Cole-Cole diagram of (PCL/SBS) blend in zone 2 [Color figure can be viewed at wileyonlinelibrary.com]

**TABLE 2** (PCL/SBS) blend with different PCL ( $M_w$ )

(40% PCL/60% SBS) blends	$M_w$ (PCL) (g/mol)
Blend 1 (reference)	$M_{w1} = 71,220$
Blend 2	$M_{w2} = 68,760$
Blend 3	$M_{w3} = 66,480$
Blend 4	$M_{w4} = 64,740$
Blend 5	$M_{w5} = 62,800$

### 3.3 | Modeling of viscoelastic behavior of reference shape memory polymer blend ( $M_{w1}$ ) in zone 2

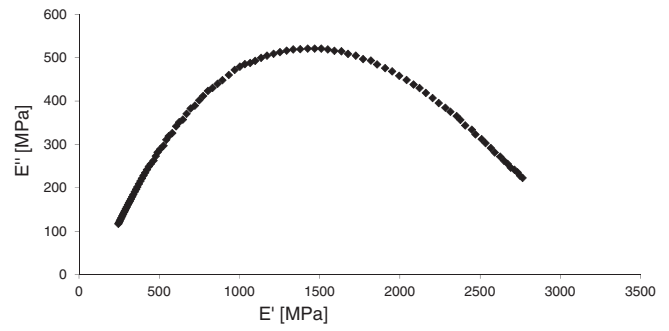
Figure 8 and Figure 9 show the Cole-Cole diagram of pure SBS and pure PCL, respectively.

The Cole-Cole diagram ( $E'' = f(E')$ ) presents a symmetrical curve for amorphous polymers (homogeneous morphology) and an asymmetric parabola for semi-crystalline polymers.<sup>16</sup> This outcome is in harmony with the form of curves as shown in Figure 8 for the SBS and in Figure 9 for the PCL.

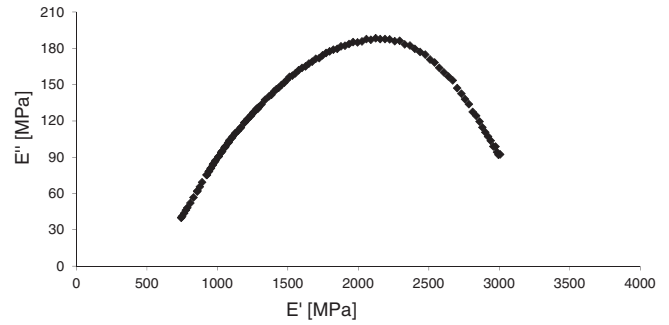
The experimental Cole-Cole diagram of the reference blend ( $M_{w1}$  (PCL) = 71,220 g/mol), is compared to the biparabolic model (Equation 10). The modeling is realized using MATLAB software. Parameters of biparabolic model are determined as follow:

$E_0$  and  $E_\infty$  are determined graphically from the intersection of experimental Cole-Cole curve with storage modulus axis.

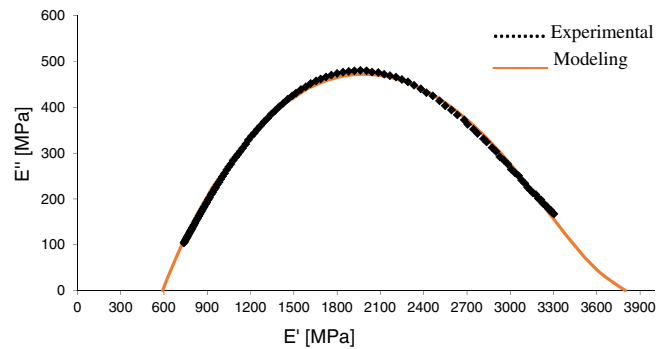
$k$ ,  $k'$  and  $\tau$  from Equations 14, 15 and 16 respectively. Experimental and modeling Cole-Cole curves, in zone 2, are depicted in Figure 10. The two diagrams form a slightly asymmetric parabola. Overall, results are in good accordance with minor differences. This outcome confirms the validity of the bi-parabolic model.



**FIGURE 8** Cole-Cole curve of pure Styrene-Butadiene-Styrene



**FIGURE 9** Cole-Cole curve of pure PCL (with  $M_{w1} = 71,220$  g/mol)



**FIGURE 10** Cole-Cole diagrams for reference (PCL/SBS) blend ( $M_{w1}$ ) [Color figure can be viewed at wileyonlinelibrary.com]

### 3.4 | Modeling of viscoelastic properties of shape memory polymer blend with different $M_w$ in zone 2

The biparabolic model is also used to predict the viscoelastic behavior of (PCL/SBS) blends with different  $M_w$  of PCL. Results are illustrated in Figures 11–14.

Results represented in Figures 11–14, indicate that the bi-parabolic model describes well the behavior of blends for different  $M_w$  of PCL. Relationship between the



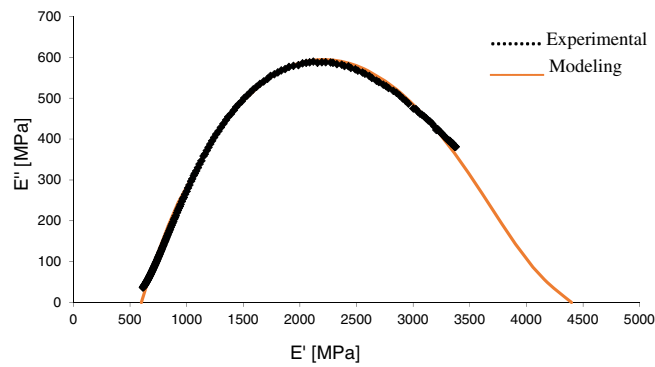
constants model ( $E_0$ ,  $E_\infty$ ,  $k'$ ,  $k$  and  $\tau_k$ ) and the molecular of the PCL is then established. Table 3 summarizes the values of the biparabolic model parameters for different  $M_w$  of PCL.

According to results in Table 3, different phenomena can be observed:

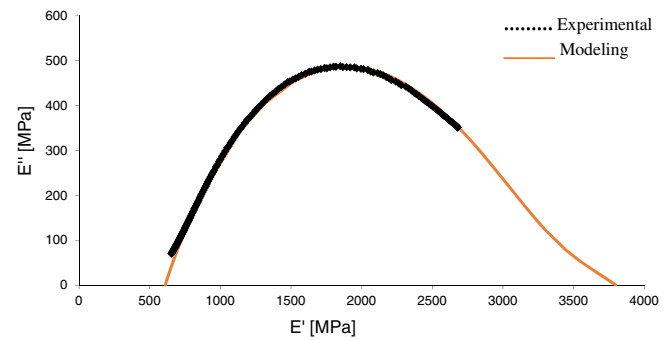
1.  $k$ ,  $k'$ ,  $Q$  and  $\tau$  remain almost constant for different  $M_w$  of PCL.
2. There is no obvious relation between  $E_\infty$  and the change in the  $M_w$ . In fact, no trend appears in  $E_\infty$  values due to their high scatter.  $E_\infty$  is determined by linear extrapolation from the Cole–Cole diagram, using the best compromise between the broadness of

the modulus range and the value of the correlation coefficient. Therefore, its values carry strong uncertainties.

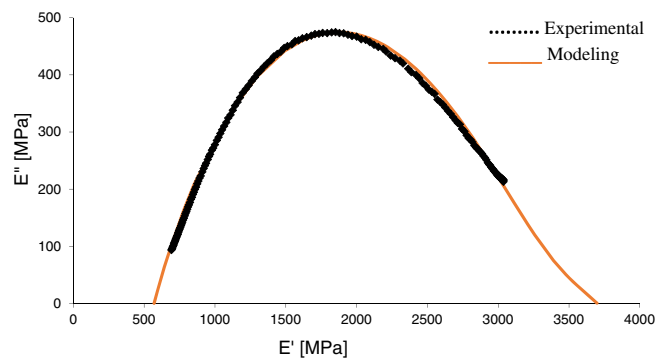
3.  $E_0$  characterizes the elasticity of the material.  $E_0$  increases with the drop of the  $M_w$  of PCL. In fact, with the diminution of  $M_w$  from 71,220 to 62,800 g/mol,  $E_0$  rises from 590 to 720 MPa. This augmentation in  $E_0$  indicates that the rigidity of the blend rises. This result is in harmony with the experimental results reported in our previous works.<sup>5,17</sup> In this research, we have shown that by decreasing  $M_w$  from 71,220 to 62,800 g/mol, the young modulus  $E$  increases from 116 to 187–219 MPa which evinces the growth of the rigidity of blend (SBS/PCL). The rise of the rigidity is essentially due to the increase of the crystallinity  $X_c$ .<sup>5,17</sup> In fact,



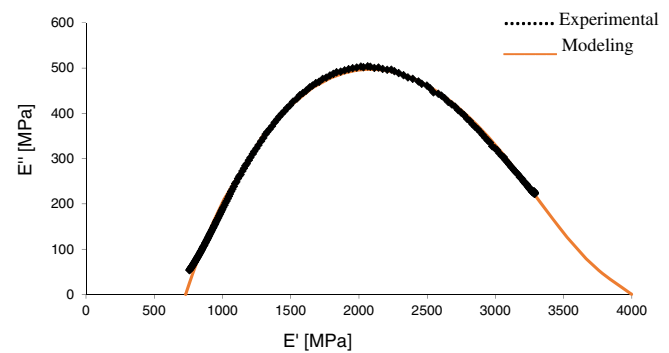
**FIGURE 11** Cole–Cole diagrams for (PCL/SBS) blend with  $M_{w2}$  [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



**FIGURE 13** Cole–Cole diagrams for (PCL/SBS) blend with  $M_{w4}$  [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



**FIGURE 12** Cole–Cole diagrams for (PCL/SBS) blend with  $M_{w3}$  [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



**FIGURE 14** Cole–Cole diagrams for (PCL/SBS) blend with  $M_{w5}$  [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

**TABLE 3** Evolution of biparabolic model parameters with  $M_w$  of PCL

$M_w$ (g/mol)	$E_0$ (MPa)	$E_\infty$ (MPa)	$k'$	$k$	$\tau_k$ (s)	$Q$
$M_{w1} = 71,220$	590	3850	0,14	0,46	1,44	0,40
$M_{w2} = 68,760$	600	4450	0,14	0,48	1,31	0,40
$M_{w3} = 66,480$	600	3750	0,14	0,49	1,50	0,45
$M_{w4} = 64,740$	610	3800	0,14	0,49	1,50	0,45
$M_{w5} = 62,800$	720	4000	0,14	0,48	1,34	0,39




$X_c$  of the blend goes up from 51.4% to 69.7% when  $M_w$  of PCL drops from 71,220 to 64,740 g/mol.<sup>17</sup>

## 4 | CONCLUSION

In this paper, a biparabolic model with six parameters ( $E_0$ ,  $E_\infty$ ,  $k$ ,  $k'$ ,  $Q$ ,  $\tau$ ) has been used to predict the viscoelastic behavior of the SMP blend (40% PCL/60% SBS). Experimental Cole–Cole diagram ( $E'' = f(E')$ ) was compared to the Cole–Cole curve obtained via biparabolic model. Experimental  $E'' = f(E')$  spectrum of the SMP mixture has been determined from DMA tests. This diagram presents four different zones associated to the physical state and the rigidity of both SBS and PCL phases. It has been revealed that the 2nd zone shows the highest variation of both elastic ( $E'$ ) and viscous ( $E''$ ) modulus. Thus, the modeling of the viscoelastic behavior of (PCL/SBS) blend, with different  $M_w$  of PCL, has been realized only in this zone. It has been demonstrated that the biparabolic model predicts well the viscoelastic behavior of the mixture for different  $M_w$ . The evolution of the model parameters with the  $M_w$  of PCL has been evaluated. Results have indicated that, when  $M_w$  of PCL decreases, the relaxation modulus  $E_0$  rises. This outcome evinces that the rigidity of the SMP blend is increasing with the drop of  $M_w$ . The change in the  $M_w$  influences the shape memory effect of the blend as reported in our previous study.<sup>17</sup> It becomes then possible to develop a new blend with specific SME and mechanical properties by changing the  $M_w$ .

## ORCID

Abir Ben Abdallah  <https://orcid.org/0000-0002-4164-369X>

Achraf Kallel  <https://orcid.org/0000-0001-6387-925X>

## REFERENCES

- [1] A. Toussaint, L. Dhont, *Prog. Org. Coat.* **1983**, *11*, 139.
- [2] S. Farzaneh, J. Fitoussi, A. Lucas, M. Bocquet, A. Tcharkhtchi, *J. Appl. Polym. Sci.* **2013**, *128*, 3240.

- [3] Q. Meng, J. Hu, *Compos. Part A* **2009**, *40*, 1661.
- [4] M. Behl, A. Lendlein, *Shape Memory Polym. Mater. Today* **2007**, *10*, 20.
- [5] A. Ben Abdallah, *Influence du vieillissement sur l'effet mémoire des polymères/Modélisation du mécanisme de mémoire de forme*, ENSAM de Paris, Paris, France **2020**.
- [6] A. Tcharkhtchi, S. A. Elhritsi, J. Fitoussi, K. Ebrahimi, M. Shirinbayan, S. Farzaneh, *Polymer* **2014**, *6*, 1144.
- [7] H. Rong, Z. Shoujing, L. Zishun, Y. N. Teng, *Int. J. Appl. Mech.* **2020**, *12*, 1.
- [8] E. Chabert, *INSA de Lyon* **2002**, *1*, 1.
- [9] Y. X. Li, S. S. Guo, Y. H. He, Z. S. Liu, *Mater. Sci. Eng.* **2015**, *04*, 1550001.
- [10] Y. C. Chena, D. C. Lagoudas, *J. Mech. Phys. Solids* **2008**, *56*, 1752.
- [11] S. A. Elhritsi, J. Fitoussi, B. J. Rashm, K. Prashantha, S. Farzaneh, M. F. Lacrampe, *Polym. Compos.* **2015**, *1145–51*, 1145.
- [12] A. Tcharkhtchi, A. S. Lucas, J. P. Trotignon, J. Verdu, *Polymer* **1998**, *39*, 1233.
- [13] J. C. Carret, *ENTP de Lyon* **2018**, *2*, 1.
- [14] R. K. Gupta, *Polymer and Composite Rheology*, Taylor & Francis Inc., Bosa Roca, USA, **2001**, p. 408.
- [15] R. Diaz-Calleja, M. J. Sanchis, J. Mostos, *Polymer* **1996**, *37*, 4003.
- [16] N. Zeggai, *Etude thermophysique des copolymères réticulés à base de l'Isobornylacrylate: Approche expérimentale et modélisation*, Université Abou-Bekr Belkaid, Tlemcen, Algérie **2018**.
- [17] A. Ben Abdallah, F. Gamaoun, A. Kallel, A. Tcharkhtchi, *J. Appl. Polym. Sci.* **2020**, *138*, e49761.
- [18] A. B. Abdallah, A. Kallel, F. Gamaoun, A. Tcharkhtchi, *J. Polym. Environ.* **2019**, *27*, 2341.
- [19] A. Kallel, A. Ben Abdallah, F. Gamaoun, S. Farzaneh, H. BenDaly, J. Fitoussi, A. Tcharkhtchi, *J. Polym. Res.* **2021**, *28*, 308.

**How to cite this article:** A. Ben Abdallah, A. Kallel, T. Hassine, F. Gamaoun, A. Tcharkhtchi, *J. Appl. Polym. Sci.* **2022**, *139*(13), e51859. <https://doi.org/10.1002/app.51859>