NUMERICAL SIMULATION OF THERMAL EXPANSION PROCESS FOR THERMOSET COMPOSITE LAMINATES USING SILICONE RUBBER TOOLS

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Abstract:

In thermal expansion process tooling design is very important for the time of applying pressure and the generation of pressure by silicon rubbers, and determines the quality of cured composites. In this paper, a finite element model was developed to simulate the thermal expansion process for thermoset composite materials using silicone rubber tools. Simulations and experiments were carried out on the thermal expansion course of a kind of silicone rubber and the model predictions agree well with the experimental measurements. Furthermore, simulations were performed on the cure course of composite laminates using silicone rubber thermal expansion process and the compaction pressure exerted by the rubber with different process gaps was predicted. The results indicate that there is a close relationship between the process gaps and the thermal expansion pressure, as well as the time of applying the consolidation pressure on the laminate during the process.

1 Introduction

Thermal expansion process is an effective method to fabricate thermoset composite parts having closed structures using convex dies, in which materials having high thermal expansion coefficient (CTE) and certain modulus are used, such as silicone rubbers. During the process, a composite laminate is placed between a silicone rubber and a closed metal tool. Due to the difference between the CTE of rubber mold and metal tool, the thermal expansion of silicone rubber during the curing process forces the laminate to the metal tool, and provides a pressure needed for laminate compaction [1]. After the composite material is cured and cooled to room temperature, the silicone rubber shrinks and is easy to remove from the composite part.

In the past three decades, many researchers investigated theoretically and experimentally the methods to control the thermal expansion process [1-6]. Cremens and Reinert [1] developed a quantitative analysis method to express the consolidation pressure as a function of temperature and parameters of rubber molds, metal tools and laminates. An experimental study of the influence of changing the volume of rubber mold on the thermally-generated pressure was performed by Kromrey [2], and the results showed that a free expansion for the rubber mold could reduce the pressure. Kemp [3], Cull et al. [4] and Penado et al. [5] presented different techniques to control the pressure, respectively. Kim [6] applied KWW (Kohlrausch-Williame-Watts) non-linear viscoelastic model to the characteristics of flexible mold and composite laminate, and the consolidation pressure was predicted by the equation. These studies above demonstrated that the tooling design of thermal expansion process is very important for the generation of pressure and the pressure transfer during the process, and determines the quality of final parts. However, to date the process is generally based on the trial and error method or accumulation of experiences assisted with the theoretical formula, which are time-consuming and costly. With the development of computer technology, numerical simulation of composite processing provides a new avenue to optimize process parameters and tooling, and has been successfully adopted in autoclave process and RTM.

In this paper, a finite element model was developed to simulate the thermal expansion process for composite laminates using silicone rubber molds. Meanwhile, experiments were performed to investigate the changes with temperature of material properties for a kind of silicone rubber. Based on these material parameters, simulations and experiments on the thermal expansion course of a silicone rubber mold were compared and the sensitivity of parameters on the expansion behavior of the rubber was analyzed. Furthermore, simulations were performed for carbon fiber/bismaleimide resin laminates using silicone rubber thermal expansion process and the effect of different process gaps on the consolidation pressure generated by the silicon rubber was investigated. These results are valuable for optimizing the thermal expansion process and can guide the design of silicone rubber tools.

2 Numerical models

Based on the thermo-elastic theory [7], a two-dimensional plain strain finite element model was developed to simulate the thermal expansion process for thermoset composite materials using silicone rubber tools. The process models include two parts as follows, i.e. temperature field computation and deformation field computation.

2.1 Temperature field computation

The Fourier equation was adopted as the basic equation for transient heat conduction:

$$\frac{\partial(\rho C_P T)}{\partial t}=\frac{\partial}{\partial x}\left(k_x\frac{\partial T}{\partial x}\right)+\frac{\partial}{\partial z}\left(k_z\frac{\partial T}{\partial z}\right)+\dot{Q} \qquad (1)$$

where T is the temperature; $\dot{Q}$ is the internal heat source density; ρ and C_p are the density and specific heat of materials; k_x and k_z are thermal conductivities along the x and z directions, respectively.

For silicon rubber materials there was no internal heat source ($\dot{Q}$ = 0). For composite laminates, the effect of resin flow on the temperature was negligible and the resin and fiber were at the same temperature at any specific time. With these assumptions, Eq. (1) can be rewritten as follows using the Kamal cure kinetic model [8] to represent the curing process of a laminate:

$$\begin{gathered}\frac{\partial\left(\rho_c C_{cp}T\right)}{\partial t}=\frac{\partial}{\partial x}\left(k_{cx}\frac{\partial T}{\partial x}\right)+\frac{\partial}{\partial z}\left(k_{cz}\frac{\partial T}{\partial z}\right)+\rho_c H_u\frac{d\alpha}{dt}\\ \frac{\partial\alpha}{\partial t}=Ae^{\frac{-E}{RT}}\alpha^m\left(1-\alpha\right)^n\end{gathered} \qquad (2)$$

where the subscript c represents composite materials; H_u is the ultimate heat of reaction during cure and α is the degree of cure; A is pre-exponential factor; E is activation energy; R is universal gas constant; m and n are cure kinetic parameters.

2.2 Deformation field computation

Regardless of body force, the equilibrium equation can be written as

$$\begin{gathered}\frac{\partial\sigma_{xx}}{\partial x}+\frac{\partial\tau_{zx}}{\partial z}=0\\ \frac{\partial\tau_{xz}}{\partial x}+\frac{\partial\sigma_{zz}}{\partial z}=0\end{gathered} \qquad (3)$$

where σ_{xx}, σ_{zz} and τ_{xz} are the stress carried by materials.

The linear elastic constitutive equation was used to describe the mechanical behavior of silicon rubber molds and composite laminates. As the deformation during the process is small, the geometric equation of materials is:

$$\varepsilon_{xx}=\frac{\partial u}{\partial x};\qquad \varepsilon_{zz}=\frac{\partial w}{\partial z};\qquad \gamma_{xz}=\frac{\partial u}{\partial z}+\frac{\partial w}{\partial x} \qquad (4)$$

where ε_{xx}, ε_{zz} and γ_{xz} are the strain carried by materials, u and w are the two displacement components in the x and z directions at any point of materials.

The silicon rubber was assumed to be isotropic and the thermo-elastic strain-stress relationship is:

$$\begin{aligned}\begin{Bmatrix}\sigma_{rxx}\\ \sigma_{rzz}\\ \tau_{rxz}\end{Bmatrix}&=\frac{E_r}{(1+\nu_r)(1-2\nu_r)}\begin{bmatrix}1-\nu_r & \nu_r & 0\\ \nu_r & 1-\nu_r & 0\\ 0 & 0 & (1-2\nu_r)/2\end{bmatrix}\begin{Bmatrix}\varepsilon_{rxx}\\ \varepsilon_{rzz}\\ \gamma_{rxz}\end{Bmatrix}\\ &-\frac{E_r}{(1+\nu_r)(1-2\nu_r)}\times\alpha_r\times\Delta T\times(1+\nu_r)\times\begin{Bmatrix}1\\ 1\\ 0\end{Bmatrix}\end{aligned} \qquad (5)$$

where the subscript r represents silicon rubber materials; α_r is the CTE of silicon rubber; E_r and ν_r are the elastic modulus and Poisson ratio of rubber mold; ΔT is the temperature rise.

For anisotropic composite materials, the constitutive equation can be written as

$$\begin{Bmatrix}\sigma_{cxx}\\ \sigma_{czz}\\ \tau_{cxz}\end{Bmatrix}=\begin{bmatrix}E_{xx} & \nu & 0\\ \nu & E_{zz} & 0\\ 0 & 0 & G_{xz}\end{bmatrix}\begin{Bmatrix}\varepsilon_{cxx}\\ \varepsilon_{czz}\\ \gamma_{cxz}\end{Bmatrix}-\begin{Bmatrix}E_{xx}\\ E_{zz}\\ 0\end{Bmatrix}\times\alpha_c\times\Delta T\times\begin{Bmatrix}1\\ 1\\ 0\end{Bmatrix} \qquad (6)$$

where α_c is the CTE of composite material; E_{xx} and E_{zz} are the modulus along the x and z directions, respectively. G_{xz} is the shear modulus.

3. Experiment

3.1 Materials

The rubber mold was made of Aircast 3600 system® (Airtech Co.). The first step of casting a silicone rubber tool was to mix the liquid rubber (Part A) and curing agent (Part B) in a 10:1 ratio, and then place the material in a vacuum for 10 minutes to remove trapped air. After the deaeration, the rubber was carefully poured into the mold cavity and then cured at 60 ℃for 2 hours.

3.2 Measurement of properties of silicon rubber

To simulate a thermal expansion process and predict the compaction pressure generated by a rubber mold, thermo-elastic characterization of a silicon rubber was performed.

Temperature dependent measurements of the thermal conductivity k_r and specific heat C_{rp} of silicon rubber were performed by the LFA 427 NanoFlash®, and the bulk density ρ_r of rubber mold was measured using the method of draining water. A compressive test was performed on a cylindrical specimen (30 mm in height and 25 mm in diameter) using a universal testing machine (SANA Ltd. Co.), to measure the elastic modulus of silicon rubber. These results are listed in Table 1, in which T is the temperature.

Table 1 Material properties of the silicon rubber

Properties	Value （T(℃))
Bulk density (10^3kg/m^3)	ρ_r =1.26/(1+5.57×10^{-4}(T-27))
Specific heat (10^3J/(kg.K))	C_{rp}=3.07-0.0142T
Thermal conductivity (W/(m.k))	k_r=-6.52×$10^{-7}T^3$＋2.61×$10^{-4}T^2$-0.03T+1.85
Elastic modulus (MPa)	E_r=941.5-2.351T

A free thermal expansion test was carried out on the Aircast 3600, to measure the CTE of silicon rubber. The temperature was increased from room temperature (about 20 ℃) to 180 ℃ at a heating rate of 2 ℃/min, and was maintained at 30 ℃ increments during the cycle for 30 minutes to obtain a even temperature distribution in the sample. The unheated rubber cylinder had a height of 30 mm and a diameter of 18 mm. The results are in good linearity for two samples and the average value of linear CTE is 2.5×10^{-4} /℃. The bulk modulus K_r of silicon rubber was also measured by experiments and the Poisson ratio v_r of rubber mold related to E_r and K_r was calculated by the linear elastic equation.

4. Results and discussion

4.1 Simulations and experimental verification

To verify the validation of the numerical models for thermal expansion process, simulations and experiments were performed for the Aircast 3600 silicon rubber. The specimen with a height of 30mm and a diameter of 25 mm was heated in a cylindrical heating mold. The temperature was increased from room temperature (about 20 ℃) to 180 ℃ at a heating rate of 2 ℃/min, and was maintained at 60 ℃, 90 ℃, 120 ℃, 150 ℃ and 180 ℃ for 15 minutes, respectively. The thermal expansion was then measured by the dial indicator. A schematic of the testing device is shown in Fig. 1 (a).

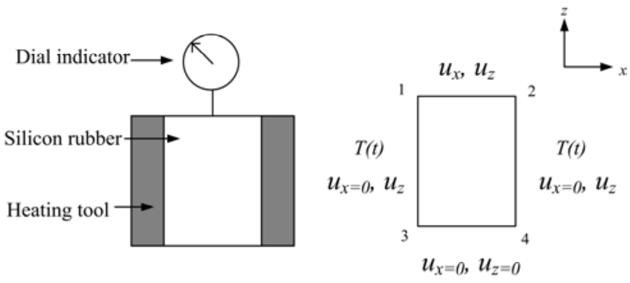


Fig. 1 Schematic of testing device and geometry for the simulation

The simulation was performed correspondingly and Fig. 1 (b) presents the two-dimensional geometry. Initially, the temperature was assumed to be 20℃ in the rubber mold. The displacements for the curve line 3-4 were fixed in the direction x and z, respectively (i.e. u_i=0, i=x, z). The displacements for the curve line 1-3 and 2-4 were only fixed in the direction x. Displacement boundary condition for the line 1-2 was free. The thermal load was applied on the line 1-3 and 2-4. 4-node elements were used for the model. The material properties of silicon rubber used in the simulation can be found in part 3.

Comparisons of the thermal expansion between the results from model predictions and experimental measurements are listed in Fig. 2. The predicted data are in good agreement with the experimental results, which proves the validity of established models.

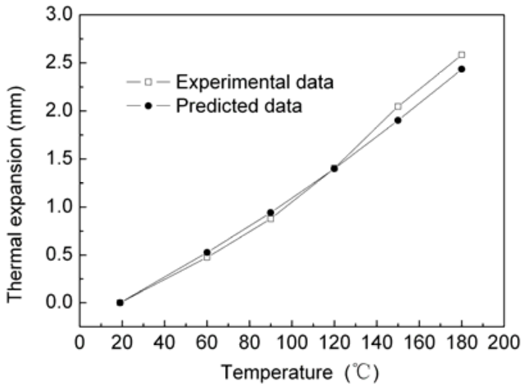


Fig. 2 Comparisons of the thermal expansion between experimental and predicted data

4.2 Parametric effects on the thermal expansion process

In thermal expansion process, the free expansion of a rubber mold is important to identify the pressure applying moment. Therefore, in this section a parametric study was performed to examine the sensitivity of material properties on the expansion behavior of the silicon rubber.

The thermal conductivity, specific heat and bulk density of materials all affect the ability of heat transfer in silicon rubbers. So the thermal diffusivity α_r is introduced to represent the thermal conduction properties of materials, including coupling effects of the three parameters above (Eq. (7)). Table 2 gives the properties of the rubber mold used in the simulations for the parametric study. The values were constant and chosen based on the experimental results. Boundary conditions were the same as the ones in section 4.1.

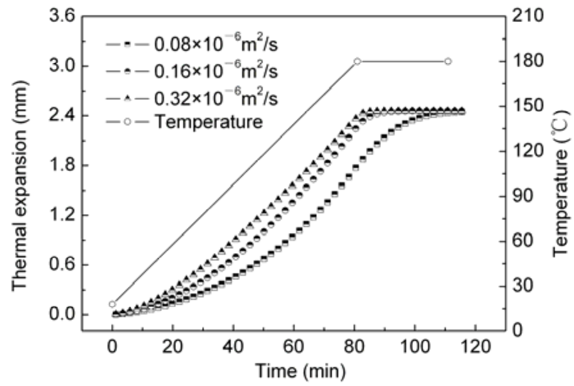
$$\alpha_r = \frac{k_r}{\rho_r \cdot C_{rp}} \tag{7}$$

Fig. 3 shows the temperature cycle for the simulations and the effect of parameters on the thermal expansion process of the silicon rubber. From the first graph (Fig. 3 (a)) it can be seen that altering the thermal diffusivity of silicon rubber has no influence on the final thermal expansion of the rubber mold, but affects the thermal expansion during the temperature cycle. When the thermal diffusivity of silicon rubber is 0.08×10^{-6} m^2/s, the increase of thermal expansion is lagged behind the temperature rise, while when the thermal diffusivity goes higher (like 0.32×10^{-6} m^2/s) the thermal expansion reaches the final as soon as the temperature gets to 180 ℃. This is because that the thermal diffusivity represents the heat transfer ability of silicon rubbers. The faster the heat transfers, the sooner the temperature distribution of the rubber mold reaches the balance.

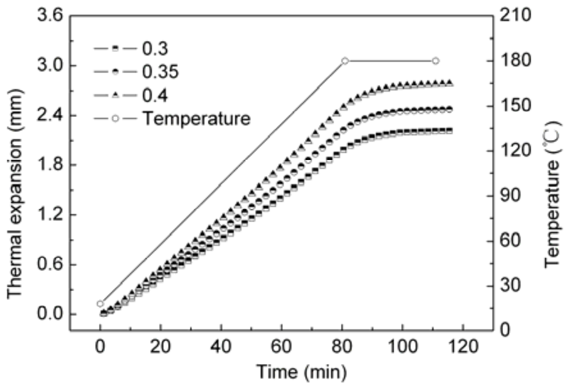
On the other hand, from Fig. 3 (b) and Fig. 3 (c) the results show that increasing 13% of the Possion radio of silicon rubber results in 11% growth of thermal expansion for the rubber mold, while increasing 13% of the CTE of silicon rubber causes 14% increase of thermal expansion. This can be concluded that the Possion radio and CTE of silicon rubber have a great influence on thermal expansion of the rubber mold not only during the process but also at the finish of the cycle. The effect of the elastic modulus on thermal expansion of the rubber mold was also carried out using different elastic modulus, such as 500MPa and 800MPa. However, the results have no obvious difference.

Table 2 Properties of rubber mold used in the simulations

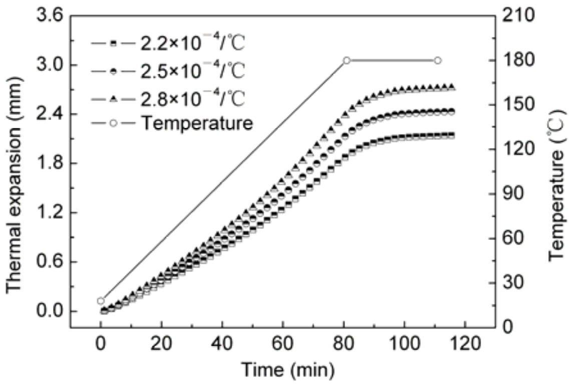
Simulations	α_r (10^{-6}m^2/s)	v_r	CTE (10^{-4}/℃)
1	0.08	0.3	2.5
2	0.16	0.3	2.5
3	0.32	0.3	2.5
4	0.16	0.3	2.5
5	0.16	0.35	2.5
6	0.16	0.4	2.5
7	0.16	0.3	2.2
8	0.16	0.3	2.5
9	0.16	0.3	2.8



(a) Effect of the thermal conductivity on thermal expansion of the silicon rubber



(b) Effect of the Poisson ratio on thermal expansion of the silicon rubber



(c) Effect of the CET on thermal expansion of the silicon rubber

Fig. 3 Results of the parametric study

This outcome indicates that the changes with temperature of parameters for the rubber mold can have great impacts on the free expansion course of materials, which are important for determining the pressure applying moment, therefore demonstrates the necessity of characterize accurately the silicon rubber for the modeling of thermal expansion process.

4.3 Simulations for manufacturing composite laminates using rubber molds

The consolidation pressure generated by a silicon rubber develops as the rubber mold finishes the free expansion and contacts the composite laminate. To analyze this thermal expansion process and predict the pressure acting on the laminate, simulations were performed on the cure course of carbon fiber/bismaleimide resin laminates using silicone rubber thermal expansion process. The effects of different process gaps on the compaction pressure exerted by the rubber were also investigated.

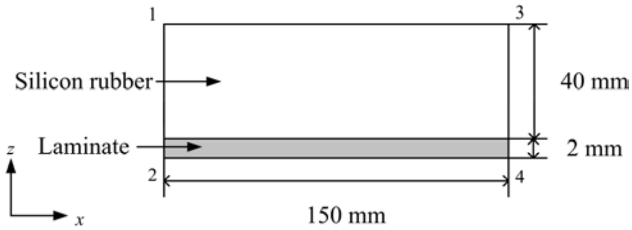


Fig. 4 Schematic of geometry for the simulation and dimensions of materials

Fig. 4 shows a schematic of the two-dimensional geometry for the simulation and the dimensions of the materials. Process gaps are the free thermal expansion along the direction z in the temperature cycle. During the process, it was assumed that there was no slippage between the laminate and the rubber mold. The thermal contact resistance between the composite part and the silicon rubber was ignored. Before the rubber mold filled the process gap the stress in the silicon rubber was supposed to be zero. Initially, the temperature was assumed to be 20 ℃ in the whole domain. The displacements for the curve line 2-4 were fixed in the direction x and z, respectively. The displacements for the curve line 1-2 and 3-4 were only fixed in the direction x. Displacement boundary condition for the line 1-3 was free when the rubber mold was in free expansion condition. After the expansion of the rubber filled the process gap, the displacement boundary condition for the line 1-3 was fixed in the direction x and z. The thermal load was applied on the line 1-3 and 2-4. 4-node elements were used for the whole model.

Material properties of the silicon rubber and the composite laminate used in simulations can be found in part 3 and ref. 9. E_{xx}, E_{zz} and G_{xz} for the composite part were set as 1.0×10^5, 10.0 and 50.0 MPa, respectively [10]. The temperature was increased from room temperature to 160 ℃ at a heating rate of 2 ℃/min, and was maintained at 160 ℃ for 2 hours.

Results of the consolidation pressure generated by the rubber mold with different process gaps are shown in Fig. 5. The data indicate that there is a close relationship between the process gaps and the thermal expansion pressure, as well as the time of applying the consolidation pressure on the laminate during the process. When the process gap is 1.8 mm, the time of applying the pressure is the earliest and the pressure is more than 2.0 MPa, which may cause excessive resin flow and even the destruction of the metal tool. When the process gap is 2.6 mm, the time of applying the pressure is too late to eliminate the void inside the laminate and the pressure is far from the desire consolidation pressure for the composite part. These results are valuable for optimizing the thermal expansion process and can guide the design of silicone rubber tools.

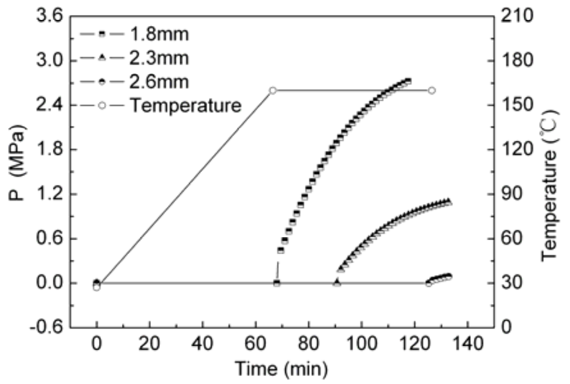


Fig. 5 Effect of different process gaps on the pressure exerted by the rubber

5. Conclusions

A two-dimensional finite element model was developed to simulate the thermal expansion process for thermoset composite materials using silicone rubber tools. It is found that the predicted thermal expansion of silicon rubber is consistent with the experimental measurements, which validates the numerical models for thermal expansion molding. The effects of parameters on the expansion behavior of the rubber were analyzed based on this model. The results show that the heat transfer abilities of rubber molds do not affect the final thermal expansion, whereas the Possion radio and CTE of silicon rubbers influence both of the free expansion during the cycle and the final thermal expansion. This outcome demonstrates the necessity of characterize accurately rubber molds for the modeling of thermal expansion process.

Furthermore, simulations were performed on the cure course of composite laminates using silicone rubber thermal expansion process and the effects of different process gaps on the compaction pressure exerted by the rubber were investigated. It is found that the process gaps have great impacts on the thermal expansion pressure and also the time of applying the consolidation pressure on the laminate during the cure, which emphasizes the importance of designing proper process gaps in thermal expansion process.

References

[1] W.S. Cremens and H.S. Reinert. "A general look at thermal expansion molding". *Society for the Advancement of Material and Process Engineering*, L.A., 21, pp 635-649, 1976.

[2] R.V. Kromrey. "New capabilities with therm-X process". *Society of Manufacturing Engineers, tooling for composite '88 Conference*, pp 10-11, 1988.

[3] D.N. Kemp. "Trapped rubber molding". Society of *Manufacturing Engineers, Fabricating Composites '88 Conference*, Philadelphia, pp 7, 1988.

[4] R.A. Cull, L. Jacobson and D.F. McMahon. "Out of autoclave processing of advanced composites utilizing silicone elastomers". *36th International SAMPE symposium*, San Diego, pp 944-958, 1991.

[5] F. Ernesto, P.J. Waydo and T.R. Kornfeld. "Manufacturing of Composite Tubes Using a Simulated Autoclave Technique". *33rd International SAMPE Technical Conference*, Covina, Calif., pp 728-738, 2001.

[6] D.O. Kim, S.W. Keum, J.H. Lee and J.D. Nam. "Thermal-expandable elastomer molding process for thermaset composite materials". *Composites Part A*, 37, pp 2121–2127, 2006.

[7] B.A. Boley and J.H. Weiner. "*Theory of thermal stresses. Summary of the formulation of thermoelastic problem*". John Wiley & Sons, New York, pp 243-271, 1960.

[8] M.R. Kamal and S. Sourour. "Kinetic and thermal characterization of thermoset cure". *Polymer Engineering and Science*, 13(1), pp 59-64, 1973.

[9] Z.S. Guo, S.Y. Du and B.M. Zhang. "Temperature field of thick thermaset composite laminates during cure process". *Composites science and technology*, 65, pp 517-523, 2005.

[10] J. Sun, Y.X. Li, M. Li, Y.Z. Gu and Z.G. Zhang. "Numerical study on effects of interaction between rubber mould and lay-up on consolidation of L-shaped laminates in autoclave process". *Polymers & Polymer Composites*, 19, pp 271-277, 2011.