Chapter 9
Effect of Processing Conditions on the Mechanical and Morphological Properties of Composites Reinforced by Natural Fibres

Abouelkacem Qaiss, Rachid Bouhfid, and Hamid Essabir

Abstract In the last years, the exploitation of different natural fibres in the plastic industry has become increasingly essential for the introduction of new composites and products. The properties of polymers reinforced with natural fibre composites are generally governed by the fibre and matrix properties, the compatibility between the compounds, the process of treatment of fibres, and the manufacturing process of composites. Enhancements and innovations in manufacturing technology and assembly of fibre-reinforced polymer composite materials and structures are required to achieve the objectives of cost and performance to allow wider adoption in many sectors. In this chapter, natural fibres from doum, coir, and bagasse were added to polypropylene by twin-screw extrusion and molded by injection machine, to study the influence of manufacturing process especially during the extrusion and injection processes. The influence of extruder screw configuration (corotating and counterrotating), screw speed (60, 80, and 120 rpm), and temperature (190, 200, and 210 °C) was studied on the pressure of matter and torque, and then an optimized configuration was selected to evaluate the mechanical properties of injected composite. On the other hand, the preparation of the injected composite samples was performed by adopting different placing directions according to flow and tensile directions: parallel and transversal placements and the effect of fibre orientation in the mechanical properties of manufactured composites were evaluated. Advanced knowledge of the relationship between structure, composition, and characteristics of material composite based on natural fibres made possible the development of high-performance materials with excellent mechanical properties. We have demonstrated some aspects concerning the influence of the manufacturing process of the fibres in the thermoplastic matrix.

Keywords Doum fibre • Coir fibre • Bagasse fibre • Composites • Twin-screw extrusion • Processing conditions

A. Qaiss (✉) • R. Bouhfid • H. Essabir
Moroccan Foundation for Advanced Science, Innovation and Research, Rabat, Morocco
e-mail: a.qaiss@mascir.com

M.S. Salit et al. (eds.), *Manufacturing of Natural Fibre Reinforced Polymer Composites*, DOI 10.1007/978-3-319-07944-8_9

9.1 Introduction

For several years, the appearance of some concepts such as sustainable development, industrial ecology, and green chemistry has pushed manufacturers to develop new materials from renewable resource (Unterweger et al. 2014; Zakikhani et al. 2014; Pan and Zhong 2014). Wherever advancing technology has created a need for combinations of properties no single material can provide, composites are becoming the material of choice (Pan and Zhong 2014; Rojo et al. 2015). By dispersing fibres or particles of one substance in a matrix, or binder, of another, the designer of a composite can reach properties neither material shows on its own. The future polymeric products must be processed and developed taking into account the environmental impact from the raw materials used in the synthesis, until the end of product life (Shouha et al. 2014; Merkel et al. 2014; Węcławski et al. 2014). Developments in the production of composite concern the improvement of the evaluation of structural defects to ensure durability, reliability, cost reduction, and increased production rates. For the strengthening of polymers, natural filler has significant specific mechanical properties and many advantages. Several worldwide laboratories have begun work on the development of new composite materials based on thermoplastic matrices reinforced with natural fillers (bio-fillers) (Merkel et al. 2014; Węcławski et al. 2014). These bio-fillers are renewable resources, biodegradable, less abrasive, neutral as CO_2 emissions into the atmosphere, and requiring little energy to be produced. These fibres can replace glass fibres in many areas or allow reaching new markets (Arrakhiz et al. 2012a, b, 2013a).

Composite materials are composed of two materials of different properties but complementary. The reinforcement usually allows support of the load of the structure, reducing thermal stress, and ensures rigidity and macroscopic resistance. The matrix serves to link the reinforcing providing protection against external agents and transfer loads to the reinforcement via the connection between the filler and the matrix (Arrakhiz et al. 2012a, b, 2013a). The fibres used for the reinforcement of polymer matrix are generally classified into two groups: continuous fibres and discontinuous fibres. Continuous reinforced composites are reserved for applications with high performance. The costs of raw materials, methods of implementation, and low production capacity make these composites expensive and are limited to industries such as aeronautics and aerospace. Carbon fibres (high modulus and high strength) (Shi et al. 2014) and Kevlar (very light) (Shi et al. 2014; Ou et al. 2010) are more used in such composites. Composites reinforced with discontinuous fibre are used to bridge the gap between the properties of composites with long fibres (continuous) (Malha et al. 2013) and non-reinforced polymers used in applications with low loads. If the fibres are sufficiently long, the stiffness of the composite may approach that of a continuous fibre system. On the other hand, the implementation of the similar non-reinforced polymers is always possible, by injection, extrusion, or compression, allowing large production capacity (Fernandes et al. 2014; El-Sabbagh et al. 2014; Valente et al. 2011). The final properties of an advanced composite are shaped not only by the kind of matrix, reinforcing materials it contains (fillers), and synergy between the fillers and the matrix but also by the filler size, filler morphology, filler loading, dispersion/distribution of fillers into the matrix, and the manufacturing process (implementation condition, used process) (Salleh et al. 2014).

It is known that characteristics of composite materials depend essentially on technology of manufacturing processes of materials. The processing of composite materials is also one of the largest research areas where the academic community and industry should make a concerted effort (Feldmann and Bledzki 2014; Al-Maadeed et al. 2014). The development of composite materials is a complicated process and requires the simultaneous consideration of different parameters such as the geometry of the components, the volume of production, the type of reinforcement and the matrix, the requirements of tooling, and process market economy (Al-Maadeed et al. 2014). Improvements and innovation in manufacturing and assembling techniques for fibre-reinforced polymer composite materials and structures are necessary to achieve materials with low cost and high performance to allow wider adoption in many industrial sectors (Gamon et al. 2013; Ho et al. 2012). This chapter highlights the various aspects of secondary processing of polymer matrix composites.

9.2 Processing of Polymer Composites Based on Short Fibre

Polymer composite materials based on natural fibre have been fabricated by using several manufacturing techniques which are designed for conventional fibre-reinforced polymer composites. These techniques comprise compounding, extrusion, and injection molding, compression molding, resin transfer molding (RTM), and vacuum infusion. It is known that characteristics of composite materials depend strongly on technology of manufacturing processes. The extrusion, injection, and compression moldings are the key process to produce polymer/filler composites with a high homogeneity and a good dispersion/distribution of fillers into thermoplastic matrix (Gamon et al. 2013; Ho et al. 2012). It is important to note that the process parameters are the rotating speed of the mixer, the temperature, and time. Therefore, the investigation of the techniques is very important for the research and application of thermoplastic composites.

9.2.1 Compounding

In the compounding step, the composite is manufactured by blending the filler with the thermoplastic matrix. Usually, the compounded material can be molded or pressed into an end product or formed into pellets for further processing. The composite products can be manufactured using thermoforming, extrusion, calendaring, injection molding, or compression molding. Most polymer composites are made by extrusion, in which the fillers are incorporated into the molten thermoplastic using a screw system (Nekhlaoui et al. 2014a; Arrakhiz et al. 2013b). When more complex forms are desired, other methods such as injection or compression molding can be used. Injection molding is the process used in the automotive industry to produce plastic products. A major advantage of this manufacturing technique may be found in the ability to form complex shapes, at a high rate of production, due to the cyclical nature of this process. During the process of injection molding of thermoplastic materials reinforced with fibres, the fibre orientation is produced by the flow conditions. The molded product is anisotropic due to the fibre orientation. The material properties and characteristics of the final part are determined by the fibre orientation (Kim and Lee 2014; Notta et al. 2014; Hashimoto et al. 2012; Oumer and Mamat 2012).

9.2.2 Extrusion Condition: Screw Configuration and Temperature

Generally, the mechanical properties of composite materials depend strongly on the fibre properties, fibre content, fibre orientation, fibre distribution/dispersion into polymer matrix, interfacial adhesion, and the manufacturing process (Essabir et al. 2013a; Kakou et al. 2014). During the extrusion process, the torque was measured to compare the extrusion force required to transmit and blend the filler and matrix. Also, the matter pressure was controlled, as a response of compound state into the die. This parameter can be influenced by many variables such as the geometry of the die, screw configuration, screw speed, the fibre content, the temperature, or the resulted viscosity of the compound.

In this study, the machine used in the optimization of extrusion process parameter is the micro-extruder Thermo Scientific HAAKE Minilab II. This device was designed for mixing small-volume samples (7 cm^3). It is a conical twin-screw extruder with a recirculation channel. Two pressure sensors are positioned in the recirculation channel for evaluating the pressure in a given velocity gradient. It can be used easily with corotating and counterrotating screws. In general, for a given geometry of the screws, the micro-extruder is used to control various parameters such as temperature, mixing time, and screw speed. In this section, the effect of temperature, screw speed, and screw configuration (corotating and counterrotating screws) on the torque and pressure of the matter was studied.

For various concentrations (5, 10, and 15 wt%) of alkali-treated fibre (bagasse, coir, and doum), three different temperatures were tested (190, 200, and 210 °C), and for each temperature three screw speeds (*n*) were tested: 60, 80, and 120 rpm. Tables 9.1 and 9.2 exhibit the evolution of torque (N cm) and pressure of matter (bars) for all composites based on natural fibres (bagasse, coir, and doum) under different temperatures and screw speeds. Results show for all types of fibres and for two screw configurations (corotating and counterrotating screws) that a linear torque increases with fibre concentration and screw speed in the compound. The more fibre added and the increase in screw speed, the more torque needed and electricity consumed in the

Table 9.1 Counterrotating extrusion screw parameters for various fibre types and loadings versus a screw speed and temperature

Bagasse fibre					Coir fibre					Doum fibre				
T (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)	T (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)	T (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)
190	60	0	33	6	190	60	0	33	6	190	60	0	33	6
		5	32	6			5	33.5	8			5	39	7
		10	34.5	7.5			10	34.5	8			10	39	8
		15	39	9			15	39	9			15	42.5	9.5
	80	0	42	8		80	0	42	8		80	0	42	8
		5	43	8			5	45	10			5	50	10
		10	44	9.5			10	47	10			10	54	10.5
		15	49	10			15	53	11			15	58	11.5
	120	0	60	11		120	0	60	11		120	0	60	11
		5	62	11			5	72	13.5			5	71	12
		10	70	13			10	79	14			10	80	13
		15	77.5	14			15	84	14			15	83	15
200	60	0	30	5	200	60	0	30	5	200	60	0	30	5
		5	29	5			5	37	7			5	32	6
		10	31	6			10	41	7			10	35	7
		15	33	7			15	45	7.5			15	35	8
	80	0	44	8		80	0	44	8		80	0	44	8
		5	43	8			5	47	8.5			5	44	8
		10	45	8			10	47	8.5			10	46	8.5
		15	45	9			15	48	9			15	48	9
	120	0	64	10		120	0	64			120	0	64	10
		5	62	10			5	77	11			5	64	10
		10	72	10.5			10	77.5	11.5			10	67	11
		15	90	12			15	77.5	12			15	69	12

(continued)

Table 9.1 (continued)

Bagasse fibre					Coir fibre					Doum fibre				
T (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)	*T* (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)	*T* (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)
210	60	0	40	5	210	60	0	40		210	60	0	40	5
		5	38	5.5			5	24	5			5	28	5
		10	40	6			10	28	5			10	32	6
		15	45	6.5			15	31	5.5			15	32	7
	80	0	46	6		80	0	46			80	0	46	6
		5	45	6.5			5	37	7			5	39	7
		10	45	6.5			10	37	7			10	41	8
		15	47	7			15	38	7.5			15	46	9.5
	120	0	74	8		120	0	74			120	0	74	8
		5	72	8			5	57	8.5			5	58	9
		10	74	9			10	57	8.5			10	60	10
		15	74	10			15	64	9.5			15	62	11

Table 9.2 Corotating extrusion screw parameters for various fibre types and loadings versus a screw speed and temperature

Bagasse fibre					Coir fibre					Doum fibre				
T (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)	*T* (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)	*T* (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)
190	60	0	44	9	190	60	0	44	9	190	60	0	44	9
		5	40	9			5	41	9.5			5	38	10
		10	42	10			10	43	9.5			10	45	11
		15	49	11			15	45	11			15	48	11.5
	80	0	62	11		80	0	62	12		80	0		12
		5	59	11.5			5	59	12			5	58	12
		10	61	12			10	60	12			10	63	13
		15	70	13			15	63	12			15	67	13.5
	120	0	43	8		120	0	43	8		120	0	43	8
		5	84	14			5	87	14			5	88	15
		10	89	15			10	94	14.5			10	95	16
		15	91	15			15	94	15.5			15	97	16
200	60	0	40	9	200	60	0	40	9	200	60	0	40	9
		5	34	7.5			5	41	8			5	35	7.5
		10	39	8			10	41	8.5			10	39	8.5
		15	41	9			15	44	9			15	41	10
	80	0	60	11		80	0	60	11		80	0	60	11
		5	51	9			5	58	10			5	51	10
		10	51	10.5			10	59	10			10	57	10
		15	55	11			15	61	10			15	60	10.5
	120	0	77	12.5		120	0	77	12.5		120	0	77	12.5
		5	81	13			5	74	13			5	65	13
		10	85	13.5			10	79	13			10	68	13.5
		15	87	14			15	79	13			15	69	13.5

(continued)

Table 9.2 (continued)

Bagasse fibre					Coir fibre					Doum fibre				
T (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)	T (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)	T (°C)	Speed (rpm)	Content (wt%)	Torque (N cm)	Pressure (bars)
210	60	0	39	7	210	60	0	39	7	210	60	0	39	7
		5	30	7			5	37	7			5	34	7
		10	35	7.5			10	39	7.5			10	35	7
		15	40	8			15	39	7.5			15	37	8
	80	0	47	7.5		80	0	47	7.5		80	0	47	7.5
		5	50	8.5			5	47	7.5			5	48	9
		10	51	9.5			10	49	8			10	48	9
		15	55	10			15	50	8.5			15	52	10
	120	0	67	10		120	0	67	10		120	0	67	10
		5	63	11			5	69	11			5	69	11
		10	67	11			10	71	11			10	72	11
		15	67	11			15	71	11			15	78	12.5

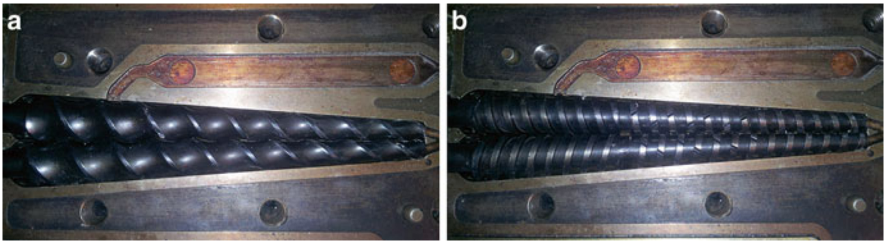

Fig. 9.1 Twin-screw extruder configuration: (**a**) corotating screw, (**b**) counterrotating screw

compound preparation. The temperature profile has an impact not only on the thermal properties of composites (in terms of degradation of fibre and matrix) but also on the manufacturing parameter. Results (Tables 9.1 and 9.2) show that increasing temperature, the torque values are decreased and at higher temperature the compound becomes more fluid (less viscous) and the compound requires low torque. Blending PP with natural fibres at different conditions (screw speed and fibre concentration) also led to a pressure rise. This result comes to join the torque study in confirming the influence of implementation parameter on manufacturing process parameter.

Moreover, it was observed that the corotating screw configuration shows a higher torque and pressure for all composite systems under different parameters (temperature and screw speed) compared to counterrotating screws. Figure 9.1 shows the screw geometry in corotating and counterrotating screw configuration. It was observed that in the two configurations, the screws are conical with the same dimension, but the difference is in the screw pitch length. The corotating screw configuration has a larger pitch than the counterrotating configuration which leads to a higher amount of mixed matter which leads to a higher pressure and torque. As screw speed and temperature were kept constant, torque and pressure of the compound must depend to two different parameters, i.e., screw geometry and the compound viscosity. First, if the feed rate was maintained constant, it was related to the matter weight. It is known that PP and fibres have different densities that could cause volume variations depending on the fibre/polymer ratio; however, the screw geometry has an important impact in terms of the amount of the transported, compressed, and mixed matter. In the case of counterrotating screw, the matter was more homogenized which allows a high shear stress, compared to corotating configuration when the matter undergoes less shear stress due to the large pitch of screw, leading to higher pressure and torque of corotating configuration.

9.2.3 Melt Rheology

Another parameter was the viscosity of matter. Therefore, the rheological behavior of produced composites was analyzed to complete the observation made during compounding. Rheology is the science that studies the response of materials (solid, liquid, gas) under applied stresses and deformations. It has been widely used to determine the rheological properties of the material such as viscosity and the elastic and viscous modulus, which lead to assess the percolated network structure, dispersion state of filler into the matrix, and the interaction between components (Elkhaoulani et al. 2013; Fernandes et al. 2013; Kabir et al. 2012). Many authors have studied the rule of rheological analysis as a potentially valuable technique to model and evaluate the microstructure, dispersion/distribution status, and the interfacial properties between fillers and the polymer matrix.

Using the MCR 500 (Physica) rheometer equipped with CTD600 device, previous strain sweep test at 1 Hz was applied to materials in order to determine strain at linear viscoelastic behavior; 5 % was chosen as strain. Frequency sweeps ranging from 500 to 0.05 Hz were applied. The measurements were carried out at 200 °C under small-amplitude oscillatory shear mode using parallel plate–plate geometry (25 mm diameter). The 2 mm thick sample disks were used for all tests. The dynamic rheological properties of composites were measured to characterize the dispersion state of the fibre within the polymer matrix, the degree of interaction between fibres and the polymer matrix, and the viscosity of the melt. In addition, the processing behavior of the polymer composites could also be obtained from their rheological characteristics. The dynamic rheological properties of three composites (PP/doum, PP/coir, PP/bagasse) were measured, and the PP/doum was taken as an example. Figure 9.2 shows the changes of the dynamic moduli (G′ and G″) and viscosity of

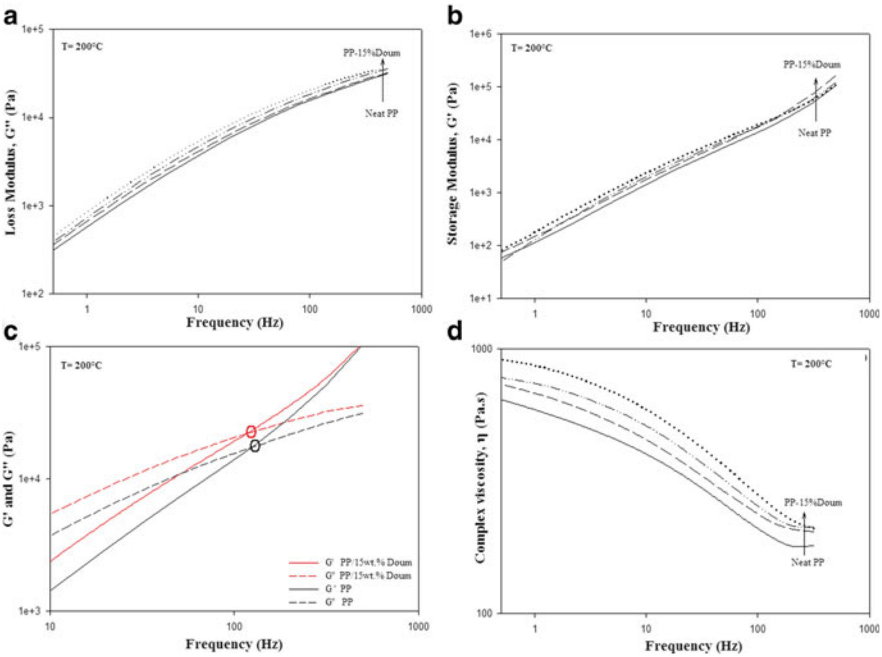

Fig. 9.2 Melt rheological properties as a function of fibre content and frequency at temperature of 200 °C: (**a**) loss modulus, (**b**) storage modulus, (**c**) crossover point, and (**d**) complex viscosity

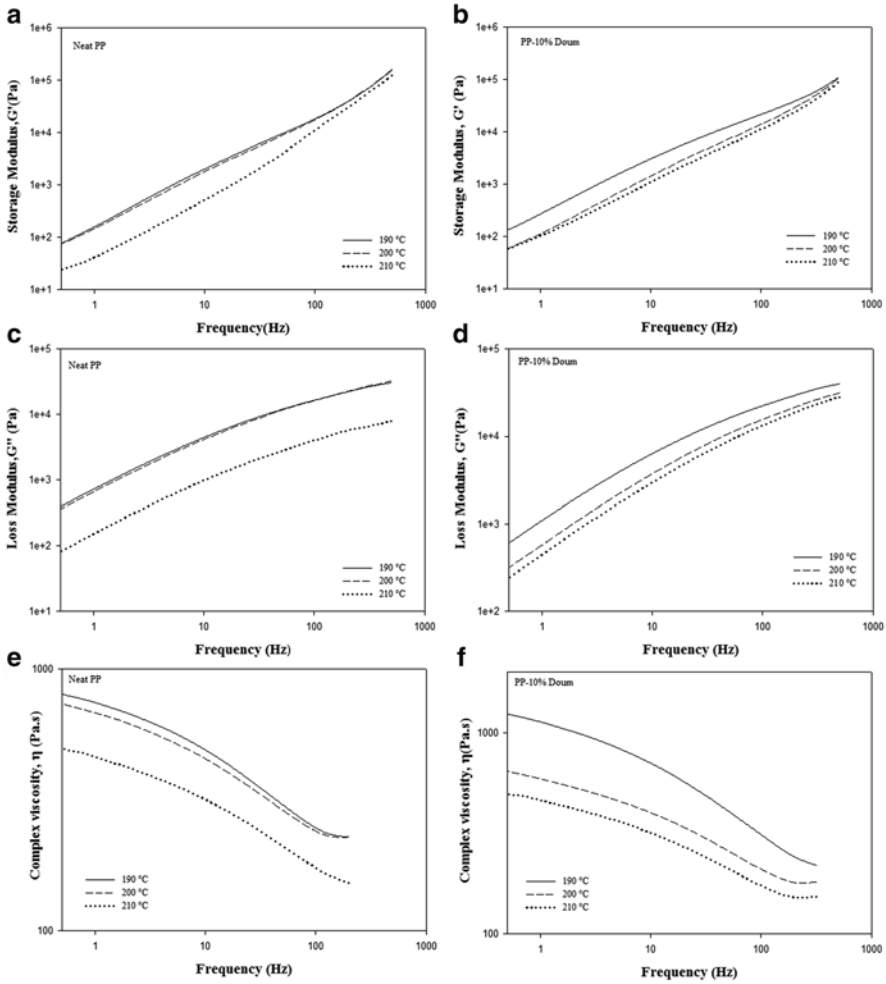

Fig. 9.3 Melt rheological properties as a function of frequency and temperature (**a**, **a**′): storage modulus, (**b**, **b**′) loss modulus, and (**c**, **c**′) complex viscosity

the melt as a function of doum fibre content and frequency. It can be obviously seen that the incorporation of fibres increases the dynamic properties of composites. This increase in complex modulus is attributed to the addition of rigid fibres leading to a change in the polymer chain displacement and the change in the molecular dynamics (Nekhlaoui et al. 2014a); this result reflects also the good interfacial adhesion between alkali-treated fibres and polymer matrix; removal of noncellulose compounds during alkali treatment exposes hydroxyl groups on the fibre's surface and favors interfacial bonding between the fibre's surface and polymeric matrix (Boujmal et al. 2014); this improvement in the interfacial adhesion affects the final properties of manufactured composites. In addition, as can be seen from Fig. 9.3, the storage modulus of the composites (5, 10, and 15 wt% fibre content) is higher than that of neat PP, indicating the stress transfer from the matrix to the fibres. It was also observed from Fig. 9.3 that, increasing frequency, both G′ and G″ increase; the noted result is related to the relaxation time response of polymeric chains (Essabir et al. 2013b, c). At high frequencies, the material behavior will be more like a solid because the polymeric chains will not get enough time to attend permanent deformation (irreversible flow) (Essabir et al. 2013b, c). But at low frequency, the polymer chains have enough time to relax. On the other hand, at higher frequencies, the elastic behavior is dominant compared to the viscous ones, whereas at low frequencies the viscous behavior is the predominant. It is important to notice that with addition of fibre, the storage and loss moduli increase, while the crossover frequency where G′ = G″ decreased. Figure 9.2c illustrates the crossover frequency, and it is noted that the crossover frequency values go from 125.99 to 116.98 Hz when doum fibre content increased from 0 to 15 wt%, respectively. This reduction in crossover frequency indicates that G″ becomes smaller than G′ and shows a plateau-like nonterminal behavior which implies elastic characteristic of the melt. This elastic behavior at lower frequency can be explained by a good interaction between the fibre and matrix which is enhanced by the chemical modification of fibres.

The complex viscosity curves as a function of fibre content and frequency are shown in Fig. 9.2d. These measurements showed a marked increase in complex viscosity with the introduction of fibres from 0 to 15 wt%. As can be seen in the figure, at 15 wt% fibre content, the complex viscosity of the melt is 100 times higher than the neat PP viscosity which makes the blending and processing of composite more difficult. Indeed, the distribution and dispersion of solid and rigid fibres into molten polymer prevent its flow (increase in the flow resistance) which is reflected by an increase in viscosity. It was found in previous works (Gamon et al. 2013) that the viscosity is closely related to the fibre–fibre interaction, fibre–matrix interaction, and fibre orientation. Moreover, it was observed that the melt viscosity decreases with increasing frequency, indicating the existence of a yield stress. At low frequency, shear rate was insufficient to ensure the mobility of the system. The presence of fibres perturbs normal polymer flow and hinders the mobility of chain segments, the fibres are randomly oriented, and polymer chains are entangled, and the fibre–fibre interaction and the frictions were more important. Gamon et al. (2013) observed that a good affinity between the compounds leads to a higher viscosity. In our case, the used alkali treatment enhances the compatibility between the fibre and matrix and ensures a good interfacial adhesion.

However, at higher shear rate, the fibres got oriented in the flow direction, and the fibre–fibre interaction was diminished; the lower perturbations in melt flow resulted in viscosity decrease. In conclusion, the increase of viscosity with fibre concentration is most likely due to an increase of the matter pressure and torque observed during the compounding process.

The problem in the compounding process is the difficulty to compound the fibres with thermoplastic polymer at high processing temperatures without thermally decomposing the natural fibre. Thus, the viscosity of the matter is also an important parameter in the manufacturing process; high viscosity makes the processing and blending of material composites more difficult. It is known that the temperature affects significantly the rheological dynamic properties (G′, G′, and complex viscosity) of the polymer composites. Figure 9.3 shows the changes in the dynamic properties of manufactured composites compared to neat PP as a function of temperature (Tazi et al. 2014; Parparita et al. 2014). As can be seen in Fig. 9.3, the dynamic modulus decreases with increase in temperature; this may be due to the softening of materials which led to the increase in the ability of the matrix chains to move freely. Moreover, it was observed that the viscosity decreases significantly in the composites compared to neat PP when temperature increases (Fig. 9.3c, c′). The presence of fibres makes the viscosity more sensitive to temperature; this is may be due to the internal humidity of fibres (Tazi et al. 2014; Parparita et al. 2014).

9.3 Composite Manufacturing

9.3.1 Extrusion

The thermoplastic polymer composites reinforced by short fibre are generally manufactured by extrusion processes and compression or injection molding (Arrakhiz et al. 2013b; Qaiss et al. 2014). These material composites provide improvements in mechanical properties such as strength, stiffness, and impact strength and have the advantage that can be shaped by the same processes as thermoplastic unfilled and without large modification tools. In every composite, there is an optimal fibre content where mechanical and rheological properties of composites are desirable. This optimum is affected by diverse parameters, among which are the fibre and matrix nature, the fibre aspect ratio, the distribution/dispersion of the fibre into a matrix (fibre agglomeration), the fibre–matrix interfacial adhesion, the fibre orientation, and the processing technique. A good dispersion and distribution of the fibres into the matrix can be achieved by effective compounding of the various components and by a suitable compounding process (extrusion) (Essabir et al. 2013a, b; Arrakhiz et al. 2013c; Nekhlaoui et al. 2014b; Sebaibi et al. 2014). The interfacial adhesion can be enhanced by using physical, chemical modification or by coupling agents or compatibilizers. However, a fibre modification and correct processing will produce homogeneous structure and fibre matrix interface that improve the final properties of manufactured composite. The mechanical properties for composite materials are carried out from tensile, flexural, compression, torsion, and dynamic analysis. The uniaxial tensile test is the most frequent mechanical test used for the characterization of materials. The tensile test is used to determine the following characteristics: The Young's modulus or stiffness modulus (E in MPa), the tensile strength or maximum stress (reached during the stress) (σM in MPa), the strain at yield (εM in mm/mm), and the plastic energy. Young modulus is determinated from stress versus strain curve, between 0.0025 and 0.005 % strain, according to ISO 527-1 (2012), which is seen to be linear for these tapes.

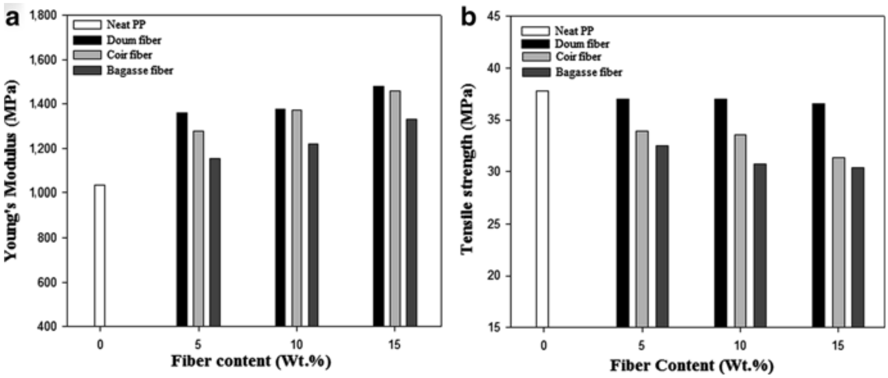

Fig. 9.4 Tensile properties of various fibre content for each fibre type as function of fibre content: (**a**) Young's modulus and (**b**) tensile strength

Figure 9.4 shows the influence of fibre loading and fibre species (doum, coir, and bagasse,) on the mechanical properties of polymer matrix. All fibre was alkali treated to remove noncellulosic components from the fibre surface and to enhance the compatibility between the hydrophilic fibres and the hydrophobic matrix. When compared to neat PP, the trend clearly shows an increase in the Young's modulus with results depending on the fibre type and loading. For all used fibres (Doum, Coir and Bagasse), the maximum Young's modulus gain is found in composites having 15 wt% fibre content. The Young's modulus shows a remarkable improvement from 1,034 MPa for neat PP to a higher value of 1,479 MPa for a 15 wt% doum fibre composite (Fig. 9.4a). This level of enhancement in the Young's modulus is assigned to a high quality of stress transfer from the matrix to fillers as natural fibres (Qaiss et al. 2014). A good dispersion and distribution of fibres into matrix ensured by the optimized manufacturing process parameter used, including screw configuration (corotating), screw speed (80 rpm), and temperature profile (200 °C), also explain this increase in the Young's modulus. Moreover, from Fig. 9.4b, tensile strength shows a decreased trend with the incorporation of fibres, and the maximum tensile strength loss was found at 15 wt%. The losses measured in tensile strength compared to neat PP are 4 % for the doum composite, 17 % for a coir composite, and 19 % for the bagasse composite. This decrease in tensile strength is quite a typical characteristic of natural fibre-reinforced composites (Qaiss et al. 2014). However, it was observed that the tensile strength of doum composite remains almost constant for all content. This is due to the good wetting of the treated fibre by the PP matrix, which leads to a good interfacial adhesion between compounds (Qaiss et al. 2014).

As a conclusion, the process used for composite production has a high importance too. Good fibre dispersion is needed to aim good material performance. Fibre orientation play a role in the mechanical properties of the materials, the longitudinal direction of fibres (compared to the stress direction) provides à higher properties (Wang et al. 2011). Twin-screw extrusion is a high shear process that can help to match good fibre dispersion (Arrakhiz et al. 2012c).

9.3.2 Injection

Injection molding consists in injecting using a rotating screw, a molten polymer in a cavity whose walls are kept at a temperature below the solidifying temperature or glass transition temperature of the polymer used. It is composed generally of two functional groups (Fig. 9.5): the injection group and the mold group. The injection process comprises a first step of mixing and plasticizing composite granules when the screw plasticized is rotated, when the screw plasticized is rotated and the molten polymer or polymer composite is forced through an orifice (gate) into a mold cavity, which is, namely, a filling stage, where it solidifies under pressure in the shape of the mold cavity, and then injecting the melt into the cavity by translation of the polymer screws, after cooling the polymer material in the mold and the final product is ejected (Arrakhiz et al. 2012c; Essabir et al. 2014).

During the injection processes, each fibre is transported by the flow, and its direction evolves according to the stress imposed by the matrix and the die walls. However, the fibres orient themselves and show a privileged orientation, which results in an anisotropic property of final composite materials. The anisotropy induced by the presence of fibres is an important property that must be taken into account in the design of materials and mold. In effect, the final characteristics and properties of the material composites are determined by the fibre orientation. At the same time, induced by the heterogeneous distribution and nonhomogeneous concentration of fibre length in the flow is often a source of defects that can induce early fatigue of the composite material. It is therefore essential to study the effect of the fibre orientation on mechanical properties of the manufactured composites.

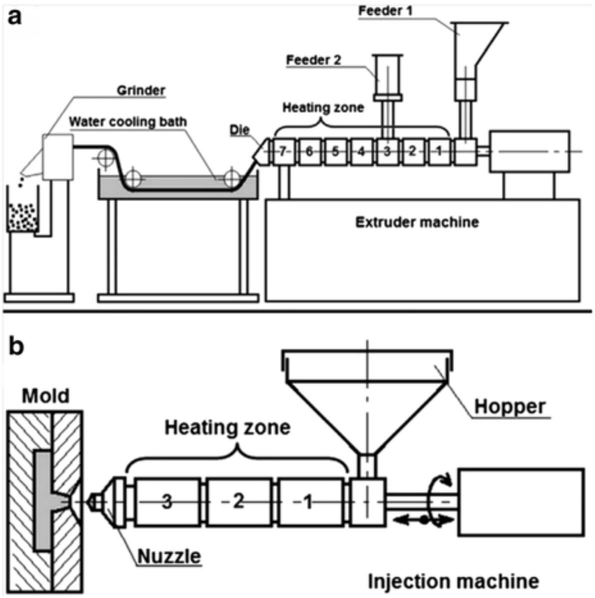

Fig. 9.5 (**a**) Extrusion process and (**b**) injection process

In this chapter, the composite is prepared by extrusion and injection molding process. Composites of polypropylene (PP) filled with *5*, *10*, and *15* wt% of alkali doum fibre were blended using a LEISTRITZ ZSE-18 twin-screw extruder (LEISTRITZ EXTRUSIONSTECHNIK GMBH, Germany). The melt-extruded composites were cooled in a water bath and then pelletized into granules of about *2* mm length, from which test samples were molded using an injection molding machine (ENGEL e-Victory). The principle of operation of the extruder and injection is shown in Fig. 9.5.

9.3.3 Mechanical Properties: Fibre Orientations

The compounded materials (polypropylene (PP) filled with *5*, *10*, and *15* wt% of alkali doum fibre) are also molded by injection process to the tensile specimens as well as the rectangular plate's samples of $100 \times 100 \times 3$ mm^3 (Fig. 9.6). However, it is known that during the injection process, the fibres are transported by the flow and its direction evolves according to the stress imposed by the matrix and the die walls.

In order to investigate the effect of the fibre orientation distribution on the tensile properties, each rectangular sample was cut into five specimens according to ISO 527-3 ($60 \times 8 \times 2$ mm^3) in different placing (Fig. 9.6). Two types of fibre orientation distribution were induced by adopting different placing directions. Two cases were considered for the placing directions according to the flow and tensile direction: parallel placements where the flow direction and the tensile direction are parallel (P1, P2, P3, P4, and P5) and transverse placements where the flow direction and tensile directions are transversal (T1, T2, T3, T4, and T5). Figure 9.6 illustrates each of the placing directions adopted for the manufacture of the specimens. The tensile

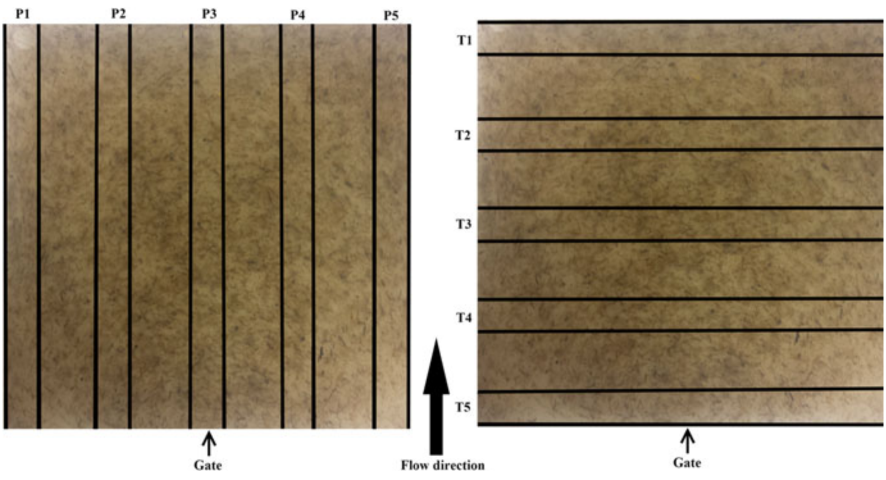

Fig. 9.6 Cutoff mechanical samples (along the parallel and transversal direction of the flow and tensile direction) of the composite

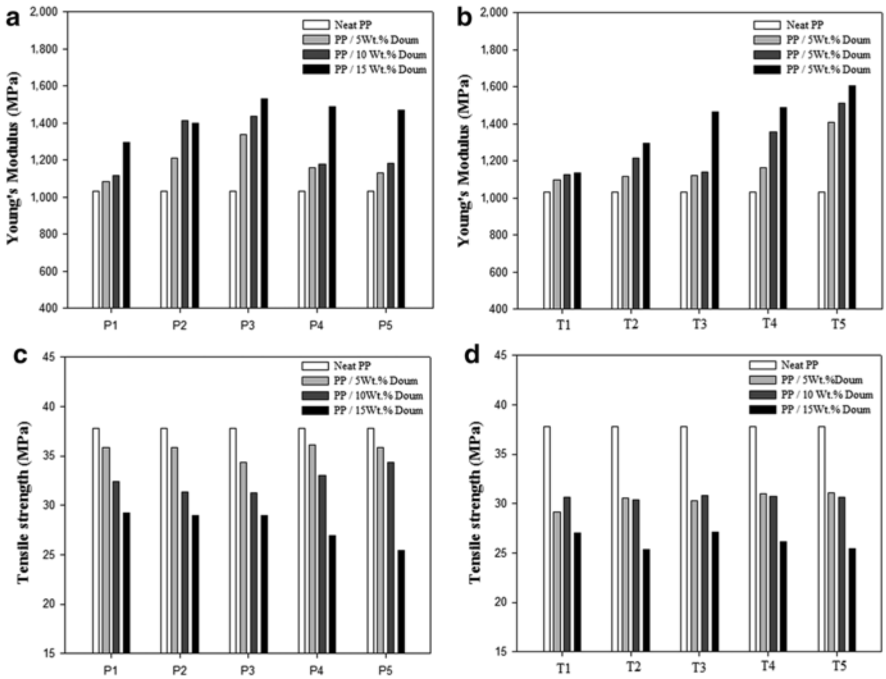

Fig. 9.7 Young's modulus and tensile strength of different doum fibre content, adopting different placing directions: (**a**, **b**) parallel placements where the flow direction and the tensile direction are parallel and (**a**′, **b**′) transverse placements where the flow direction and tensile directions are transversal

properties of samples were carried out under room temperature conditions (25 °C, 40 % relative humidity). Tensile tests were carried out according to ISO 527-3 using a universal testing machine (Instron 8821S, USA) at a crosshead speed of 5 mm/min using 5 kN load cell without extensometer.

Many factors influence fibre orientation in injection molding. Numerous studies have examined the orientation of fibres in short fibre-reinforced thermoplastic moldings (Nekhlaoui et al. 2014b; Sebaibi et al. 2014; Meyer et al. 2014). The modulus of short fibre-reinforced plastics is significantly affected by the fibre orientation against the loading direction, and the fibre orientation was generated by the polymer flow during the molding process. For this reason, it is important for strength prediction to analyze the fibre orientation in the reinforced plastics.

Figure 9.7 shows the evolution of tensile properties as the Young's modulus and tensile strength as a function of fibre loading and fibre direction against the flow and tensile direction. In the first observation, it is clear that the fibre content influences the tensile properties; this is due to the reinforcing effect of natural fibre. In addition, the increased fibre content did not bring a homogeneous distribution and could not contribute to the tensile property in longitudinal direction to achieve a maximum value. However, from Fig. 9.7, the Young's modulus of the manufactured composite is influenced by flow direction against the tensile direction. In the case of parallel placements where the flow direction and the tensile direction are parallel (P1, P2, P3, P4, and P5) and the P3 placement shows the higher Young's modulus for all fibre content, this position corresponds to the core of samples; however, at the extremity position (P1 and P5), the Young's modulus is lower. This can be explained by the fibre orientation at different positions; at core position, fibres are mostly aligned along the flow and tensile direction; in this case, the stress is directly applied to the fibres, and the response of materials shows a higher rigidity. While at extremity position, the fibres are mostly aligned perpendicular to the flow and tensile direction, and the stress is applied to the interface between the fibre and matrix leading to the lowest Young's modulus.

In the transverse configuration where the flow direction and tensile directions are transversal, it was found from Fig. 9.7a′ that the Young's modulus decreases by moving away from the injection point (gate) from maximum value at T5 position to lower one at T1 position, for all fibre content. The losses measured in tensile strength are 22, 29, and 20 % at 5, 10, and 15 wt% fibre content, respectively (Fig. 9.7b, b′). It is found that in the gate extremity, fibres tend to orient toward the transversal flow direction (and parallel to tensile direction), while at the core position the fibres are oriented transversally to the tensile direction. This highly anisotropic fibre orientation in the composites away from the injection point (gate) could be the main reason for the lower mechanical property, where the stress is applied directly to the interface between the fibre and matrix.

Figure 9.7b, b′ shows the tensile strength of manufactured composites as a function of flow and tensile direction. It is noted that the tensile strength curve shows a stabilized trend at various positions and for two configurations (parallel and transverse to flow and tensile direction). However, it was concluded that the tensile strength has not been affected by fibre orientation, because the tensile strength gives an idea of the degree of interfacial adhesion; however, in our case, the interfacial adhesion between fibres and matrix was assured by chemical treatment of fibres which ensure a good affinity and compatibility between them.

9.4 Conclusion and Perspective

The aim of this chapter was to study the processing of natural fibre-reinforced thermoplastic matrix with twin-screw extrusion and injection process using polypropylene as the matrix polymer and alkali-treated fibres (doum, coir, and bagasse). In the first part of the work, natural fibre-reinforced polypropylene composites were manufactured using twin-screw extrusion. The effect of twin-screw extrusion conditions as temperature, screw configuration and screw speed on the manufacturing process parameter on the extruder device as matter pressure and torque during the compounding process of composites was investigated. The second part of the work focused on the fibre orientation during the injection process; during the injection processes, the fibres are transported by the flow, and it orient themselves leading which to an anisotropic property of final composite materials. The study showed that a better reinforcement is achieved by using the alkali-treated fibres (doum, bagasse, and coir) with a higher aspect ratio, when the manufacturing process was optimized. The extrusion conditions have a remarkable influence of the processing, and the corotating screw configuration shows an optimized value in terms of pressure and torque. However, the study of fibre orientation shows that the increased fibre content did not bring a homogeneous distribution. In addition, it was found that for parallel placements at core position, fibres are mostly aligned along the flow and tensile direction and at extremity position the fibres are mostly aligned perpendicular to the flow and to the tensile direction, while, for the transversal placements, fibres tend to orient parallel to the tensile direction in the gate extremity and transversally in the core position.

As a second step of this study and due to anisotropic properties of the molded composites, it is very important to clarify the relationship between the fibre orientation and mechanical properties by using a distribution of fibre orientation by a numerical analysis method performed by the numerical simulation in comparison with the results obtained from the image processing.

References

Al-Maadeed MA, Shabana YM, Khanam PN (2014) Processing, characterization and modeling of recycled polypropylene/glass fibre/wood flour composites. Mater Des 58:374–380

Arrakhiz FZ, El Achaby M, Kakou AC, Vaudreuil S, Benmoussa K, Bouhfid R, Fassi-Fehri O, Qaiss A (2012a) Mechanical properties of high density polyethylene reinforced with chemically modified coir fibers: impact of chemical treatments. Mater Des 37:379–383

Arrakhiz FZ, Elachaby M, Bouhfid R, Vaudreuil S, Essassi M, Qaiss A (2012b) Mechanical and thermal properties of polypropylene reinforced with alfa fiber under different chemical treatment. Mater Des 35:318–322

Arrakhiz FZ, El Achaby M, Benmoussa K, Bouhfid R, Essassi EM, Qaiss A (2012c) Evaluation of mechanical and thermal properties of Pine cone fibers reinforced compatibilized polypropylene. Mater Des 40:528–535

Arrakhiz FZ, Malha M, Bouhfid R, Benmoussa K, Qaiss A (2013a) Tensile, flexural and torsional properties of chemically treated alfa, coir and bagasse reinforced polypropylene. Compos Part B Eng 47:35–41

Arrakhiz FZ, Benmoussa K, Bouhfid R, Qaiss A (2013b) Pine cone fiber/clay hybrid composite: mechanical and thermal properties. Mater Des 50:376–381

Arrakhiz FZ, El Achaby M, Malha M, Bensalah MO, Fassi-Fehri O, Bouhfid R, Benmoussa K, Qaiss A (2013c) Mechanical and thermal properties of natural fibers reinforced polymer composites: doum/low density polyethylene. Mater Des 43:200–205

Boujmal R, Essabir H, Nekhlaoui S, Bensalah MO, Bouhfid R, Qaiss A (2014) Composite from polypropylene and henna fiber: structural, mechanical and thermal properties. J Biobased Mater Bioenergy 8:246–252

Elkhaoulani A, Arrakhiz FZ, Benmoussa K, Bouhfid R, Qaiss A (2013) Mechanical and thermal properties of polymer composite based on natural fibers: Moroccan hemp fibers/polypropylene. Mater Des 49:203–208

El-Sabbagh A, Steuernagel L, Ziegmann G, Meiners D, Toepfer O (2014) Processing parameters and characterisation of flax fibre reinforced engineering plastic composites with flame retardant fillers. Compos Part B Eng 62:12–18
Essabir H, Hilali E, Elgharad A, El Minor H, Imad A, Elamraoui A, Al Gaoudi O (2013a) Mechanical and thermal properties of bio-composites based on polypropylene reinforced with nut-shells of Argan particles. Mater Des 49:442–448
Essabir H, Nekhlaoui S, Malha M, Bensalah MO, Arrakhiz FZ, Qaiss A, Bouhfid R (2013b) Biocomposites based on polypropylene reinforced with almond shells particles: mechanical and thermal properties. Mater Des 51:225–230
Essabir H, Elkhaoulani A, Benmoussa K, Bouhfid R, Arrakhiz FZ, Qaiss A (2013c) Dynamic mechanical thermal behavior analysis of doum fibers reinforced polypropylene composites. Mater Des 51:780–788
Essabir H, Bensalah MO, Bouhfid R, Qaiss A (2014) Fabrication and characterization of apricot shells particles reinforced high density polyethylene based bio-composites: mechanical and thermal properties. J Biobased Mater Bioenergy 8:344–351
Feldmann M, Bledzki AK (2014) Bio-based polyamides reinforced with cellulosic fibres – processing and properties. Compos Sci Technol 100:113–120
Fernandes EM, Mano JF, Reis RL (2013) Hybrid cork–polymer composites containing sisal fibre: morphology, effect of the fibre treatment on the mechanical properties and tensile failure prediction. Compos Struct 105:153–162
Fernandes EM, Correlo VM, Mano JF, Reis RL (2014) Polypropylene-based cork–polymer composites: processing parameters and properties. Compos Part B Eng 66:210–223
Gamon G, Evon P, Rigal L (2013) Twin-screw extrusion impact on natural fibre morphology and material properties in poly(lactic acid) based biocomposites. Ind Crop Prod 46:173–185
Hashimoto M, Okabe T, Sasayama T, Matsutani H, Nishikawa M (2012) Prediction of tensile strength of discontinuous carbon fiber/polypropylene composite with fiber orientation distribution. Compos Part A Appl Sci 43:1791–1799
Ho M, Wang H, Lee JH, Ho C, Lau K, Leng J, Hui D (2012) Critical factors on manufacturing processes of natural fibre composites. Compos Part B Eng 43:3549–3562
ISO 527-1 (2012) Plastics—determination of tensile properties—part 1: general principles
Kabir MM, Wang H, Lau KT, Cardona F, Aravinthan T (2012) Mechanical properties of chemically-treated hemp fibre reinforced sandwich composites. Compos Part B Eng 43: 159–169
Kakou CA, Arrakhiz FZ, Trokourey A, Bouhfid R, Qaiss A, Rodrigue D (2014) Influence of coupling agent content on the properties of high density polyethylene composites reinforced with oil palm fibers. Mater Des 63:641–649
Kim JW, Lee DG (2014) Study on the fiber orientation during compression molding of reinforced thermoplastic composites. Int J Precis Eng Manuf Green Technol 1:335–339
Malha M, Nekhlaoui S, Essabir H, Benmoussa K, Bensalah M-O, Arrakhiz F-Z, Bouhfid R, Qaiss A (2013) Mechanical and thermal properties of compatibilized polypropylene reinforced by woven doum. J Appl Polym Sci 130(6):4347–4356
Merkel K, Rydarowski H, Kazimierczak J, Bloda A (2014) Processing and characterization of reinforced polyethylene composites made with lignocellulosic fibres isolated from waste plant biomass such as hemp. Compos Part B Eng 67:138–144
Meyer KJ, Hofmann JT, Baird DG (2014) Prediction of short glass fiber orientation in the filling of an end-gated plaque. Compos Part A Appl Sci 62:77–86
Nekhlaoui S, Essabir H, Bensalah MO, Fassi-Fehri O, Qaiss A, Bouhfid R (2014a) Fracture study of the composite using essential work of fracture method: PP–SEBS–g–MA/E1 clay. Mater Des 53:741–748
Nekhlaoui S, Essabir H, Kunal D, Sonakshi M, Bensalah MO, Bouhfid R, Qaiss A (2014b) Comparative study for the talc and two kinds of Moroccan clay as reinforcements in polypropylene-SEBS-g-MA matrix. Polym Compos. doi:10.1002/pc.22986

Notta-Cuvier D, Lauro F, Bennani B (2014) An original approach for mechanical modelling of short-fibre reinforced composites with complex distributions of fibre orientation. Compos Part A Appl Sci 62:60–66

Ou R, Zhao H, Sui S, Song Y, Wang Q (2010) Reinforcing effects of Kevlar fiber on the mechanical properties of wood-flour/high-density-polyethylene composites. Compos Part A Appl Sci 41:1272–1278

Oumer AN, Mamat O (2012) A study of fiber orientation in short fiber-reinforced composites with simultaneous mold filling and phase change effects. Compos Part B Eng 43:1087–1094

Pan Y, Zhong Z (2014) Modeling of the mechanical degradation induced by moisture absorption in short natural fiber reinforced composites. Compos Sci Technol 103:22–27

Parparita E, Darie RN, Popescu C-M, Uddin Md A, Vasile C (2014) Structure–morphology–mechanical properties relationship of some polypropylene/lignocellulosic composites. Mater Des 56:763–772

Qaiss AEK, Bouhfid R, Essabir H (2014) Natural fibers reinforced polymeric matrix: thermal, mechanical and interfacial properties. Springer, Switzerland

Rojo E, Alonso MV, Oliet M, Del Saz-Orozco B, Rodriguez F (2015) Effect of fiber loading on the properties of treated cellulose fiber-reinforced phenolic composites. Compos Part B Eng 68:185–192

Salleh FM, Hassan A, Yahya R, Azzahari AD (2014) Effects of extrusion temperature on the rheological, dynamic mechanical and tensile properties of kenaf fiber/HDPE composites. Compos Part B Eng 58:259–266

Sebaibi N, Benzerzour M, Abriak NE (2014) Influence of the distribution and orientation of fibres in a reinforced concrete with waste fibres and powders. Constr Build Mater 65:254–263

Shi S, Sun Z, Hu X, Chen H (2014) Carbon-fiber and aluminum-honeycomb sandwich composites with and without Kevlar-fiber interfacial toughening. Compos Part A Appl Sci 67:102–110

Shouha P, Swain M, Ellakwa A (2014) The effect of fiber aspect ratio and volume loading on the flexural properties of flowable dental composite. Dent Mater 30:1234–1244

Tazi M, Erchiqui F, Godard F, Kaddami H, Ajji A (2014) Characterization of rheological and thermophysical properties of HDPE–wood composite. J Appl Polym Sci. doi:10.1002/app.40495

Unterweger C, Brüggemann O, Fürst C (2014) Effects of different fibers on the properties of short-fiber-reinforced polypropylene composites. Compos Sci Technol 103:49–55

Valente M, Sarasini F, Marra F, Tirillò J, Pulci G (2011) Hybrid recycled glass fiber/wood flour thermoplastic composites: manufacturing and mechanical characterization. Compos Part A Appl Sci 42:649–657

Wang J, Geng C, Luo F, Liu Y, Wang K, Fu Q, He B (2011) Shear induced fiber orientation, fiber breakage and matrix molecular orientation in long glass fiber reinforced polypropylene composites. Mater Sci Eng A 528:3169–3176

Węcławski BT, Fan M, Hui D (2014) Compressive behaviour of natural fibre composite. Compos Part B Eng 67:183–191

Zakikhani P, Zahari R, Sultan MTH, Majid DL (2014) Extraction and preparation of bamboo fibre-reinforced composites. Mater Des 63:820–828