

Chapter 16

Green Composite Film Synthesized from Agricultural Waste for Packaging Applications



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1 Introduction

The extensive uses of non-biodegradable plastics have crossed the permissible limits of plastics wastes and created environment imbalance condition all over the world. The incinerated or land filled plenty available agro-wastes released greenhouse gases, smoke and particulate matters directly in to the environment and breached environment laws. So, researchers should find the appropriate way in order to reduce the plastics wastes generation by using alternate biodegradable material such as agricultural wastes, i.e., wheat straw, rice straw, hemp, kenaf, jute, sisal, etc. [1–4]. The cellulosic property of agro-waste has motivated the researchers to synthesize biodegradable composites due to their renewable properties, provided remarkable mechanical and thermal stabilities as compared to non-biodegradable plastics. Generally, polymers such as polyethylene, polylactic acid, polypropylene, starch and agro-wastes have remarkable stability for green packaging applications. The present chapter explores the reliable use of abundantly available biodegradable agricultural wastes for packaging applications such as food, sprout and active packaging's. The perfect use of agricultural wastes as a reinforcing agent in polymer matrix is the eye opener for current researchers. Many researchers promoted the use of agro-waste to synthesize biodegradable packaging film due to their cheap, lightweight, high tensile strength and benchmark thermal stability. Enhanced cellulose percentage area in agro-waste encourages suitability of this fiber to minimize the contribution of polymers with green agro-waste. Thus, the suitable use of agro-waste to synthesize green composites for packaging application is need of the hour [5, 6].

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Native-agro-waste incorporated polymeric films have low mechanical and impact strengths as compared to petroleum-derived materials due to their hydrophilic nature. Inherent composition (lignin and hemicellulose) does not permit agro-waste to reinforce the polymeric film successfully. This behavior of agro-waste showed incompatibility found between the materials resulting in low desirable properties present in synthesized film. Moreover, this signified the less mechanical stability of the agro-waste reinforced polymeric film. However, many chemical modification techniques have enhanced surface property of agro-waste for appropriate blending in polymer matrix. This strategy helped to remove undesirable contents from agro-waste and created some void openings at the surface for improving blending characteristics. Chemical, physical and biological treatments are some of the frequently used popular techniques for reducing recalcitrance nature of agro-waste. The findings of many scientists elucidated that the mechanical and thermal characteristics of modified agro-waste incorporated polymeric film had considerably higher as compared to native-agro-waste reinforced polymeric film [7]. So the surface advancement in agro-waste is an emerging field in the polymeric composite research (Fig. 1). The prepared composite film is reliable as compared to synthetic packaging film in terms of contact angle, water vapor migration rate, flexural strength, water vapor permeability, mechanical stability and impact strength for green composite packaging application.

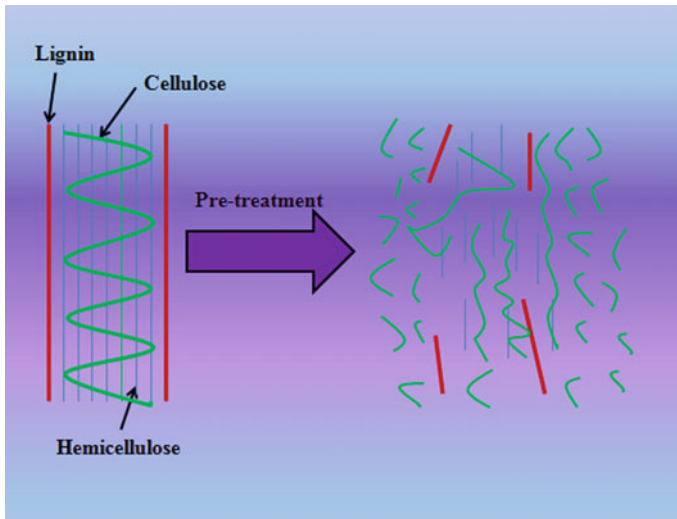


Fig. 1 Pretreatment of agricultural waste (*Source* Author)

1.1 Agro-waste

Agricultural wastes are generally produced from various from agricultural activities by the farmers. Agro-waste is waste which has no economic value. In India, the annual production of agro-waste is around 910 metric tons. Moreover, agro-waste has lightweight, readily available, comparatively low cost product and simple to process material. So, the safe disposal and better utilization of agro-waste are the key prospects for green life. Many scientists claimed that agro-waste had a property to use as filler in polymer matrix to increase composites tensile stability for various applications such as automotive, packaging, household and building industries [8]. Thus, it will build a new green environment for human life.

1.2 Classification of Agricultural Waste

In present juncture, the environment aspects encouraged researchers to increase the use of agro-waste as reinforcing agent in polymer matrix and maintain the environment balance condition. Agro-waste can be categorized in two ways, i.e., field residues and process residues (Fig. 2). Field residue is classified such as stems, stalks, leaves and seed pods and process residues are classified such as husks, seeds, roots and bagasse. [9]. Agro-wastes embedded products are required low maintenance with long life durable ability. However, these virtues are attracted industrialist to surging use of agro-waste for green composite preparation.

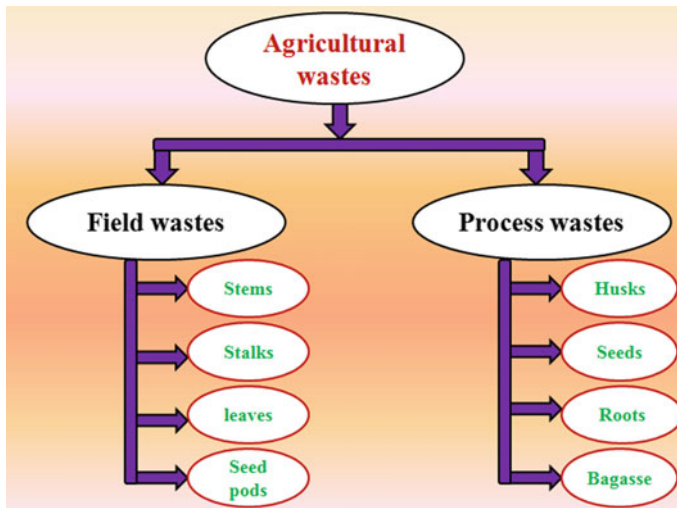


Fig. 2 Classification of agricultural wastes (Source Author)

1.3 Chemical Composition of Agricultural Waste

Agro-waste is composed of cellulose tightly embedded with hemicellulose protected by a strong wall of lignin. The percentage of cellulose, lignin and hemicellulose in agro-waste is varying with the type of agro-waste [9]. In order to attain the desirable properties, the higher cellulosic percentage in agro-waste has provoked researchers to use them for synthesizing green packaging film. The chemical composition of different types of agro-waste is depicted in Table 1.

2 Green Packaging Film Synthesized from Agricultural Waste

The required combination of agro-waste and polymers, i.e., green composites sustained a promising alternative to replace the non-biodegradable petroleum-derived materials for packaging applications. The main key factor is reduced the consumption of synthetic plastics, while researchers are moving toward renewable sources that are agricultural wastes to synthesize green plastics [4]. Recently, polymers are used for several applications like packaging, automotive and furniture goods. Thus, the use of agricultural wastes opens the way for biodegradable composite film and prepares a more sustainable waste-based packaging film that also helps to boost our economic sector. The various polymers such as polyethylene, polypropylene, polylactic acid

Table 1 Chemical composition of several types of agricultural waste

Agro-waste	Cellulose (%)	Hemicellulose (%)	Lignin (%)	References
Wheat straw	38–45	15–31	12–20	[10]
Hemp	57–77	14–22.4	3.7–13	[10]
Banana	53.45	28.56	15.46	[11]
Coir	43.44	0.25	45.84	[11]
Jute	59–61	22.1	15.9	[11]
Seed flax	43–47	24–26	21–23	[10]
Sisal	65–68	10–22	9.9–14	[12]
Bagasse	40–55.2	25.3	16.8	[10]
Kenaf	72	20.3	9	[13]
Rice husk	35–45	19–25	20	[13]
Rice straw	41–57	33	8–19	[13]
Flax	62–72	18.6–20.6	2–5	[14]
Ramie	68–76	13–16	0.6–0.7	[13]
Bamboo	26–65	30	5–31	[14]

Source Author

and polyvinyl alcohol have been frequently used for the packaging sector. Moreover, many agricultural wastes, i.e., wheat straw, hemp fiber, rice straw, kenaf, banana, jute etc. have been used as a filler in polymer matrix due to their considerable mechanical strength [4]. Although, the use of biocomposites for packaging applications made from lignocellulosic fiber is the most adaptable technique for green market (Table 2).

3 Some Improving Interfacial Interaction Techniques for Polymer/Agro-waste Composites

Pretreatment is the imperative technique involved in the effective reinforcement of agricultural waste in the polymer matrix. Several strategies are available such as physical, chemical, biological and physiochemical treatments for enhancing the interfacial interaction between fiber and polymer [30]. In the present scenario, these strategies have been reported in many literatures to attain the higher mechanical and thermal characteristics of the agro-waste incorporated green composite film (Fig. 3). Moreover, literature confirmed that treated-agro-waste was a promising substitute of polymer to synthesize green composites for packaging applications and serve as a potential alternative of non-biodegradable petroleum-derived materials [31]. In other words, these interfacial interaction improving techniques are the possible solution of poor mechanical stability of native agricultural waste-based polymer composite film.

3.1 Physical Pretreatment

In order to change the specific surface area or degree of polymerization of agro-waste, many physical treatment processes such as grinding, milling and chipping machines are used. The main focus of using this treatment is to avoid the wastage of chemicals use for requiring changes in lignocellulosic biomass. This treatment has reduced the particle size that increases the bulk density of treated-agro-waste and makes it appropriate for blending in the polymer matrix. The main disadvantage of this treatment is required higher energy consumption to operate the process [32].

3.2 Chemical Pretreatment

This pretreatment involved the reaction changes in agricultural waste. This treatment is easily removed undesirable materials (hemicellulose and lignin) from agro-waste and increases its cellulose digestibility for polymer adhesion. Many chemicals such

Table 2 Green packaging films synthesized from agricultural waste

Raw materials (reference)	Parameters	Pre-treatment	Method
Polystyrene (PS) [15]	Wheat straw (WS) (0–20%)	Chemical pre-treatment	Solvent casting method
Polyethylene (PE), polypropylene (PP) [4]	Wheat straw (0–20%)	Alkali treatment	Solvent casting method
Starch, polyvinyl alcohol (PVA) [16]	Banana fiber (BF) (0–30%)	NaOH treatment	Solvent casting method
Polylactic acid (PLA) [17]	Kenaf (KF)	Silane, alkaline treatment	Melt blending method
Cinnamon essential oil (CE), Bisphenol-A-based benzoxazine (BOZ) [5]	Hemp fiber (HF) (6–24%)	NaOH treatment	Hydraulic hot-press
PLA [18]	Glycidyl methacrylate-grafted (GMA) (1%), tert-butyl perbenzoate (TBPB) (0.5%), hemp hurd (HH) (10–30%)		Extrusion and injection
High density polyethylene(HDPE) [19]	HF (10–30%), BF (10–40%), maleic anhydride modified high density polyethylene (MAPE) (0.8–1.2%)		Injection
PP [20]	HF (5–15%)	NaOH treatment	Hand lay-up

(continued)

Table 2 (continued)

Raw materials (reference)	Parameters	Pre-treatment	Method
Polyhydroxy butyrate (PHB) [21]	Almond shell, rice husk, seagrass (10–20%)	NaOH treatment	Melt blending
Polylactic acid (PLA) [22]	WS (0–30%)	Solid state shear milling process (SSSM)	Extrusion
Green epoxy [23]	Sisal	Mechanical pressing	Unidirectional single layer laminates
Polyvinyl alcohol [24]	Rice straw (RS) (1–3%)	NaOH treatment	Solvent casting method
Hydroxy propyl methyl cellulose [25]	Jute fiber (0–3%)		Solvent casting method
Starch [26]	Rice husk fiber (2–10%)	Sulphuric acid treatment	Solvent casting method
Polylactic acid (PLA), poly (butylene succinate) PBS [27]	Cellulose crystals (CNC) (extracted from Hemp fiber)	Sulphuric acid treatment	Solvent casting method
Low density polyethylene (LDPE) [28]	Doum fiber (DF) (5–30%)	NaOH treatment	Extrusion
PP [29]	Alfa fiber (AF)	Chemical treatment	Hot-press

(continued)

Table 2 (continued)

Results		Applications		
PS Tensile strength (TS) \approx 3 MPa Elongation at break (EAB) \approx 210%	PS/20% WS TS \approx 2.3 MPa EAB \approx 110%	PS/20% Alkali treated-WS TS \approx 3.56 MPa EAB \approx 140 %	PS/20% Acid treated-WS TS \approx 3.14 MPa EAB \approx 130%	Packaging
PE/PP TS \approx 46.5 MPa EAB \approx 122% Contact angle \approx 121° WVTR \approx 47.50 g m ⁻² day ⁻¹	PE/PP/30% Native-WS TS \approx 34 MPa EAB \approx 116.20% Contact angle \approx 92° WVTR \approx 132 g m ⁻² day ⁻¹		PE/PP/30% Alkali treated-WS TS \approx 45.010 MPa EAB \approx 119.98% Contact angle \approx 117° WVTR \approx 51.89 g m ⁻² day ⁻¹	Packaging
PVA/Starch/BF TS \approx 30.8 MPa	PVA/Starch/20% Alkali treated-BF TS \approx 34.4 MPa		PVA/Starch/20% Acid treated-BF TS \approx 19.2 MPa	Packaging
PLA TS \approx 55 MPa Impact strength (IS) \approx 38 J/m		PLA/Alkali treated-KF TS \approx 8 MPa IS \approx 70 J/m		Packaging
CE/BOZ TS \approx 28.05 MPa Tensile modulus (TM) \approx 1.60 GPa Strain \approx 2.10% Water absorption (WAB) \approx 0.75%		CE/BOZ/Treated-HF (20%) TS \approx 55.74 MPa TM \approx 3.47 GPa Strain \approx 1.38% WAB \approx 5%		Packaging

(continued)

Table 2 (continued)

Results		Applications	
PLA TS \approx 65 MPa EAB \approx 104.5% Flexible strength (FS) \approx 100 MPa	PLA/30% HH TS \approx 45 MPa EAB \approx 102% FS \approx 60 MPa	GMA-g-PLA/30% HH TS \approx 50 MPa EAB \approx 102.1% FS \approx 80 MPa	Packaging and disposable goods
	HDPE TS \approx 20 MPa Strain \approx 30%	65% HDPE/30% BF/5% MAPE TS \approx 50 MPa Strain \approx 7.25%	65% HDPE/30% HF/5% MAPE TS \approx 37.5 MPa Strain \approx 7.5%
PP TS at 10 mm thickness \approx 40 MPa Fracture toughness (FT) at 10 mm Thickness \approx 4.2 MPa m ^{0.5}	95% PP/5% Alkali treated-HF TS at 10 mm thickness \approx 60 MPa FT at 10 mm thickness \approx 4.3 MPa m ^{0.5}	85% PP/15% Alkali treated-HF TS at 10 mm thickness \approx 44 MPa FT at 10 mm thickness \approx 3.95 MPa m ^{0.5}	Automobiles, military, construction, packaging, aerospace, railway coach
	PHB TS \approx 40 MPa EAB \approx 101.7% WVP \approx (2.1) \times 10 ⁻¹⁵ kg s ⁻¹ Pa ⁻¹ m ⁻¹	PHB/20 % Almond shell TS \approx 35 MPa EAB \approx 101.25% WVP \approx 5 \times 10 ⁻¹⁵ kg s ⁻¹ Pa ⁻¹ m ⁻¹	PHB/20% Seagrass TS \approx 40 MPa EAB \approx 100.75% WVP \approx 1.75 \times 10 ⁻¹⁵ kg s ⁻¹ Pa ⁻¹ m ⁻¹
PLA TS \approx 70 MPa EAB \approx 107%		PLA/30% WS TS \approx 60 MPa EAB \approx 103.85%	PLA/30% WS (SSSM) TS \approx 68 MPa EAB \approx 103.1%

(continued)

Table 2 (continued)

Results		Applications
Epoxy/73% native-SF TS \approx 320 MPa	Epoxy/77% treated-SF TS \approx 410 MPa	Packaging
PVA TS \approx 41 MPa Transparency \approx (68–95)%	PVA/NaOH treated-RS TS \approx 98 MPa Transparency \approx (52–90)%	Packaging
Hydroxypropylmethyl cellulose film TS \approx 41.56 MPa EAB \approx 141% Transparency at 600 nm \approx 89% WVTR \approx 0.053 g/cm ² /24 h	3% Jute fiber based hydroxypropylmethyl cellulose film TS \approx 57.86 MPa EAB \approx 145% Transparency at 600 nm \approx 78% WVTR \approx 0.039 g/cm ² /24 h	Packaging
Starch TS \approx 2.5 MPa Water uptake \approx 250%	Starch/10% cellulose nanocrystals Tensile strength \approx 3.4 MPa Water uptake \approx 100%	Packaging
PLA WVP \approx 0.071×10^{-11} g mm kPa ⁻¹ h ⁻¹ m ⁻² Transmittance at 450 nm \approx 88.2%	PLA/20% PBS WVP \approx 0.042×10^{-11} g mm kPa ⁻¹ h ⁻¹ m ⁻² Transmittance at 450 nm \approx 86%	Packaging
LDPE TS \approx 12.75 MPa EAB \approx 140%	70% LDPE/30% TDF TS \approx 10 MPa EAB \approx 40%	Packaging
PP Young modulus (YM) \approx 700 MPa TS \approx 35 MPa	Raw-AF/PP YM \approx 1000 MPa TS \approx 31 MPa	Etherified-AF/PP YM \approx 900 MPa TS \approx 32 MPa
	Alkali treated-AF/PP YM \approx 1200 MPa TS \approx 31 MPa	Esterified-AF/PP YM \approx 1400 MPa TS \approx 32 MPa
	PLA/20% PBS/3% CNC WVP \approx 0.042×10^{-11} g mm kPa ⁻¹ h ⁻¹ m ⁻² Transmittance at 450 nm \approx 81%	Packaging

Source Author

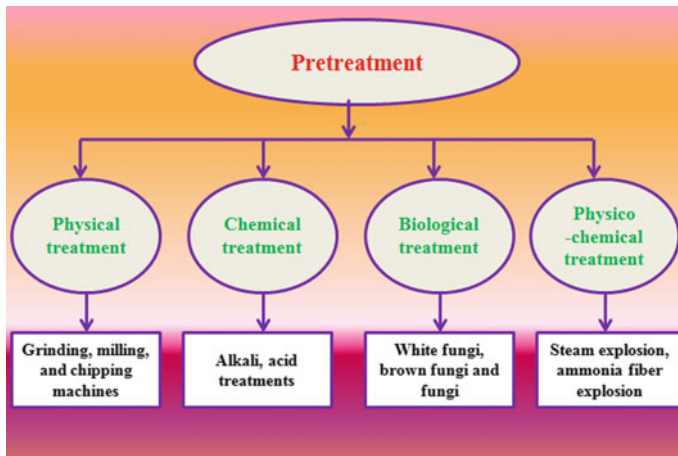


Fig. 3 Types of pretreatment (*Source* Author)

as alkalis (KOH, NaOH, $\text{Ca}(\text{OH})_2$ etc.), acids (HCl , H_2SO_4 , H_3PO_4 , etc.) and ionic liquids have been commonly used for hydrolysis of hemicellulose with delignification process. This pretreatment can be considered in one of the most effective treatments for making more cellulose accessible [32].

3.3 *Biological Pretreatment*

Biological pretreatment is considered as a minimum energy required process in which agro-waste is kept under a control atmosphere with preferred microorganism. That microorganism has created the desirable surface changes of the agro-waste that makes biomass more acceptable for polymer adhesion. In this treatment, many organisms, i.e., white, brown and fungi have been used for enhancing the suitability of agro-waste in the polymer matrix. But this process has required larger residence time with an effective control atmosphere represent this treatment less attractive as compared to other pretreatment processes [33].

3.4 *Physicochemical Pretreatment*

This category of treatment is classified as a combination of physical and chemical changes. In this treatment, agro-waste is treated at higher temperatures and pressure which breaks the recalcitrance structure of agro-waste and creates some desirable surface changes in the agro-waste. Many physicochemical treatment processes are steam explosion, ammonia fiber explosion, liquid hot water hydrothermal treatment,

wet oxidation, etc., are commonly used for improving interfacial interaction between polymer and agro-waste [32].

4 Various Testing Methods for Analyzing the Characteristics of Green Packaging Film

Several types of testing methods are used for analyzing the properties of agro-waste-based green composites for packaging application. Various testing methods such as water vapor transmission rate (WVTR), water vapor permeability (WVP), tensile test, optical characteristic test, impact test and contact angle have been used by researchers for analyzing the characteristics of packaging film [15].

4.1 Water Vapor Transmission Rate and Water Vapor Permeability

WVTR is an essential property for analyzing the quality of packaging film. This test explores the water vapor migration rate through the green composite film. A low value of this rate signifies synthesized film is applicable to keep warm products warm. In this test, a glass cup of 100 ml was taken and filled with distilled water. After that, cup is covered by synthesized film tightly and observed the weight of that wet cup chamber. This chamber is placed in an incubation chamber at a known temperature with fixed relative humidity. The changes in a total weight of wet cup chamber are analyzed at a regular interval of a time period. Water vapor transmission rate can be determined using the following equation.

$$\text{WVTR} = \frac{\text{WC}_1 - \text{WC}_2}{\text{WC}_1 * A * \text{day}} \quad (1)$$

where WC_1 and WC_2 represent the initial and final weights of wet cup chamber; A represents the exposing area of wet cup chamber [34].

Water vapor permeability is also determined using the following equation.

$$\text{WVP} = \frac{\text{WVTR} * \text{thickness of green film}}{P(\text{RH}_1 - \text{RH}_2)} \quad (2)$$

where P is saturation vapor pressure at constant temperature, and RH_1 and RH_2 are relative humidity of inside and outside of the wet cup chamber.

4.2 Tensile Test

This test is demonstrated the mechanical stability of green composite film using universal testing machine. This property depends on materials thickness, testing speed and method of preparation. In this test, film is cut in strip form at fixed gauge length and gauge width according to ASTM. Generally, many researchers have been used ASTM D 0882 for materials thickness less than 1 mm. A sample is fixed between the grips of testing machine and allowed to elongate a material at a known testing speed. Further, the machine calculated the ultimate tensile strength and maximum flexibility of the green film that plays a vital role in observing the mechanical stability of the material [4].

4.3 Dart Impact Test

Dart impact strength is an important property for industries of packaging sectors to access the durability of the packaging film. A sample is cut in required dimensions according to ASTM. Commonly, ASTM D 1709 has been used frequently for analyzing the impact strength of the green packaging film. In this impact test, a dart of known weight is free fell on the surface of the film at a fixed height and observed that film is punctured or not. If it is not punctured, again increase the weight of dart and repeated the process. The phenomena helped to understand the impact strength of the green packaging film. The energy required to fracture the surface of film is equivalent to drop a known dart impact failure weight at fixed height on the sample.

4.4 Contact Angle Test

In order to elucidate the hydrophobic property of green film, water contact angle is measured at vapor–liquid interface meets at solid surface of sample using drop shape analyzer machine. Sessile drop method is the simplest way to observe the water repelling quality of the prepared film. This test is measured by the wettability of the surface of the sample. In this test, a drop of distilled water is dropped on the surface of the composite film and examined the surface hydrophobicity of the sample. Higher contact angle (greater than 90°) signified the hydrophobic property of the green composite film [4].

4.5 *Optical Characteristics Test*

This test visualized the transparency of the green composite film for packaging applications using UV–Vis spectrophotometer machine. Green film is cut in strip form and placed in a cuvette using blank cuvette as a reference in the machine and scanned over the wavelength of 400–800 nm visible range. This test is provided the transparency of the sample over visible range and demonstrated the optical characteristics of the green film [35].

4.6 *Thermal Stability Test*

Thermal stability test is provided information about ability of the packaging film to resist the action of heat using PerkinElmer thermogravimetric analyzer. This test represents the maximum temperature at which our materials can sustain their mechanical property. In this test, a known weight of green film is placed in a machine and allowed to heat from 30 to 800 °C at required heating rate. This process is done in a nitrogen-controlled atmosphere for avoiding the undesirable thermal cracking of the green film. Thermal analyzer provided the graph between mass (%) with temperature. This graph helped to understand the main thermal degradation temperature range of the green composite film [21].

5 Conclusion

In summary, the effect of agricultural wastes on the packaging properties of the polymeric film was evaluated. This chapter has enlightened the perfect use of agricultural waste as a reinforcing agent in the polymer matrix for packaging applications. Various improving interfacial interaction techniques, i.e., physical, chemical, biological and physiochemical treatments have been discussed in order to enhance the suitability of agricultural waste in the polymer matrix in terms of water vapor migration rate, transparency, mechanical and thermal stabilities. Published literature confirmed the importance of various strategies for agricultural waste for polymer blending. This chapter is also explored the testing methods, i.e., water vapor transmission rate, water vapor permeability, contact angle, tensile test, impact test, optical characteristics test, thermal stability test and some ASTM standards for tensile and impact tests. This chapter described a brief introduction for the utilization of abundantly available agricultural wastes as filler in the polymer matrix for green packaging application.

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