

REVIEW PAPER

POLYMER BIOCOMPOSITES BASED ON AGRO WASTE: PART II. HUSK, STALK AND STRAW OF SOME AGRICULTURAL CROPS AS DISPERSED FILLER

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Abstract. A special place in the field of modern composite materials is occupied by the production of new types of structural and functional materials that increase energy efficiency, sustainability, safety and durability of final products, as well as reduce material consumption and cost. In this regard, an increased interest has recently been received by polymer composite materials based on renewable natural sources (biocomposites), in which plant fibers, i.e. agro waste are used as a reinforcing filler. Large amounts of lignocellulosic materials are produced around the world as a result of agricultural activities. Agricultural residues include rice straw, wheat straw, rice husk and sunflower husk, which are mostly left on the fields after harvests. This review article will focus on polymer biocomposites based on the mentioned agro-waste.

Keywords: *biocomposites, agro waste, dispersed filler, chemical treatment.*

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1. Introduction

Currently, public attention is paid to environmentally friendly composite materials made from natural fillers and polymer materials (Sarki *et al.*, 2011). Lignocellulose fillers are considered as scientific and technological innovation in the field of new materials, highlighting the importance of using agro waste as raw materials. In addition, composites made from lignocellulosic materials have received considerable attention due to their properties: high specific strength, high modulus, high volume application, low cost, low density, low energy consumption, easy workability, renewable origin and recyclability (Kiziltas *et al.*, 2011; Navas *et al.*, 2015; Zaaba & Ismail, 2019). The development of these environmentally friendly composites has accelerated rapidly because they are non-toxic, biodegradable and safer to work with (Chitra & Vasanthakumari, 2012; Ofora *et al.*, 2016). Eco-friendly composite materials, also known as biocomposites, are made from one or more phases derived from biological origin (Kuciel *et al.*, 2010). Adding plant fibers/fillers such as sugar cane (Vidyashri *et al.*, 2019; Agunsoye & Aigbodion, 2013), soy protein (Thakur & Kessler, 2014a; Thakur & Kessler, 2014b; Thakur *et al.*, 2014c; Thakur *et al.*, 2014d), wheat straw (Mengeloglu & Karakus, 2008), rice husk (Chen *et al.*, 2015a; Kumar *et al.*, 2019), peanut shell (Sareena *et al.*, 2012), coconut husk (Mishra, 2017), cotton stalk (Bekele *et al.*, 2017), etc. as a natural filler, important for reinforcing polymers. Beyond that the

addition of natural filler to the composite material, i.e. reuse and recycling of agricultural waste can minimize environmental problems associated with their accumulation (Prithivirajan *et al.*, 2015; Nguyen *et al.*, 2019).

There have been many studies in the literature on the development of new composites containing both long and short natural fibers such as: bamboo (Md Shah *et al.*, 2016; Banga *et al.*, 2015; Muhammad *et al.*, 2019), jute (Wang *et al.*, 2019; Singh *et al.*, 2018; Kumar & Srivastava, 2017), linen (Amiri & Ulven, 2016; Moudood *et al.*, 2019), kenaf (Radzuan *et al.*, 2019; Dashtizadeh *et al.*, 2019; Sarifuddin *et al.*, 2012; Tholibon *et al.*, 2019), sisal (Senthilkumar *et al.*, 2018; Ibrahim *et al.*, 2016), agave (Cisneros-López *et al.*, 2017; Glouia Y *et al.*, 2019; Cisneros-López *et al.*, 2017; Annandarajah *et al.*, 2019), ramie (He *et al.*, 2008; Zhang *et al.*, 2015), coconut (Mulinari *et al.*, 2011; Kumar *et al.*, 2016), cotton (Raftoyiannis, 2012; Lin *et al.*, 2011), kapok (Mani *et al.*, 2012; Sharma *et al.*, 2017) etc.

At present, with the exception of fibrous natural fillers, much attention is paid to the loading of dispersed agricultural waste in the polymer matrix. In this regard, waste of lignocellulosic plants, such as shells, husks and stems of agricultural crops can be used not only as a source of biomass, but also as inexpensive dispersed fillers for polymer composites after special treatment. From a number of agricultural crops in which the shell or husk, as well as the stalk is used as a natural filler, the following can be distinguished: rice (Yiga *et al.*, 2020), sunflower (Ashori & Nourbakhsh, 2010), wheat (Zena *et al.*, 2009), buckwheat (Andrzejewski *et al.*, 2019), different nuts (e.g. walnuts, almonds, pistachios, hazelnuts, peanuts, etc.) (Sutivisedsak *et al.*, 2012; Tufan *et al.*, 2015; Zhang *et al.*, 2019).

2. Husk, stalk and straw of some agricultural crops as dispersed filler

2.1. Rice (lat. *Oryza*) is one of the staple crops grown all over the world and has the same value as wheat as a staple food (Foo & Hameed, 2009; Soltani *et al.*, 2015). It is the second most consumed food in the world (Fernandes *et al.*, 2016) and covers about 1% of the Earth's surface (Adam *et al.*, 2012). Globally, the annual total production of paddy rice is about 600 million tons, and on average 20% of this, or about 120 million tons, is husk (Louis & Thomas, 2013). It is estimated that on average, paddy rice is composed of approximately 20-22% husk, 5-8% bran and 70-72% rice (Somvanshi *et al.*, 2017). Rice husk is the main agricultural waste of the rice industry. This is the outermost layer of the rice grains and is removed from the rice grains during the milling process (Fig. 1).

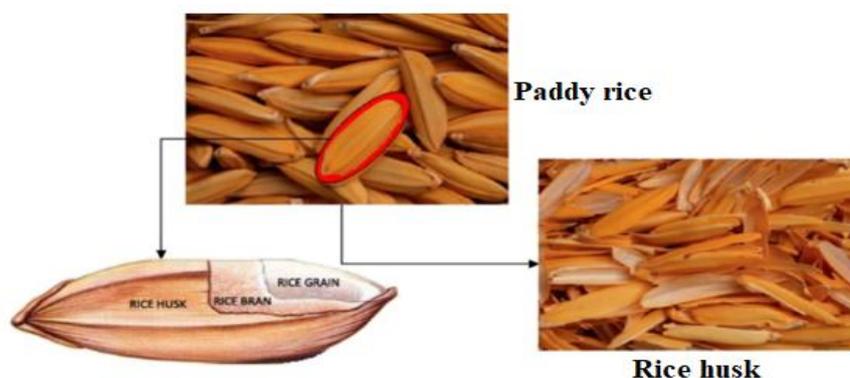


Figure 1. Rice grain: the corresponding structures and rice husk (Fernandes *et al.*, 2016)

The chemical composition of rice husk depends on many factors, such as the type of paddy rice, the type of fertilizer used, the chemical composition of the soil, the harvest year, climate and geographic conditions (Pode, 2016; Babaso & Sharanagouda, 2017). The main components and physical properties of rice husk are summarized in Table 1, as reported by various researchers (Arjmandi *et al.*, 2015; Ludueña *et al.*, 2011; Ugheoke & Mamat, 2012; Ishak, 2011; Shukla *et al.*, 2014). The use of rice husk is very limited due to its undesirable properties such as resistance to degradation, low nutritional value and high ash content (Athinarayanan *et al.*, 2015).

Table 1. Components and physical properties of rice husk

| Properties | Rice Husk |
|----------------------------------|-----------|
| Components, % | |
| Cellulose | 25-35 |
| Hemicellulose | 18-21 |
| Lignin | 26-31 |
| SiO ₂ (silica) | 15-17 |
| Soluble substances | 2-5 |
| Moisture content | 5-10 |
| Physical properties | |
| Particle size (µm) | 26.64 |
| Surface area (m ² /g) | 0.92 |
| Density (g/cm ³) | 1.00 |

The presence of silica in rice husk is advantageous compared to other biomass due to potential applications in electronics, ceramics and polymer materials (Carmona *et al.*, 2013), while the high silica content makes rice hulls abrasive (Ghosh & Bhattacharjee, 2013), resistant to water penetration and fungal decomposition (Rahman *et al.*, 2010).

There are studies in the literature that use thermoplastic polymers such as polypropylene, (Rosa & Santos, 2009; Raghu *et al.*, 2018; Mohamed *et al.*, 2020), polyethylene (Bilal *et al.*, 2013; Atuanya *et al.*, 2013), polyvinyl chloride (Chand & Jhod, 2008; Ramle *et al.*, 2013; Crespo *et al.*, 2008; Chand *et al.*, 2012) and polystyrene (Rozman *et al.*, 2000; Victor, 2020) as matrices for rice husk composites, as well as thermoset matrices such as epoxy (Lai *et al.*, 2017; Bisht & Gope, 2015; Jain *et al.*, 2019) and polyamides (Batteggazzore *et al.*, 2016). In the production of composites filled with rice husk, both thermoplastic and thermosetting plastic can be used, in both virgin and recycled forms. Because waste plastics offer a promising source of raw materials for composites due to the huge amount generated each day and lower cost than virgin plastics, the use of recycled plastics for the production of composites filled with rice husk has been studied by a number of authors (Orji & McDonald, 2020; Chen *et al.*, 2015b; 245, p. Hong *et al.*, 2016; Abdul Azam *et al.*, 2020; Chen *et al.*, 2018; Ghofrani *et al.*, 2012; Chen *et al.*, 2016; Tong *et al.*, 2014; Chen *et al.*, 2020). These studies confirm the importance of recycling plastic waste and natural fiber residues into sustainable composite products for industrial and domestic applications.

The tensile stress of rice husk composites generally decreases with increasing filler concentration due to poor interfacial bonding between the hydrophobic polymer matrix and the hydrophilic filler (Yang *et al.*, 2004). To increase and improve the adhesion between these chemically different components of polymer composites, it is necessary to optimize the properties of the interface. This is often achieved either by treating or modifying the fiber surface with appropriate chemicals or by using agents

that improve compatibility (Huner, 2017). Chemical modifications also improve dimensional stability and reduce water absorption of composites.

Alkaline treatment is one of the most commonly used chemical treatments for natural fillers. Alkaline treatment, i.e. mercerization and acetylation, fibrillate lignocellulosic fibers, reducing their aspect ratio, increasing surface roughness and giving them access for better penetration into the matrix, which leads to increased adhesion. The mercerization and acetylation of rice husks were used to study these effects (Ugheoke *et al.*, 2011). The higher tensile strengths exhibited by the mercerized rice husk composite specimens are greater than the acetylated rice husk specimens, due to excessive fibrillation. This led to a decrease in the mechanical properties of polypropylene-based composites filled with acetylated rice husk compared to composites filled with mercerized rice husk. The results of the study of the physical and thermal characteristics of polypropylene composites reinforced with rice husk treated with alkali showed that the physical properties, as well as the thermal stability of the composites, are significantly improved when adding alkali-treated rice husk to the polypropylene matrix (Luna *et al.*, 2015).

Double treatment with alkali and acetylation on rice husk for the preparation of composites showed that the introduction of rice husk into a polyethylene matrix significantly increases the tensile and flexural modulus of a composite prepared from 10 wt% mercerized and acetylated rice husk fiber and unmodified polyethylene matrix (Fávaro *et al.*, 2010). This composite showed a 35% increase in Izod impact strength over the pure matrix. SEM analysis showed interfacial interaction between rice husk fibers and unmodified polyethylene and better phase compatibility provided by double treatment. Rassiah and Ali used rice husk mercerization after boiling rice husks at 100°C in water for about 1 hour (Rassiah & Ali, 2016). The results of the study showed that boiled and NaOH treated rice husks showed the highest tensile strength of 10.41 MPa (PP 90/Rice husk 10), tensile modulus of 1122.1 MPa (PP 60/Rice husk 40), hardness 12.0 (PP 60/Rice husk 40) and impact strength 2.20 J/mm² (PP 90/Rice husk 10). The presence of boiled and sodium hydroxide treated rice husk has been shown to be indicative of better properties compared to boiled and untreated rice husk.

The results of a study of the chemical and thermal stability of rice husk against alkaline treatment with 2% to 8% NaOH showed that mercerization of rice husks causes significant degradation of lignin and hemicellulose when the sodium hydroxide concentration is at least 4% (Ndazi *et al.*, 2008). Evaluation of the chemical resistance of rice husk to alkaline treatment using direct analysis showed that the proportion of lignin and hemicellulose in rice husk treated with 4-8% NaOH significantly decreased by 96% and 74%, respectively. The silica ash content also drops slightly with alkaline treatment, which indicates possible degradation of silica with alkaline treatment. The thermal stability and final decomposition temperatures of alkali-treated rice husk were also reduced by 24-26°C due to the decomposition of hemicellulose and lignin during alkaline treatment. This leads to the conclusion that alkaline treatment of rice husk with more than 4% NaOH causes significant chemical degradation of rice husk, which subsequently reduces their thermal stability. The authors also processed rice husk using steam and NaOH in order to study their effect on the properties of surface functional groups and the characteristics of composite panels based on phenol-formaldehyde resin (Ndazi *et al.*, 2007). Chemical modification of rice husk with NaOH improves the adhesion of rice husk in composites made from phenol-formaldehyde resin by removing surface impurities such as silicon dioxide and carbon compounds that block reactive

chemical groups. Thermogravimetric analysis of unmodified and NaOH-treated rice husk showed that untreated rice husk exhibited superior thermal stability compared to NaOH-treated rice husk. Decreased thermal stability of NaOH-treated rice hulls is an indicator of possible degradation of rice husk as a result of using concentrated NaOH.

Other authors, in order to solve the problem of rice husk hydrophilicity, chemically treated it with benzenediazonium salt in various media (in alkaline, acidic and neutral media) at different pH values, which in turn improved the mechanical properties of the composites (Rahman *et al.*, 2010). It has been established that the mechanical properties of composites prepared from rice husk treated in an alkaline environment significantly increase compared with those that were treated in an acidic and neutral environment. Based on the filler concentration, the composites reinforced with 35% filler had the optimal set of mechanical properties among all the composites produced.

Interface bonding is further improved when the surface of the rice husk is chemically modified with a reagent such as glycidyl methacrylate. Rozman and *et al.* observed an increase in tensile strength and toughness for rice husk/polystyrene composites by chemically modifying rice husks with glycidyl methacrylate (Rozman *et al.*, 2000). The strength of the composites decreased with increasing rice husk content. The toughness also improved as the rice husk was modified with glycidyl methacrylate. The improvement is due to improved interfacial bonding between the fiber and the polymer matrix, thus increasing the binding of the filler to the matrix due to the decreased mobility of macromolecules.

On the other hand, improved adhesion between the natural filler and the polymer matrix is often achieved through the use of compatibilizers. Compatibilizers have the ability to react in any way with both organic fillers and polymers, creating bridges at the interface. Examples of compatibilization include silanization, peroxide treatment, isocyanate, maleated polyolefin, acrylation, and latex coating treatment.

As mentioned earlier, the hydrophilic nature of the natural fibers generated by hydroxide groups and the hydrophobicity of the matrices make this combination incompatible. In this case, silane treatment improves the interfacial adhesion between natural fibers and the matrix (Arrakhiz *et al.*, 2012; Yanjun *et al.*, 2010). Silane molecules have bifunctional groups, where one of them reacts with fibers, and the other with polymer.

To determine the effect of a silane binding agent, agricultural waste was used as reinforcing fillers in thermosetting polymer composite materials (Imoisili *et al.*, 2012). The mechanical properties of the rice husk-polyester composite were improved by using the bonding agent triethoxyvinylsilane for surface treatment.

Treatment of rice husk with γ -aminopropyltrimethoxysilane improves adhesion between the hydrophilic fiber of rice hulls and hydrophobic polymer matrices polypropylene/recycled acrylonitrile-butadiene rubber, which led to a decrease in water and oil absorption in biocomposites polypropylene/recycled acrylonitrile-butadiene rubber/rice husk (Santiagoo *et al.*, 2011). And also the results of the study showed that composites based on rice husk treated with γ -aminopropyltrimethoxysilane demonstrate higher tensile strength, tensile modulus and lower elongation at break.

Good interfacial interaction between rice husk and recycled polyvinyl chloride (PVC) is achieved by introducing aminosilane as a coupling agent (Ramle *et al.*, 2013). At the same time, the tensile strength and elastic modulus of the 45% rice husk/55% recycled PVC composite increase to 66% and 84%, respectively. Similar results also

prevailed in cases where rice husk were double-treated with alkali and silanes to prepare polylactic acid-based composites (Tran *et al.*, 2014). In this case, two different silanes were used. The results showed that no difference was observed according to the nature of the silane.

The authors of work (Sun *et al.*, 2019) proposed a simple and effective method for improving the interfacial adhesive properties of rice husk and high-density polyethylene compositions using a silane coupling agent γ -aminopropyltriethoxysilane and maleated polyethylene (PE-g-MA) compatibilizer with additional modification. It was found that the binding agent γ -aminopropyltriethoxysilane crosslinks with a hydroxyl group on the husk surface and attaches to a high polymer by forming $-\text{NH}-$, $-\text{C}=\text{O}-$ functional groups. The PE-g-MA compatibilizer enhances the two phases by covalently bonding to the ester bond and reduces the cross-sectional roughness of the composites. While the modification enhances the dispersibility and mechanical properties of the polymeric compound system with high rice husk content, the flexural strength is improved by 11.5% and 28.9% with the addition of a silane coupling agent and PE-g-MA, respectively. The flexural strength of the composite with one rice husk surface modifier increased by 11.5%, and the strength of the composite containing the binder and compatibilizer increased by 40.7%.

Commonly used binders for biocomposites are copolymers containing maleic anhydride such as maleated polypropylene (PP-g-MA) (Techawinyutham *et al.*, 2016; Razavi Nouri *et al.*, 2006) or maleated polyethylene (PE-g-MA) (Petchwattana *et al.*, 2012). The anhydride groups of the copolymers can react with the surface hydroxyl groups of natural fibers to form ester bonds, while the other part of the copolymer interlaces with the polymer matrix due to their equal polarity (Simao *et al.*, 2016).

With the addition of a compatibilizer such as PP-g-MA, the interfacial bond between the rice husk and the matrix polymer is improved, resulting in improved dimensional stability and water absorption properties (Yang *et al.*, 2006), as well as strength properties (Yang *et al.*, 2007). The presence of PP-g-MA reduces the size of voids and makes the surface more uniform, confirming its effect on improving adhesion in the interfacial region (Rosa *et al.*, 2009). This improvement has even been observed in rice husk composites where a recycled thermoplastic blend of low density polyethylene and polyethylene terephthalate was used as a polymer matrix, and PE-g-MA as a compatibility agent (Chen *et al.*, 2015c).

It was also observed that the simultaneous addition of 2 wt.% PP-g-MA and 1 wt.% maleic anhydride grafted styrene-ethylene-butylene-styrene (SEBS-g-MA) to the rice husk/polypropylene composite leads to a synergistic effect (Yehet *et al.*, 2015). The impact strength and ultimate strength of the composite increased by 35 and 41%, respectively. The results of moisture absorption tests have shown that both PP-g-MA and SEBS-g-MA are effective in reducing the rate of moisture diffusion.

The authors of the work (Panthapulakkal *et al.*, 2005a) used a terpolymer based on ethylene-acrylic ester-maleic anhydride. The addition of 2.5% coupling agent increased tensile and flexural strengths by 54.8 and 20%, respectively. This indicates that the terpolymer binder improves the interaction, like any other maleated polyolefin, between rice husk and high density polyethylene. Similar results were obtained using four different coupling agents based on terpolymers of ethylene (acrylic ether)-(maleic anhydride) and terpolymers of ethylene (acrylic ether)-(glycidyl methacrylate) (Panthapulakkal *et al.*, 2005b). Terpolymer-based binders are effective in improving the performance of high density polyethylene composites filled with rice hulls. The degree

of adhesion between the filler and the high density polyethylene is determined by the chemical structure of the binder. The stiffness created by the side groups results in a less flexible interface and reduces stress transfer between the filler and the matrix.

From morphological studies of the low density polyethylene-based composite film filled with rice hulls and nanoclay, it can be concluded that the PP-g-Ma compatibilizer helps the polymer molecules to penetrate and exfoliate into clay plates in order to realize a large aspect ratio of the filler (Majeed *et al.*, 2013). Likewise the addition of the compatibilizer enhances the natural dispersion of the fiber in the continuous low density polyethylene phase and the rice husk aggregates begin to disappear with increasing compatibilizer concentration, which in turn improves homogeneity. The compatibilizer improved the strength properties of the composite film, for example, adding only 2 wt% PE-g-MA to the composite system improved the tensile strength of the film by about 22% compared to uncompatibilized films. Similar results were obtained for low density polyethylene-based composites filled with rice husk and montmorillonite (Majeed *et al.*, 2018).

The compatibilizer content plays a very important role in the formation of the mechanical properties of biocomposites. The mechanical properties of composites tend to increase in accordance with the change in the PP-g-MA content from 0 to 4 wt%, and then it decreases at 6 and 8 wt% PP-g-MA, but it is still higher compared to composites without compatibilizer (Tran *et al.*, 2013). The positive effect at a low content of PP-g-MA may be due to the reaction of hydrophilic hydroxyl groups of rice husks and anhydride groups of PP-g-MA in the esterification reaction. This phenomenon acts as an effective factor in improving the interfacial adhesion between filler and matrix. Meanwhile, at a higher content of PP-g-MA, an excess of the compatibilizer can lead to agglomeration and the formation of a new PP-g-MA phase. Therefore, it can hinder interfacial adhesion between rice husk and polypropylene.

Good interfacial interaction between rice husk and polyvinyl chloride was achieved by modifying the surface of the former using maleic anhydride as a compatibilizer (Chand & Jhod, 2008). The positive effect of rice husk reinforcement as well as maleic anhydride surface treatment is reflected in the improved tribological and mechanical properties of the polyvinyl chloride-based composite (Chand *et al.*, 2010). Surface treatment of rice husk with maleic anhydride also improves wear resistance by improving the compatibility between rice husk and polyvinyl chloride (Chand *et al.*, 2012).

Another major agricultural waste from the rice industry is rice straw, which is mostly burned or left on the field until next plowed. Rice straw is one of the many natural fiber sources that have been extensively tested for composite applications (Yao *et al.*, 2008; Ismail *et al.*, 2011; Liu *et al.*, 2012; Jayamani *et al.*, 2015; Low *et al.*, 2018; Grozdanov *et al.*, 2006).

Rice straw has been investigated for disposal in cement-based composites (Xie *et al.*, 2015). It has also been used as natural filler for injection-molded high density polyethylene biocomposite materials (Low *et al.*, 2017). Optimization efforts were also undertaken to achieve the highest possible toughness when using polymer composites (Al Amin *et al.*, 2017; Bouasker M *et al.*, 2014; Saidahet *et al.*, 2019).

Thus, rice husk and straw have been used in combination with a variety of polymer matrices including polyethylene, polypropylene, polyvinyl chloride and polylactic acid to form polymer composites. The addition of rice husk resulted in a decrease in the tensile strength of the composites. Therefore, the use of different types

and amounts of binder/compatibilizer has been reported to improve the mechanical properties of composites. In addition, second fillers such as montmorillonite, along with compatibilizers, are used to improve the mechanical properties of composites. In most of the studies conducted in composites filled with rice husk, only one polymer was used as a matrix. Limited work has been reported on the use of second fillers. Summarizing the literature, we can say that, consequently, the ability to interpret the use of polymer mixtures as a matrix and secondary fillers when creating new properties of polymer composites can open up interesting areas for research.

2.2. The sunflower (lat. *Helianthus annuus*) is the most cultivated oilseed plant, the harvest of which in the world reaches about 40 million tons, where more than 50% of sunflower seeds are husks (Salasinska & Ryszkowska, 2013a). In the literature, several authors have discussed the possibility of development of thermoplastic and thermoset composites filled with sunflower husk waste (Salasinska & Ryszkowska, 2015; Marhoon, 2017; Kaymakci *et al.*, 2013; Nerenz *et al.*, 2012; Binici *et al.*, 2014). In the study of Salasinska and co-authors, it is presented that the use of crushed sunflower husk as a filler for polyethylene composites makes it possible to obtain composite materials with improved mechanical properties (Salasinska *et al.*, 2016). Samples containing sunflower husk as a filler showed higher water absorption compared to composites modified with fillers obtained from other types of agricultural waste, such as finely ground peanut shells or pistachio shells (Salasinska & Ryszkowska, 2013b).

The mechanical, thermomechanical properties and structure of ultra-low density polyethylene in the form of matrix composites of thermoplastic elastomer filled with powdered waste of sunflower husk and the possibilities of its application are determined (Barczewski *et al.*, 2018). It was shown that the introduction of natural fillers into the ultra-low density polyethylene matrix made it possible to obtain composites characterized by an improved modulus of elasticity and tensile strength with comparable elongation at break.

The use of sunflower husk as a filler for the production of composites based on epoxy resin led to a significant increase in the viscosity of composites, which created a porous structure, which led to a decrease in the mechanical characteristics of composites based on sunflower husk and epoxy resin. Nevertheless, a decrease in strength properties is not a contraindication for the manufacture of environmentally friendly and inexpensive products (Barczewski *et al.*, 2019).

The composites based on epoxy resins with agricultural waste in the form of sunflower husk were subjected to dynamic mechanical analysis and thermogravimetric analysis, as well as measurements with a cone calorimeter (Salasinska *et al.*, 2019). Based on thermogravimetric analysis, it was found that composites have a higher thermal stability in the temperature range up to 300 °C compared to epoxy resin.

Abdullah (2017) investigated the effect of particle size and concentration of sunflower husk on tensile strength, Young's modulus and water absorption of polyester resin. The results of the study showed that the addition of 10 wt% sunflower husk as a reinforcing material to polyester resin gave the best result in terms of tensile strength and Young's modulus, where tensile strength and Young's modulus increased by 46.6% and 27.2%, respectively.

Extensive research is being carried out in the field of using sunflower husks and stems as fillers for polymer composites, but despite this, some questions still remain open.

2.3. Wheat (lat. *Triticum*) is the most widely grown crop in the world. Wheat kernels contain about 14.5% bran, which are produced in huge quantities every year as a by-product of wheat milling (Xie *et al.*, 2008). Only 10% of this byproduct is used in bakeries and breakfast cereals as a food additive. 90% of the remaining bran can be sold as animal feed, but due to high transport costs, millers often discard the bran as waste, posing a threat to the environment. The study of wheat bran as a cellulose filler in biocomposites based on natural rubber has shown that, in comparison with commercial cellulose fillers, the presence of amorphous cellulose and a high content of amino acids in wheat bran has a beneficial effect on the technological, physicochemical and morphological properties of biocomposites. In addition, the high content of macro- and microelements present in wheat bran can have a positive effect on further biodegradation of biocomposites (Formela *et al.*, 2016).

Wheat bran can be effectively used as polymer filler in the injection molding process to create low-liability items. Majewski and Cunha (2018) conducted an original study of the effect of concentration and particle size of wheat bran on the properties of low density polyethylene. The presence of wheat bran in the low density polyethylene matrix reduced the longitudinal shrinkage, which stabilized the size of the finished injection molded samples. However, it should be noted that wheat bran, due to its hydrophilic properties, has an affinity for water, which increases the water absorption of the polymer composition with an increase in the concentration of the filler. In the presence of water and in high humidity conditions, this can lead to swelling and resizing of the cast samples. It was also found that the particle size of the wheat bran has a significant impact on elongation at break. The elongation was greater when the particles were smaller.

Experimental tests have shown that the design of the screw mixing tips also has a significant effect on the characteristics of the single-screw extrusion process, as well as on the physical, mechanical and structural properties of the resulting biocomposite lignocellulose extrudate based on wheat bran and low density polyethylene (Sasimowski *et al.*, 2019). The introduction of wheat bran into polyethylene led to a slight increase in the stiffness of the sample and a significant decrease in the tensile strength and elongation. The highest strength was obtained for specimens extruded with a cutting ring tip.

World wheat statistics show that the world produces about 710 million tons of wheat straw annually [El Messiry & El Deeb, 2016]. Consequently, it has a huge impact on the environment and this problem is growing every year. In this regard, it is strongly recommended to introduce an innovative environmentally friendly process of converting agricultural waste (agricultural waste) into quality products suitable for use in industry. From this point of view, the use of straw is attracting great attention of researchers as a potential alternative lignocellulosic raw material, replacing wood for the manufacture of composites.

Panthapulakkal and Sain (2007) prepared composite materials from high density polyethylene with a high content of agricultural waste – wheat straw, corn stalk, corn cob, and wood flour (65 wt.%). Surface chemistry showed a carbon-rich surface for wheat straw compared to cornstalk, corncob, and wood flour. Wheat straw-filled high density polyethylene (HDPE) composites have shown superior mechanical properties compared to HDPE/corn stalk, HDPE/corncob, and even HDPE/wood flour composites.

Ashori and Nourbakhsh (2009) prepared completely eco-friendly, sustainable and biodegradable composites using wheat straw and rice husk as reinforcement for

thermoplastics, as an alternative to wood fibers. Mechanical properties including tensile strength, flexural strength and flexural toughness were investigated depending on the amount of fiber and binder used. Composites filled with wheat straw showed an increase in tensile and bending properties when a binder was added.

Investigation of the effect of modified wheat straw on the physicochemical properties of composites based on high density polyethylene showed that the modification with caprolactam by reducing the polarity of the fibers contributes to good compatibility and dispersion of straw fibers in the matrix (Zhang *et al.*, 2016). Composites of high density polyethylene with modified particles of wheat straw have shown excellent mechanical properties due to the greater compatibility of the filler with the matrix. It can be said that modified wheat straw fibers function as "biological steel", reinforcing high density polyethylene for the production of biocomposites.

Mengelolu and Karakus (2008) announced about thermal performance of thermoplastic composites filled with wheat straw. The authors used recycled high density polyethylene, recycled polypropylene and a 50% blend of these two polymers as a thermoplastic matrix. It has been found that wheat straw flour in a thermoplastic matrix reduces the decomposition temperature of composites. The properties of thermoplastic composites have been improved using bonding agents.

Whole and shredded wheat straws up to 10 cm in length have been used with polypropylene to make lightweight composite materials (Zou *et al.*, 2010). The influence of wheat straw concentration, wheat straw length and crushing configuration (half, quarter and mechanical crushing) on the flexural and tensile properties of composites was investigated. The sound-absorbing properties of composites from solid straw and chopped straw were studied. Compared to solid wheat straw/PP composites, mechanically shredded wheat straw/PP composites have 69% higher flexural strength, 39% higher modulus of elasticity, 18% higher impact resistance, 69% higher tensile strength and 26% higher Young's modulus, better sound absorption properties.

Agro waste can be used singly or in combination with each other to achieve the desired structure of agro waste and plastic composites. In this regard, El Messiry and El Deeb [2016] investigated the potential use of treated wheat straw/flax fiber as a reinforcing agent in polyvinyl alcohol. The research focused on whole stems of wheat straw that were reinforced with an injection of animal glue to improve the strength and crushing strength of wheat straw. Wheat straw strength increased by about 165%, tensile strain by 125%, and Young's modulus by 125%. The results support the possibilities of consuming whole wheat straw as a composite material as an engineering material. Glue-impregnated wheat straw can be used to produce low density composites with acceptable mechanical properties.

Thamae and co-authors (2010) using corn/wheat stalk flour and high density polyethylene waste prepared a composition from natural fibers. The authors have shown that through careful selection of processing methods and materials, composites can be made that can be affordable without undue loss of their properties.

A study of the effect of a separate filler – wheat straw and combined fillers – wheat straw and an inorganic filler (heavy calcium carbonate, silicon dioxide and fly ash) on the water absorption properties of hybrid composites based on recycled polypropylene showed that the hybrid system of wheat straw and inorganic fillers demonstrates better water absorption by compared to wheat straw/polypropylene composites without inorganic fillers (Yu *et al.*, 2016).

It should be noted that the use of wheat straw, on the one hand, provides a growing opportunity whenever an improved stability of the material is required, while, on the contrary, it is still a subject for discussion, since the production of composites reinforced with wheat straw only in isolated cases has reached end use, such as in the automotive industry or construction.

3. Conclusion

The use of lignocellulosic fibers has increased over the years, mainly due to environmental factors and low cost. Despite this, the systematization and analysis of the literature data on polymer composites based on lignocellulosic fillers showed that there are still many information and knowledge gaps about composites from agricultural waste that need to be closed in order to stimulate the commercial production of these new materials.

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