Use of different kinds of waste in the construction of new polymer composites: review

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Abstract:	Waste valorization is one of the current research areas that has attracted a great deal of attention over the past few years as a potential alternative to the disposal of a wide range of residues in landfill sites. In particular, the development of environmentally friendly and innovative strategies to process several kinds of waste is an area of increasing importance in our current society. Landfill, incineration and composting are common, mature technologies for waste disposal. However, they are not satisfactory due to the generation of toxic methane gas and bad odors, high energy consumption and slow reaction kinetics. This paper is aimed to summarize recent development of waste valorization strategies in polymeric matrices for the sustainable production of new composite materials (polymer matrix composites, PCMs). The incorporation of residua into polymeric composites can represent a new resource and a successful approach to creating materials suitable for specific applications promoting a circular economy approach.
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Firenze 15th October 2022

Dear Editor,

I have the honor to submit You the work titled: "**Use of different kind of waste in the construction of new polymer composites: Review"** by Taurino R. et al., that I would like You to consider for publication on the "Materials Today Sustainability.

This Review is aimed to summarize recent development of waste valorization strategies in polymeric matrices for the sustainable production of new composite materials (polymer matrix composites, PCMs). Infact, the increase of tighter regulations regarding waste, and the demand for renewable chemicals and fuels, are pushing the manufacturing industry toward higher sustainability to improve cost-effectiveness and meet customers' demand. Then, the incorporation of residua into polymeric composites can represent a new resource and a successful approach to creating materials suitable for specific applications promoting a circular economy approach. In particular, in this review paper, the possibility of reusing both agro and industrial waste in polymer composite applications are discussed taking into account their mechanical properties and environmental impact. For all of these aspects, in the authors' opinion this manuscript should will be suitable for academia and industries to gain an insight into the several wastes, which can potentially be used in developing novel polymeric composites. Then, this manuscript should receive consideration for publication in the "Materials Today Sustainability.".

We confirm that the present manuscript has never been published previously.

Thank You for Your attention.

Sincerely yours,

PhD. Rosa Taurino

Rosa Taurino

Highlights

Sustainable production of new composite materials (polymer matrix composites, PCMs).

Organic fillers treatment methods to improve the surface interaction.

Stakeholders can adopt waste-based composites to spread the concept of sustainability.

Locally sourced recycled material as filler can have a positive environmental impact.

Use of different kind of waste in the construction of new polymer composites: Review

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Abstract

Waste valorization is one of the current research areas that has attracted a great deal of attention over the past few years as a potential alternative to the disposal of a wide range of residues in landfill sites. In particular, the development of environmentally friendly and innovative strategies to process several kinds of waste is an area of increasing importance in our current society. Landfill, incineration and composting are common, mature technologies for waste disposal. However, they are not satisfactory due to the generation of toxic methane gas and bad odors, high energy consumption and slow reaction kinetics.

This paper is aimed to summarize recent development of waste valorization strategies in polymeric matrices for the sustainable production of new composite materials (polymer matrix composites, PCMs). The incorporation of residua into polymeric composites can represent a new resource and a successful approach to creating materials suitable for specific applications promoting a circular economy approach.

Keywords: solid waste; polymer matrix composite; waste recycling; mechanical properties; thermosetting resin, thermoplastic resin.

1. Introduction

In recent years, the increase of tighter regulations regarding waste, and the demand for renewable chemicals and fuels, are pushing the manufacturing industry toward higher sustainability to improve cost-effectiveness and meet customers’ demand.

The disposal of waste to landfills and the limited availability of natural resources lead to an increased interest in searching for innovative alternatives that can contribute to a circular economy.

According to ISWA data, currently around 4 billion tons of waste are produced in the world every year and this volume is expected to increase by the year 2025 [1]. Around half of these ones are municipal wastes while the residue amount arises from industrial and production activities with continued growth in both developed and developing Countries due to population, living standard and urbanization growth [2]. In particular, even if many kinds of waste are produced worldwide, special attention is paid to materials that are not biodegradable [3,4], or to residua that, even being biodegradable, are produced in great quantities and can cause high environmental impacts, including climate-harming emissions by illegal dumping, such as crop wastes, eggshells, crustacean shells and others [5]. In this perspective, the productive use of wastes represents a possible way of reducing some of the problems associated with their management, reducing the use of natural resources and in some cases resulting in the production of environmentally friendly products. In this regard several research groups are working on waste valorization and one of the main used approach is to recycle wastes, in particular solid waste, by mixing with polymeric matrices to obtain new composites materials. Composites materials are widely used in building, furniture, electric appliances, cars, and other fields, in particular they offer a broad potential to be used in furniture parts and structures. The properties of a composite material depend on the details of its structure and adhesion between the fillers and matrix. Moreover, the filler particle morphology, size and resin filler compositional ratio have a critical influence on the mechanical properties of composites [6] and can have a significant impact on the visual look (Fig. 1) of the product. Infact, both the chemical composition and the proportion of fillers with respect to the resin will affect directly the color of the final product, as well as the purity of the fillers it is essential to avoid the presence of polluting particles that can generate black dots on the composites surface.

For this reason, composites can play a key role in the interior building sector due to many design opportunities and material characteristics: material strength, the defined anisotropic/isotropic mechanical characteristics and lightweight allows for freedom of design and long yet thin overhanging structures as well as convertible parts. Then, it is important to stress that the use of solid wastes as fillers can have important impact on the process stability, look of product as well as final product odor. Infact, masking unpleasant odors originating from recycled raw materials is not always possible consequently, the end uses of the composite product become restricted. The odor is dependent on the material composition, contamination, and processing temperatures. In some cases, the effect of the composite manufacturing process can reduce the concentration of volatile organic compounds (VOCs) [7]. In this review article, different individual solid wastes derived from natural and industrial sources, including their modifications, resultant properties, and polymer composites' applications, are thoroughly reviewed and discussed. In Table 1 and 2 are reported the polymer matrix generally used in the preparation of composites, and the main physical and mechanical properties of thermoplastics and thermosets used as matrix in PCMs preparation [8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20]. As discussed elsewhere [21], material preparation methods involve in general the crushing and milling of recycled fractions into smaller particles (see the example in Fig. 4); further processing on recyclates depends on the type of material. A possible process for manufacturing composites containing waste is presented in Fig. 2.

Then, it is expected that this review will be suitable for academia and industries to gain an insight into the several wastes, which can potentially be used in developing novel polymeric composites. Furthermore, different stakeholders can adopt waste-based composites that will propagate the concept of sustainability and circular economy.

2. Solid waste

Solid wastes can be divided into two groups, agro and industrial wastes. Industrial waste, such as paper, glass, metal, textile and plastic, constitute a major part of the municipal solid wastes originated from household, schools, hospitals and business activities. About 7–10 billion tons of urban wastes are generated every year globally, in which industrial wastes account for 21% of the total [22]. In addition to the industrial waste, a massive amount of agro-waste generated during manufacturing processes is commonly disposed to landfills and its decomposition process takes a significant amount of time. Infact, referring exclusively to agro-industrial wastes-included in the "biomass" definition (European Directive 2009/28/EC), the production in Europe is almost 250 tons per year, of which 12 million tons in Italy alone. Only a small part is recovered, while the remaining quantity causes management problems: seasonal increase of organic material in separate collection, demand for new landfills, illegal dumping, microbiological instability, production of toxic compounds (land pollution), etc. In Tables 3 is reported the estimated global solid waste composition [23]. Agricultural wastes such as husks, shells, powders, grape marcs, stalks or other living organism shells can be defined as organic by-products. In Table 4 the major components of various lignocellulosic wastes. The organic waste fillers are biodegradable making them innocuous to the environment in contrast to industrial waste that are relatively more difficult to be biodegraded.

In this review paper, the possibility of reusing both agro and industrial waste in polymer composite applications are discussed taking into account their mechanical properties and environmental impact.

2.1 Agro waste valorization

In the last 15 years there has been growing research interest in agricultural residues as alternative fillers for the manufacturing of composites due to not only the increasing environmental awareness but also to their specific properties, health advantages, and recyclability [30].

Natural filler composites are indicated for application in components subjected from light to moderate loadings. Typical applications include civil construction, furniture, packing, and mainly the automotive industry [31, 32] where natural fillers were used in association with both thermoplastic and thermoset matrix to substitute fillers or fiberglass composites [33, 34]. In particular, the insertion of natural fillers in the industrial, building, and commercial market sectors has experienced a growth rate of 13% over the last 10 years with an annual use of approximately 275 million kilograms [35]. Due to their large availability throughout the world and low cost, sisal, flax, jute, coconut, and ramie are the most used reinforcing fillers. Other alternative fillers for the manufacturing of composites can be the agricultural residues such as wheat cereal straws [36], rice husk [37], bagasse [38] and kenaf [39], to mention a few examples. Jiewei Tong et al. [40], fabricated transparent composites for light-transmitting building applications using wheat straw fibers (WSF), agricultural residues produced in huge amounts around the world. Infact, wheat (Triticum aestivum L.) is a cereal crop with a cultivated area of 214 Mha and a yield of 734 Mt (FAO 218) [41]. The obtained transparent composites were successfully prepared by impregnating pre-polymerized methyl methacrylate (MMA) to WSF. The prepared bio-based composites had high light transmittance and mechanical properties similar to wood fiber transparent composites. The authors showed that the transparent composites using wheat straw (TCWS) with a thickness of 3 mm and 30 wt% WSF had a light transmittance of 74.63 %, haze of 54.87 %, tensile strength of 58.19 MPa and impact strength of 4.26 kJ/m^2. Moreover, thermal property test showed that the heat conductivity of TCWS was 0.07 $Wm^{-1}k^{-1}$, displaying excellent thermal insulation performance, excellent thermal dimensional stability and UV resistance. These results showed the possibility to reuse wheat straw fibers to obtain new composites with potential application prospects in the field of transparent buildings. However, even if wheat straw has similar chemical constituents to wood (i.e. cellulose, hemicellulose, lignin, and extractives), the comparatively poor surface properties of wheat straw with hydrophobic components can reduce the interfacial bond quality between the substrate and polymer binder [42,43]. To overcome this problem, MehdiChougan et al. [44] used on wheat straw hot water, steam and microwave, to mitigate the surface quality deficiencies for an intimate interfacial bond between substrate and polylactic acid matrix. With this procedure, the tensile strength, elongation at break, elastic modulus, and toughness increased by 166 %, 18 %, 68 %, and 285 %, respectively if compared to untreated wheat straw bio-based composites. Being one of the main problem in the use of agro waste, several treatment processes are performed to enhance the interfacial bond quality between fillers and polymer matrix. For example, rice straw (RS) is an other type of agricultural waste that can be used as substitute of wood-based raw materials [45]. RS is less homogeneous than wood and contain a relatively large amount of vessel elements (i.e. sclerenchyma fibers, epidermal cells and parenchyma cells) as compared to wood material [46], which may influence the interaction between the rice straw and polymer matrix. Jiun Hor Low et al. [47] explored the fabrication of pristine rice straw based high-density polyethylene (HDPE) bio-composites (without using any coupling agent) by using the injection molding technique. To investigate the effect of rice straw particle size on the bio-composites, the rice straw and HDPE (30:70 by weight) as well as the dispersing agent lubricant (Ultra-Plast TP01) and binding agent (Ultra-Plast TP10) were mixed in a high-speed mixer and then melt compounded using a co-rotating and intermeshing twin screw extruder at temperature of 180–190 °C. Even if the the fabricated bio-composites achieved the highest tensile strength at the particle size of 250 – 300 μm, the incompatibility between the hydrophilic rice straw and hydrophobic polymer chain leads the final mechanical properties of composites by proving that the it is necessary to perform agro waste treatment to improve the bondability of lignocellulosic materials in both particle and fiber form with synthetic resins. Then, to improve the surface interaction, Rahman et at. [48] reported the use of vinyltrimethoxysilane as coupling agent between rice straw and HDPE. In this work, vinyltrimethoxysilane (VTMO) as crosslinking agent, dicumylperoxide (DCP) as the initiator, dibutyltin dilaurate (DBTL) as the condensation catalyst were used. After drying at 70°C for 24 h, RS, ground and sieved, was mixed with HDPE and then compounded with a co-rotating and intermeshing twin screw extruder. This formulation was designed and optimized to produce a new injection grade crosslinked rice straw /HDPE biocomposite. As well as in thermoplastic matrix composites, agro waste fillers are often used for composites made from thermosetting resin. [49]. In general, thermoset resins unsaturated polyester resin (UPR) and epoxy resin are the most commonly used with those natural fillers to produce low-strength materials. In particular, unsaturated polyester resins (UPRs) are the most common in preparing composites, comprising 80% excess of all thermoset resins [50]. Though they provide excellent properties such as chemical and weather resistance with relatively low cost, in general, the mechanical properties are inadequate for many structural purposes without the addition of the appropriate fillers as load carrying constituents of composites. UPRs having good dimensional stability, easy manufacturing and limited moisture absorption are the most commonly resin used with those natural fillers to produce low-strength materials. For example, Taurino et al. investigated the possibility to use agro-residues of the vinification process as cheaper and eco-friendly filler to prepare unsaturated polyester composites for interior design building sector. Millions of tons of agro-waste are produced per year by the wine industry, in many European Countries, such as Italy, France, Spain, Germany and Portugal. Infact, in Italy, the annual potential production of wine residues is about 0.5 T as dry matter, more than 60% deriving from grape marks and stalks. Indeed, winery waste can represent an alternative source for the production of composites for interior furniture. For example, experimental unsaturated polyester–based resin composites, reinforced with grape stalks, were prepared by a simple casting technique by Taurino et al.[51]. However, the main disadvantages of using such fillers in the composites are their hygroscopicity and difficulty in achieving acceptable levels of dispersion in the polymer matrix. Uniformity of the filler/matrix surface has a major impact on the load carrying capacity, and thus on the properties of composites. This effect can be achieved using compatibilizers that reduce the tension between phases or modifying the cellulose surface to obtain a less hydrophilic one [52]. For example, the addition of a silane coupling agent was necessary to increase the interface adhesion between resin (UPR) and grape stalks [51], resulting in good mechanical and physical properties of silanized composites that are satisfactory for the production of interior furniture composites. The combination of a thermal treatment and silanzation was perfomed to increase the addition between UPRs and digestates residues from anaerobic digestion (AD). In particular, another study [53] assesed the potential for utilizing digestate residues from anaerobic digestion (AD) process of organic substrates as fillers for thermosetting matrix (UPR resin) to prepare a new solid surface material. Solid surface is a common term applied to a small range of materials, characterized by a distinctive look, with different chemical compositions or thicknesses but sharing many functional similarities. The polymeric resins used for producing solid surface materials are, in general, filled acrylic polymers, filled unsaturated polyester resins (UPRs) and filled acrylic-polyester blend polymers. The composition of a typical solid surface component is characterized by 35% of thermoset resin, and 65% of alumina trihydrate (ATH), by weight. In general solid surface materials are applied in vanity tops, sinks, shower floors [53]. To improve fillers surface adhesion, residual organic components of the digestate material were removed by thermal treatment (calcination at 550°C) and additional silane treatment was made. Here, the tensile strength and Young's Modulus decreased after the compatibilizer addition and an increase in percentage of elongation and energy at maximum load was measured, due to the formation of flexible polysiloxane (Si-O-Si) from self-condensation reaction of silane. In this case, thermoset resin was used as a matrix material with the advantage to work with a very low viscosity material which can be mixed with the agro fillers at low pressures. Infact, thermosets can be processed by simple processing techniques such as hand lay-up and spraying, compression, resin transfer, injection, compression injection, and pressure bag molding operations. By K. John et al., no significant effect on the flexural properties has been observed by silane treatment but a small increase in the mechanical properties has been observed by alkali treatment of sisal/glass hybrid composites [54].

Another thermosetting resin used as a hosting matrix for agro-residues is the epoxy resin. Epoxy resins are materials reserved for the manufacture of elements having to undergo high mechanical and thermal stresses and it is also considered as the most dominant matrix material for lightweight polymer composites. Moreover, several studies showed that epoxy composites reinforced using kenaf, coir, coconut fiber, banana, sisal, jute, flax, vakka, and pineapple leaf have better mechanical properties thus making them attractive in low load carrying applications including furniture application. For instance, ternary composites of rice husks/glass fibre/epoxy composites with better hardness, Young’s modulus, impact strength and wear resistance than the glass fibre/epoxy were studied [55, 56]. The epoxy composites of natural fillers of different materials are used for newer applications. By this way, the α-cellulosic micro filler, which are isolated from Cocos nucifera var Aurantiaca Peduncle (CAP) through chemical treatment process, was systematically utilized by K.J.Nagarajan et al, as a reinforcing material in thermoset epoxy polymers as a replacement of carbon, ceramic fillers and wood derived products. The results on mechanical properties such as tensile, flexural, impact test revealed that the properties of the α-cellulosic micro filler reinforced epoxy composites increased in linear nature for 3 wt% to 15 wt% of filler loading and, 15 wt% shows the superior behavior in their mechanical properties [57]. Since epoxy resins have excellent mechanical and chemical properties, and dimensional stability, they are widely used in lamination to prepare composite panels. Composites laminated for broader applications, such as for the furniture industry for example, based on new agro waste of 4% alkali-treated and untreated *Lagenaria siceraria* fiber (LSF) were prepared [58]. The authors investigated the effect of four different fiber lengths of 3, 5, 7, and 9 mm with five different fiber contents of 15, 20, 25, 30, and 35 wt% on the composites mechanical strength. Compared to neat epoxy resin, the mechanical properties are enhanced for increase in the fiber length up to 7 mm and 30 wt% of untreated and treated fiber; however, further increase of both fiber content and fiber length results in a decrease of the mechanical properties for both treated and untreated LSF-reinforced composites. In the study of Imana Taha et al. [59] tomato stalks-based panels have been prepared. In particular, they analyzed the effect of particle size and weight concentration on the mechanical and physical performance. The tensile strength and tensile modulus are relatively low compared to flax and bagasse based particleboards. The primary reason behind this lowering is the fair cellulose content compared to the other resources. Infact, cellulose is the main component providing strength, stiffness, and structural stability, while the low lignin percentage of tomato stalks facilitated the chopping process of stalks [60]. As suggested by Nemli G. et al. [61] chemical surface treatments such as the acetylation of fibers prior to their incorporation into the resin mix can improve bonding between hydrophobic matrix and hydrophilic fibers to meet the minimum requirement specified by the European standard EN 312:2010 for particleboards. Then, as discussed previously, the main disadvantage of using agro-waste in the composites are their hygroscopicity and difficulty in achieving acceptable levels of dispersion in the polymer matrix. Several approaches to improve the compatibility between resin and organic fillers are summarized in Fig. 3. Physical pretreatment method mainly focused on the size reduction of biomass, therefore, fragmentation methods like cutting, shredding, grinding and milling were employed [62,63]. Chemical pretreatment methods are based on chemical reactions taking place in aqueous solution between lignocellulosic biomass and various chemical compounds. Most common are treatments with acid and alkaline solutions. Acid pretreatment aims at breaking up the lignin structure as well as dissolving hemicellulose and depolymerizing cellulose. It takes place at acid concentrations of 10–30%, elevated pressures and temperatures, and reaches hemicellulose degradation rates of up to 90% [64]. Alkaline pretreatment is based on the saponification process. The presence of ammonia, as well as sodium or calcium hydroxides [65] causes swelling of the biomass. Thereby, the reduction of cellulose crystallinity results in an increase in the specific surface. Then, several physicochemical pretreatment methods combining the usage of chemicals with the application of physical forces were used. For example, Liquid Hot Water, steam explosion, and supercritical water is the simultaneous application of high temperature and pressure in aqueous solution. Lastly, microwave irradiation is a way of heating the biomass, resulting in the disruption of lignocellulosic structures [66]

2.2 Non biodegradable wastes valorization

Non biodegradable wastes, such as fly ash, tyre rubber, glass waste, and waste concrete, are other types of waste commonly found in the environment and proposed as reinforcements or fillers for the preparation of polymeric composites [67,68].

One of the main types is fly ash, a mineral by-product produced by combustion and constituted by metallic oxides, such as SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, K_2O, and many elements, such as Cr, Pb, Ba, Sr, S and Zn. This material, annually produced in large quantities (about 750 million tons for year) [69] is useless and harmful to the environment and human health, but its compatibility with other materials (e.g. polymeric ones). This means that the recovery of fly ash is an efficient way to prevent such danger and to contribute to further waste reduction and set the standard for industrial-scale recycling [70,71]. In the study by Mateusz Gargol et al. [72] utilization of fly ash and silica gel as replacement of standard fillers, was made to increase the thermal resistance, and rigidity of polymeric composites based on polypropylene matrix. Several samples with different amounts of filler and reference material were synthesized in two different ways. The data obtained during the thermal analysis confirmed that the amount of fillers affect the thermal properties of the samples. Samples without fillers were characterized by the worst thermal stability that instead increases with increasing the inorganic filler content. Moreover, the use of fly ash reduces the amount of monomers, decreasing the cost of such materials. Many experimental studies using fly ash (FA) have shown that the presence of filler increases the stiffness of the polymer composite but, like most fillers, reduces the impact resistance. To improve these properties, other components should be added to the composite formulation. Pardo et al. [73,74,75] investigated the deformation and fracture behavior of polypropylene (PP)/ash composites with different ash contents and analyzed the effect of a silane coupling agent too. They found that the incorporation of a silane coupling agent in the formulations led to composites with slightly improved tensile and fracture properties. This could be due to improved interaction between PP and ash or changes in the crystallization behavior of PP. Khan et al. [76] studied the effect of polyethylene-grafted maleic anhydride (PE-g-MAH) as a compatibilizer and showed improvement in modulus and tensile strength properties of low-density polyethylene and waste ash. Sengupta et al. [77] studied a renewable, low-cost chemical-like furfuryl palmitate as an effective coupling agent for FA-reinforced recycled PP matrix composites. The highest enhancement in properties was observed in 2 wt% furfuryl palmitate-coated FA-filled composites. Greater tensile toughness and higher resistance to deformation under higher bending loads can be obtained after the addition of silanized FA in the epoxy-based composites. In particular, in the work of Tanakorn et al. [78] the epoxy composite containing 10 wt% treated FA showed the best mechanical properties with respect to the unmodified composite. Moreover, the transmittance of visible light through all epoxy composites containing treated FA was reduced to 10% suggesting that epoxy composites containing 10 wt% treated FA are suitable for interior light partition applications. Other researchers showed that it is possible to increase the final mechanical properties of fly ash/epoxy composite, by controlling the particle size, and volume fraction of fly ash and fabrication conditions. Viscosity conditions must be optimized by controlling the temperature of the slurry (mixing resin and filler) [79]. The result of optimum fly ash content and particles size to improve the tensile properties was found to be 30 vol.% and particle size lower than 50 μm. A comparative study of the performances of fly ash with epoxy resin reported that a combination of fly ash and epoxy resin can provide higher mechanical strength than epoxy composite containing silica fume [80]. Similarly, epoxy composites containing, as fillers, the waste from the Production of Mineral Insulation Boards (RGI) showed higher flexural strength, better abrasion resistance and better impact resistance. RGI is a waste from the production of insulation boards made of mineral wool and containing a high proportion of recycled glass (>80%). This is the dry by-product of the production process that falls off from below a pulper in front of a hardening chamber so it does not contain any organic elements, and it is, therefore, possible to classify this as recycled glass without organics.

In view of the high mechanical resistance of glass and also because of its pleasant aesthetics, glass waste has been used in epoxy-related applications, such as countertops, and replacement for standard grit in friction floors [81]. Even if the glass waste is one of the most recyclable material in the world, to the best knowledge of the authors, there were only a few studies conducted on the potential valorization of glass waste in polymer matrix composites PCMs. Glass waste can be classified in two main categories: glass cullet, that in general after the collection can be reused in glassworks, and glass fines (mixtures of flat glass, Pyrex, ceramics etc difficult to reuse). One of the common applications of fine glass is to replace natural coarse and fine aggregates in concrete, in asphalt paving mixes and then in various construction applications. In the polymer matrix, glass fine powders could possibly be used as a filler, where the hardness does not disturb the processing or can be used in fiber form. For example, funnel and panel fibers obtained from the recycling of cathode ray tube, were used as reinforcement for thermoplastic resin (polypropylene, PP). Panel and funnel glass fibers/PP composites provided good mechanical and physical properties similar to the E glass fibers/PP composite. A clear disadvantage of PP matrix material for composite applications is its apolar character with respect to glass waste, which leads to limited wettability and poor fiber–matrix adhesion. Matrix modification with maleic-anhydride (MA-PP) was used to increase the thermal and mechanical properties (Izod impact properties, HDT, tensile strength and modulus) [82]. The need to identify new applications of glass wastes led Taurino et al. to the development of a technology that uses fluorescent glass waste as raw material in PCMs with a composition typical of a solid surface. The polymeric resin used for producing solid surface materials is filled unsaturated polyester (35% of thermoset resin, and 65% of alumina trihydrate (ATH), by weight). In this study ATH fillers were replaced by glass waste in coarse morphology and finely ground. In this case as well, a silane coupling agent (vinyltriethoxysilane) was added to the resin in order to increase the compatibility between the two components (fillers-resin). As expected, the presence of coarse glass particles decreases the ductility of the resin, resulting in lower absorbed energy during impact. Only the processing of a bilayer composite with fine waste glass can give an increase of the impact strength and thus in the maximum load (Wmax) that can be tolerated by the material.

Another kind of solid waste proposed as filler or reinforcement, in order to manufacture composites with thermoplastic, thermosets and rubber matrixes with interesting tensile, electrical, or acoustical properties is the tire rubber [83,84]. Infact, the increase in the number of waste tires has resulted in serious environmental concerns due to their toxicity, flammability, and non-biodegradability [85]. Additionally, discarded tires represent an ideal breeding ground for mosquitoes, vermin, and other disease-carrying insects [86]. The tyre rubber is a material obtained through vulcanization of natural and synthetic rubber blends with the addition of chemical additives such as vulcanising agents, fillers, and extenders [87]. Its use for purposes such as filler in the production of several composites (Table 5) has been established. Furthermore, the composition and the irreversible cross-linking of rubber macromolecules do not make it as easy to recover and process as most thermoplastics. To this end, there is a growing interest in recycling used tires into ground tire rubber (GTR) products and then as filler in host material [88]. This approach allows the recovery and reuse of part of a tire rubber avoiding their disposal in landfills but, as discussed above, the main challenge for composite preparation by using both non-biodegradable and biodegradable wastes is to investigate how properties and development of these materials are influenced by the compatibility between fillers and polymer matrix. In the case of tire rubber when it is blended with polymer matrices (HDPE, PP, SBS, etc.), the fiber-matrix compatibility is expected to be low. To increase the compatibility between both components a pretreatment of the waste is generally carried out. For example, grafting of polymers and compatibilizing agents were used on tire powder to obtain useful materials, improving the interfacial adhesion [89]. In the work of Yagneswaran, S et al. poly acrylic acid (PAA) grafted ground rubber tire (PAA-*g*-GRT) were obtained and used as filler in epoxy composites. The PAA-*g*-GRT/epoxy composite showed higher mechanical properties with an increase of modulus up to 180% as compared with the neat GRT/epoxy composite [90]. Another way to increase the compatibility between components is to increase the interfacial bond by pre treating with acid. For instance, Hernandez et al. [92] introduced a GTR-polypropylene composite material and studied the effect of sulfuric acid treatment on its mechanical and thermal properties. The addition of acid-treated GTR increased the specific surface area by approximately 625% in relation to the untreated GTR, improving the mechanical properties, and in particular the Young's modulus by about 275% with respect to the composite obtained with untreated GTR. Moreover, Araujo-Morera et al. [91] performed a mechanical-chemical treatment of GTR particles which were then used as fillers in Styrene-

Butadiene Rubber (SBR) composites. A combination of cryo-grinding mechanical processes along with acidic treatment was used to modify the surface of GTR and improve its adhesion and compatibility to the host polymer. These methods, however, make the final products more expensive; alternative studies by R.Mujal-Rosas et al showed that significant changes in properties can be obtained when the size of the particle reinforcement changes [84]. In particular, with reference to particle size, when the particles are small (< 200 μm) they adhere better to the matrix (polypropylene, PP) due to their high specific stiffness and to the small size of both pores and cracks. On the other hand, the increase of the real conductivity as well as the permittivity and dielectric loss factor prove that the modification of reinforcement particles size, without any prior pre-treatment with acids, is effective and cheaply. Currently, there are already some studies that explore the potential of additive manufacturing (AM) in the development of 3D printable GTR-reinforced composite materials. Different mixtures of GTR-Acrylonitrile-butadiene-styrene copolymer (ABS) composite material were fabricated, varying the amount of GTR to thoroughly investigate the effects of the rubber additive. The manufacturing process utilized an inexpensive Fused Filament Fabrication (FFF) printer. Experimental modal analysis revealed that, despite the reduction in overall mechanical strength properties, the introduction of GTR into the composite significantly improved the overall vibration isolation characteristics of the 3D printed composite. Furthermore, the density of the composite gradually decreased, producing a lightweight composite material [93] that can be easily applied to multiple industry needs and components across a wide variety of applications.

3. Potential environmental impacts of composites made of waste fillers

One of the main objectives in developing composites from recycled materials is to reduce the carbon footprint of products. The technical properties of composites made from recycled materials have been studied over a long period of time. The production of composites using waste fillers is an ecologically better alternative to production from virgin materials. The environmental benefits depend on the formulation of the composite and the waste processing methods required. Waste materials, which can be used as fillers, must have low maintenance requirements and sufficient strength, which is essential, for example, in the construction industry [94]. A high filler content in composites and the use of locally sourced recycled material can have a positive environmental and social impact in Countries with poor waste management practices [2,95]. Infact, the cost of products manufactured from recycled materials is influenced by several factors, such as transportation costs, landfilling fees, recycling process, volumes, material quality, plant operating costs, and waste taxation [96]. Measures identified for policymakers to influence the cost factors include green taxes, green public procurement and standardization of recycled materials [97]. Potential applications of new composites waste based, are slowly expanding to include even more technically demanding applications, such as compression-molded door panels, molded with stiffening ribs, thickness variations, and molded-in holes [98].

In general, there are no recent studies available to calculate product or material costs for alternative composite fillers in composites. According to studies of Petri Sormunen et al. [98], the impact of composite production ranged from 440 to 510 kg CO2-eq in all scenarios. No differences were expected between the impacts of composite production from plastic, wood, and other waste. The impacts were the same because the expected energy consumption for their pre-treatment is the same. Slightly less electricity was required for the preparation of mineral wool and plasterboard, due to their lower strength compared to the other waste materials. The production of composites has a high climate change mitigation potential, if one takes into account the replacement of conventional products made from virgin plastic. In addition, the European Commissio's investments in the Horizon 2020 projects on waste management contribute to the local economy and make collectors and informal recyclers healthier and safer in the workplace. [99]

4. CONCLUSION

Waste generation has grown rapidly with population growth, creating significant environmental problems such as soil, water and air contamination and also impacting human health. In particular, inadequate solid waste management causes the alteration of ecosystems; this study, therefore, examined recent development in solid waste valorization strategies for the sustainable production of new composite materials. As is evident, there is a considerable amount of research on the application of agricultural waste as fillers in thermoplastic and thermoset matrices to obtain composites suitable for application in components subjected to light to moderate loads. Typical applications include civil construction, furniture, packing, and automotive industry. For example, transparent composites for light-transmitting building applications, laminated composites for wider applications, based on agricultural residues have been produced. Composites reinforced with natural fiber using kenaf, coir, coconut fiber, banana, sisal, jute, flax, vakka, and pineapple leaf have shown improved mechanical properties.

Their surface pre-treatment is necessary to increase the adhesion between matrix and fillers and then the mechanical, surface, thermal and chemical properties of polymer composites. On the other hand, few reports have been found on the use of industrial wastes such as fly ash, tyre rubber waste powder and glass waste as fillers in PMCs. Thus, it is clear that better utilization of agro-wastes to manufacture composites can benefit both its performance properties and the environment.

Furthermore, the utilization of agro-waste could also benefit farmers as an additional income, which can be an important motivating factor in promoting an efficient collection and management.

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Table 1. Some physical and mechanical properties of thermoplastics matrix materials

Thermoplastics	**Density (g/cm^3)**	**Tensile modulus (GPa)**	**Tensile strenght (MPa)**	**Melting Temperature (°C)**
Polypropylene (PP)	0.90-0.91	1,1-1.6	20-40	175
Polyethylene (PE)	0.91-0.95	0.3-0.5	25-45	115
Polyvinyl chloride (PVC)	1,38	3.0	53	212
Polystirene (PS)	1.04-1.05	2,5-3.5	35-60	240

Table 2. Main physical and mechanical properties of thermosets matrix materials

Thermosets	**Density (g/cm^3)**	**Tensile modulus (GPa)**	**Tensile strength (MPa)**	**Elongation at break (%)**	**Compression strength (MPa)**
Polyester	1.0-1.5	2-5	40-90	1-3	90-250
Epoxy	1.1-1.6	3-6	28-100	1-6	100-200
Vinyl ester	1.2-1.4	3-4	69-86	4-7	86
Phenolic	1.29	3-5	35-62	1-2	210-360

Table 3. Estimated solid waste composition (projection for 2025) [1]

Waste composition	Quantity (%)
Organic	46
Paper	17
Plastic	10
Glass	5
Metal	4
Other	18

Table 4. Components of different lignocellulosic wastes

Composition (%)	**Rice husk** [24	**Peanut hull** [25]	**Hazelnut husk** [26	**Coconut shell** [27]	**Wheat straw fibers** [28]	**Grape stalks** [29]
Wax	2	-	-	-	-	-
Lignin	37	28	35	32	25	17
Holocellulose	46	69	55	-	-	-
α cellulose	-	43	35	-	-	
cellulose	-	-	-	31	39	30
Hemicellulose	14	-	-	31	24	21
Inorganics (ash)	-	-	8	-	-	-
Extractives		-	-	6	-	16

Table 5. Summary of mechanical and physical properties of several ground tyre rubber (GTR) composite materials.

Composite	**Mechanical properties**	**Physical properties**
Polypropylene (PP)/GRT [84]	Decrease of Young modulus, tensile strenght, elongation at break and toughness	Increase of conductivity
Polyvinyl chloride (PVC)/GRT [88]	Increase of stiffness and Young modulus, Decrease of tensile strenght and elongation at break	Increase of noise absorption coefficient
Polyurethane (PU)/GTR [89]	Increase of tensile strenght and elongation at break. Decrease of Young modulus.	Decrease of dynamic heat generation
Epoxy resin/Poly acrylic acid grafted-GRT) [90]	For composites with 10 wt% GRT. Increase in tensile strength,tensile modulus, flexural strength, flexural modulus. Decrease of elongation at break.	-
Styrene-butadiene rubber (SBS)/GRT [91]	Elongation at break and tensile strength increased significantly with the increment of filler content	Positive effect for wet grip
PP/GTR-MA GTR modified with sulfuric acid [92]	Increase of Young’s modulus. Reduction of the elongation at break and tensile strenght	Increase of the crystallization temperature of PP. Decrease in the percent of crystallinity in the composites.
Acrylonitrile-butadiene-styrene copolymer (ABS)/GTR [93]	Decrease of tensile strength and young’s modulus. Reduction in cut-off frequency. Improvement in the overall damping ratio.	Decrease of the overall density

Fig. 1. Extruded wood-plastic composites from recycled raw materials (LUT University and Linnaeus University)

Fig. 2. Manufacturing process of composites from waste fillers.

Figure 3. Overview of organic fillers treatment methods used to improve the mechanical, surface, thermal and chemical properties of polymer composites.

Figure 4. Glass waste converted into coarse aggregates and fine glass powder (part of the photo is taken from [84].

Fig.1

Figure 2

Click here to access/download;Figure;Figure 2.png

Figure 3

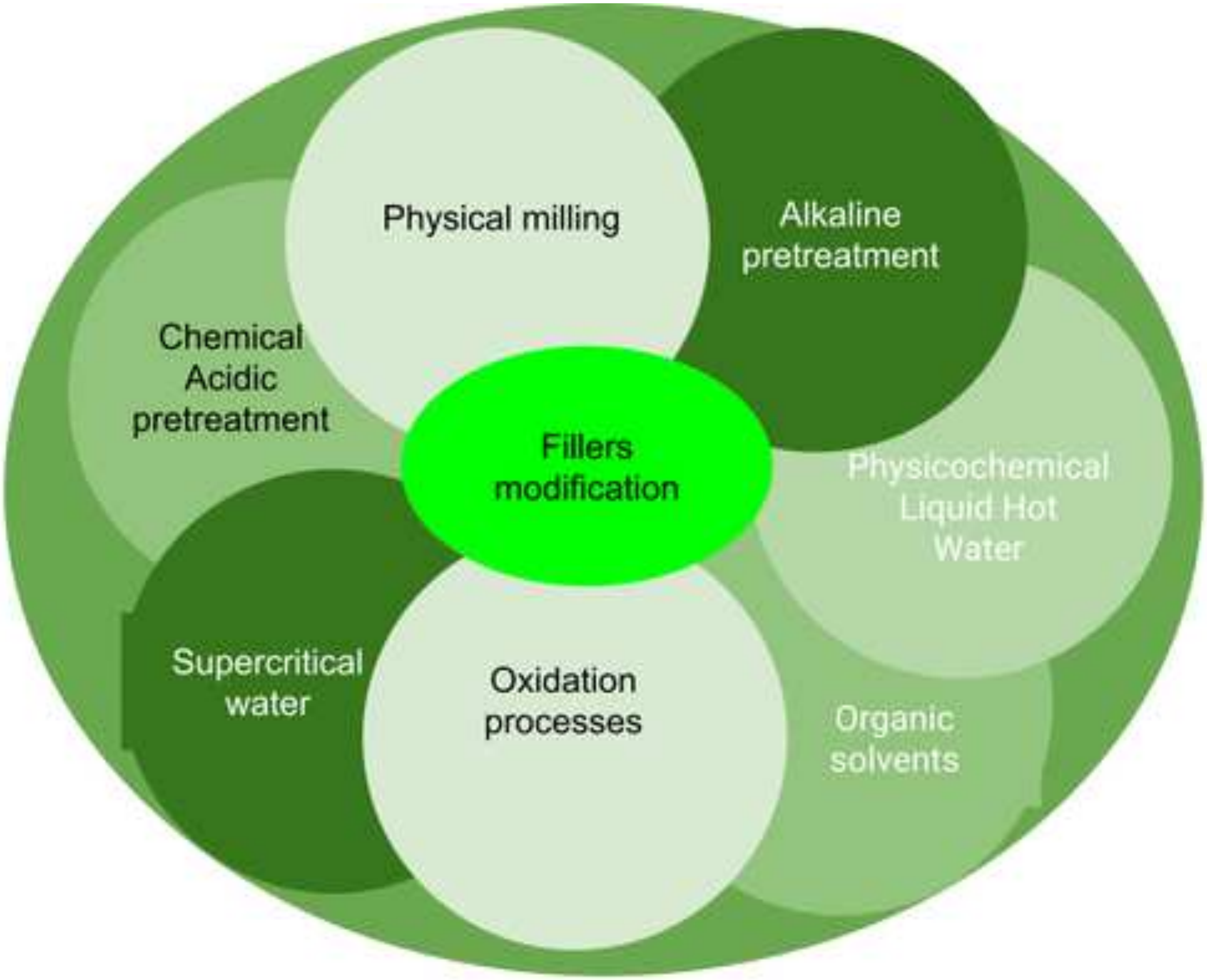
Physical milling
Alkaline pretreatment
Chemical Acidic pretreatment
Fillers modification
Physicochemical Liquid Hot Water
Supercritical water
Oxidation processes
Organic solvents

Figure 4 Click here to access/download;Figure;Figure 4.png

Declaration of interests

☒The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: