Review of the Development of Biodegradable Plastic from Synthetic Polymers and Selected Synthesized Nanoparticle Starches

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Abstract
Plastics which are made from polyethylene, due to their remarkable physical properties are among the most widely used materials in the world today with applications ranging from agriculture to food packaging and domestic uses. However, their remarkable properties are counteracted by the fact that they are not biodegradable and result in water and land pollution which are on a steady increase annually, amongst other forms of pollution. This has consequently resulted in the development of plastics which are biodegradable and at the same time are able to compete with the generic LDPE-based plastics in terms of strength and durability. This review focuses on the various results and progressions made towards developing a biodegradable plastic from a synthetic base polymer such as Low-Density Polyethylene, Polycaprolactone or Polylactic Acid by dispersing synthesized selected nanoparticle starches into the matrix of the polymer.

Key words: Polymers, LDPE, Starch, Nanoparticles, Biodegradability

1 Introduction
Polymers range from familiar synthetic plastics such as polystyrene, Polyethylene, polypropylene and acrylics to natural biopolymers such as DNA, Polysaccharides - cellulose, starch, glycogen and polypeptides in proteins – silk, keratin, and collagen [1,2]. Synthetic polymers are produced by chemical reactions known as polymerization in which individual molecules (monomers) join to form larger units. Plastics are basically just like organic polymers but with higher molecular mass [3,4]. Natural polymers have much shorter polymer chains compared to synthetic polymers. It’s the length of the chains in the synthetic polymers which give them their desirable and unique structures which complement their strength, flexibility, and lightness. Starch is a natural polymer based on glucose found in plants. It is produced by most green plants as carbohydrate reserve and energy storage. Starch belongs to a class of natural biopolymers called polysaccharides, which is the most dominant and abundant biopolymer found in the earth. Other polysaccharides include cellulose, glycogen, and chitin.

Starch is certainly one of the most versatile materials for potential use in polymer technology. It can be converted, on the one hand, into chemicals like ethanol, acetone, and organic acids; used in the production of synthetic polymers and, on the other hand, it can produce biopolymer through fermentative processes or be hydrolysed and employed as a monomer or oligomer.
Finally, it can be grafted with a variety of reagents to produce new polymeric materials, used as such or as fillers for other polymers [5].

Over the past decade, man has manufactured more plastic than we did in the century before that. Plastics production worldwide increased from 322 million tonnes in 2015 to 335 million tonnes in 2016 alone [6,7]. As the need for plastic materials steadily climbs, so does the compulsion of discovering more eco-friendly alternatives to orthodox polymers produced from petroleum. The research for more environmentally friendly alternatives has prompted scientists to develop bio-based or biodegradable polymers which are capable of degradation in certain controlled environments [8–10].

2 Review on Biodegradable Polymers from Synthetic Polymers and Natural Polymers

Biodegradable polymers widen the scope of waste control and treatment options over conventional fossil-fuel polymers, and this is corroborated by Life Cycle Assessment (LCA). The LCA of synthetic polymers such as polyethylene terephthalate (PET) and LDPE shows that disposal and recycling systems or technologies capable of eliminating or drastically reducing the environmental impact of synthetic polymers are yet to be created [11, 12]. The favoured end-of-life disposal choices for biodegradable polymers are residential and metropolitan composting. These biodegradable polymers offer substantial contributions towards material retrieval, a decrease of landfill and usage of renewable resources [13, 14].

Polyethylene and other synthetic polymers, because of their extensive variety of uses, go into open spaces ensuing their valuable life and thus aggregating in our surroundings. The inability of synthetic polymers to decompose in nature opposes disintegration and degradation making a genuine environmental issue. However, starch, a natural polymer is a well sought-after and a potential competitor to put an end to the ecological worries from the usage of non-biodegradable and harmful feedstock in modern applications which have created an expanded enthusiasm for the need to investigate and create biomaterials [15, 16]. The eco-friendliness and simplicity of compound adjustment, for example, gelatinization and mixing of starch with different materials have created attractive properties suited to particular utilizations and applications. Properties of starch have also found use as disintegrative binders, dispersants and lubricants in the medical industry due to its specific properties such as bindability and gel-ability [17, 18]. These properties of starch have been made use by different researchers in blending starch with synthetic polymers to cause disintegration of the copolymers leading to a level degradation. Arvanitoyannis et al. [19] conducted an analysis of the use of biodegradable films made from LDPE/rice or potato starch for food packaging purposes. The water vapour transmission rate, mechanical, and gas permeability attributes were estimated and linked to their morphology. Higher starch levels increased fragility and crumbliness that led to the diminished rigidity, modulus and higher gas permeability; leading to the degradability of the specimens. A considerable dip in mechanical qualities, tensile and flexural modulus and also percentage elongation was noticed. In summary, the gas permeability and water vapour transmission were enhanced in correspondence to starch levels in the blend and the biodegradation frequency of the blends was bolstered as the starch levels surpassed 10% of the total mass [19].
In another research on the biodegradation of LDPE, which was aimed at quickening the biodegradation process of low-density polyethylene by mixing it with starch and subjecting the produced blends to ultra-violet radiation and then treatment by submersion [20]. It was observed from the results that the tensile strength of the samples subjected to UV radiation and submerged in soil was diminished by 20 % to 66 %, while there was a reduction of about 10 % to 26 % in ductile strength of samples which were only submerged in soil, the percentage reductions being from the original amount of starch contained in the sample blend. It was concluded that there was a symbiotic influence on the LDPE/starch biodegradation activity when the sample blends were subjected to UV-radiation and then subjected to soil burial action, thus leading to the oxidization of the blends and making them more vulnerable to biotic reactions, therefore accelerating the biodegradation rate of LDPE/starch blends [20].

The ratio of LDPE to starch in a biopolymer blend also influences the frequency at which degradation of the polymer occurs. This was proven in a study by Abioye et al. The sample blends produced each had different starch content to LDPE content ratios and the degradation of each sample after burial in sandy-loam soil at a specific depth of 4 inches for a period of 28 days was analysed by measuring and comparing the degradation rate and ratio of the samples. The degradation ratio was simply calculated as the percentage of the ratio of weight loss to initial weight, while the degradation rate (Dt) was calculated as the degradation ratio per day. Six sample blends were produced, and they had the following percentage weight of cassava starch (CaS); 5 wt.%, 10 wt.%, 15 wt.%, 20 wt.%, 40 wt.%, and 50 wt.%. The degradation ratio of all the samples increased overtime although at an inconsistent rate, while the degradation rate (Dt) for the samples waivered, increasing and decreasing after each measurement. Although, only the sample with 20 wt.% CaS showed a consistent increase in biodegradation ratio and the most consistent change in degradation (Dt) amongst other samples. Thus, it can be concluded that the optimal ratio regarding the degradability of biopolymer in sandy-loam soil is 80 wt.% LDPE blended with 20 wt.% cassava starch. The authors also conclude from the biodegradation results that there is a linear relationship between the biodegradability of a biopolymer blend of starch and low-density polyethylene and the percentage weight of starch in said biopolymer blend [7].

The influence of starch on the mechanical properties and degradability of LDPE were analysed by Datta and Halder. As usual, different samples with various ratios of LDPE/corn starch were synthesized and each of their performances compared with that of pure LDPE without any additive. The samples were synthesized utilizing a crosshead extruder with a screw diameter of 42 millimetres. It was obtained from the analysis that the film with a composition ratio of 60 wt.% LDPE, 30 wt.% starch, and 10 wt% additive exhibited the best properties for packaging purposes with 16.2 MPa ultimate ductile strength and 140 % elongation at break when compared with unaltered LDPE. Soil burial test for a period of 4 months to examine the degree of biodegradability of the blends indicated a 10 % reduction in ductile strength and 32 % elongation of the film for 20 % starch level, and for 35 % starch content there was 46 % reduction in ductile strength and 45 % elongation, also the scanning electron microscope results depicted the increase in isometric granular shaped corn starch in the polymer matrix, thus bolstering the hygroscopic nature of the film. Basically, the integration of the bio-filler (corn starch) into the matrix of LDPE increases the degradability of the LDPE film. It was pointed out that the use of corn starch nanoparticles as the bio-filler would have bolstered the quality of the blend in reference to its mechanical properties and thus provided them with better results [21].
Recent trends in this area of research made us of bio-fillers (the biodegradable component of the polymer blend) which have been converted to nanoparticles or nanocrystals. This is because they infuse themselves into the polymer matrix and mix better compared to when they are used in standard phase, thus producing better quality blends with enhanced mechanical properties. Organic nanocrystals, such as cellulose nanocrystals (CNCs) have been generally considered as reinforcement segment in biodegradable polymers because of their biodegradability, easy accessibility, very crystalline form, and capacity to enhance the qualities of bio-composites. The thermal, rheological, mechanical, biodegradation and barrier attributes of the polymer are enhanced following the inclusion of CNCs [22, 23, 24, 25].

In another research by Gray et al., CNC was added to TPS firstly to ensure proper dispersion and then the fortified TPS were mixed with LDPE at different loading levels, and it was observed that all mechanical attributes, comprising ductility and hardness, and modulus of elasticity were significantly upgraded by CNC. Glass transition temperature and melting point of the nanocomposites was also improved considerably when compared to CNC free nanocomposites. Also, the barrier strength of the nanocomposite of LDPE and TPS was bolstered because water absorption showed substantial decrease upon addition of the CNC. Finally, the researchers came up with a value of 1% CNC as the optimal amount required to be added to the blend for the highest improvement in strength and barrier performance [26].

It is quite obvious from this review, that different kinds of starches can be used to produce a partially degradable copolymer from a synthetic polymer. However, some starches offer superior properties to others for producing such bio-composite blends and this was observed according to [24] research. The research examined the combination of various starches, namely native, adipate, acetylated and cassava starch, into the matrix of low-density polyethylene (LDPE) to obtain partially biodegradable product by means of a high shearing mixer for the mixing of the LDPE and the starches to guarantee the homogeneity of the blends. The sample blends were characterized by using mechanical and structural analysis. From the mechanical analysis; the tensile tests indicated that the blends comprising of cassava starch displayed slight changes, in terms of absolute figures, for mechanical quantities. Although taking into account the average deviation of all the samples, it is impossible to establish with sufficient accuracy what kind of starch had sounder operation. It was noted from flexural tests that cassava starch exhibited finer outcomes than any other kind of starch compared to the other starches used. From the structural and morphological analysis, it was observed that for native starch, the LDPE/starch interface was not homogenous because of the contradicting properties of the constituents (hydrophobic LDPE versus hydrophilic starch), while the acetylated starch (RD125) displayed a further homogeneous interface and enhanced interfacing amongst LDPE and the reformed starch particles. Also, the blends from adipate starch presented improved interfacial attraction forces when contrasted with native and acetylated starch, nevertheless the blends comprising of cassava starch had the finest values to the qualities indicating the best interface. Ultimately, from mechanical, morphological, and structural analysis and SEM micrographs, it can be concluded that blends comprising of adipate and cassava starches offered sounder outcomes for biodegradation and mechanical tests. For this cause, they are more designated to usage with LDPE [27].
It could be deduced from this review that biodegradation of polymers blends has been studied by numerous researchers by means of soil burial. However, there are different means to simulate and study biodegradation in polymers. Such as burial in compost, elongated submerging in sea water, UV exposure to simulate sunlight or controlled actions of specific microorganisms. For example, *Stenotrophomonas pavanii* was used in this photo-biodegradation investigation of LDPE films altered with food grade dye-sensitized Titania nanoparticles (TNPs) and starch blends. *Stenotrophomonas pavanii* is a bacterium which is screened from sugar cane stems, and they belong to the genus of *Stenotrophomonas*. The study recommends the suitability of *Stenotrophomonas pavanii* (CC18) as a catalyst for quickened decomposition. The investigation by Mehmood *et al.* revealed that the quickened photocatalytic degradation of LDPE films, utilizing colour sharpened Titania nanoparticles, created a substantial amount of carbonyl gathers which bolsters biodegradation. Considering evolution results on polyethylene exterior, hydrophobicity, metabolic action and Fourier-transform infrared spectroscopy analysis information, it was observed that *Stenotrophomonas pavanii* (CC18) is more effective compared to other microscopic organisms. The outcomes convincingly indicated the potential use of LDPE/Titania-starch blend for making of shopping bags which can easily disintegrate in open surroundings after use. Furthermore, it could also be inferred that the studied methodology renders LDPE eco-friendlier and more likely to decompose at a quickened rate under suitable conditions [28].

Another genus of microorganisms called *Pseudomonas* was used to study the degree of biodegradability of low-density polyethylene by Kyaw *et al.* The study compared four unique species of *Pseudomonas* microorganisms. Degradation of low-density polyethylene was dictated by the reduction in weight of the samples, morphological deviations, mechanical and spectroscopic changes. The eluted blends subsequent to corruption were examined using gas chromatography combined with mass spectroscopy. The outcomes demonstrated that *Pseudomonas* species degrade LDPE films. Biofilm formed on the surface of the LDPE films after around 40 days of incubation in all the four breeds of *Pseudomonas* species, although for the “*Pseudomonas aeruginosa*” species, the formulation of biofilm was quicker. Hence, due to the reduction of hydrophobicity of the LDPE by the biofilm, the degradation rate is enhanced [29].

LDPE is the most exploited option for creating biodegradable blends which is apparent from the amount of research conducted heretofore. However, other polymers such as polyurethane can be considered for this purpose. Researchers Jašo *et al.* created a mix of polylactic acid (PLA) and thermoplastic polyurethane (TPU) with differed proportion utilising dissolve mixing technique. Tests of unadulterated PLA and TPU that experienced a similar thermal treatment were likewise arranged. A respirometer was utilized to analyse the biodegradation of the mixes with various PLA/TPU proportions and distinctive morphologies. Biodegradation of unadulterated PLA began following 20 days of brooding time and continues quickly, degrading totally following 70 days. While, Polyester urethane begins to decompose in a split second and then, backs off and at last does not show over 30 % mineralization following 70 days. It was discovered that structure has a solid effect on the speed of decomposition of PLA/TPU mixes. Mixes with a co-continuous structure decomposed at a higher preliminary rate than different mixes that had a globular structure. The PLA segment in the mix impacts its definitive level of decomposition [30].
Polypropylene Carbonate (PPC) is a biodegradable and part sustainable polymer synthesized from CO₂ and can be utilized as a base polymer for producing a biodegradable polymer. PPC can serve as an appropriate option to fossil fuel plastics and a conceivable choice for adequately reusing CO₂. In research by Bahramian et al., the attributes of PPC were evaluated in comparison with that of common petroleum-based polymers. It was observed in this study that the inclusion of starch bolstered the frequency of degradation of PPC in the soil after a half year to 50 % and the inclusion of cellulose to the PPC bolstered the biodegradation frequency of PPC in soil by 20 %. It was also established that the mechanical attributes of PPC are equivalent to LDPE and PPC has astounding barrier properties that are significant to extending the timeframe of realistic usability or shelf-life of numerous items. Notwithstanding its practicality and equivalent physical attributes with different plastics in the market, PPC is chemically impervious or resistive to various simulated media [31].

Compatibilization in polymer science is the inclusion of an ingredient called compatibilizers to an incompatible mix of polymers thus enhancing their stability. Polymer blends are normally depicted by bristly, unstable phase structures.Compatibilization is a procedure by which the blend attributes are improved while enhancing union between the segments, decreasing the interfacial tension and steadying the morphology [32]. In a study by Pack et al., it was ascertained that the addition of RDP (resorcinol diphenyl phosphate)-coated starch as a compatibilizer can enhance the miscibility and stability of a PLA/Ecoflex blend thus increasing the tensile attributes like yield strength and impact toughness. It was also discovered that the mixture of RDP-coated starches with nanoclays could make the blends flame retardant [33]. In order to determine the compatibilizer best for polyethylene-TPS blends, Claire Cerclé and her partners studied five different compatibilizers for PE-TPS blends using the emulsification curve in 2012. Fine morphologies (volume diameter = 1.4 μm) were obtained for the polyethylene–maleic anhydride terpolymers (PE-Rx-MA) comprising of elastomeric pieces. The PE-Rx-MA copolymers, when used as compatibilizers, offered extremely elevated elongations at break within the span of 700%, the uppermost values testified for PE/TPS structures. Also, those copolymers inverted the typical trend of compatibilized PE/TPS blends, which display a reduction in impact strength with compatibilizer content, and rather lead to substantially upgraded notched impact resistance [34].

3 Conclusion
It can be concluded from this review that more work is still needed in creating a conspicuous and undoubtable substitute for the fossil-based synthetic polymers which are polluting our planet at a rapidly increasing rate. Technically, the use of starch obtained from staple foods for polymer blends diminishes the quantity of those foods available for human and livestock consumption, which could pose a possible problem in the near future. The use of starch from non-staple sources which are not consumed routinely by human beings and livestock can help avoid this issue. Thus, more attention should be given to finding alternative sources of polysaccharides asides the conventional starch consequently de-stressing their usage duality as they also serve as staple food sources. This would be a bodacious effort towards achieving a suitable biodegradable copolymer substitute for conventional petroleum-based polymers.

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