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Characterization of functional properties of starch based nanobiocomposite films containing montmorillonite and titanium dioxide

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Introduction: Biopolymers are a class of polymer, which are disintegrated by an enzymatic or bio-path and the products disseminated to the surroundings do not induce negative effects. Nowadays, non-degradable polymers are quid pro quo with biodegradable ones particularly in agricultural applications, environmental and food industry use. Starch is an example of natural biopolymers, biocompatible, which is completely biodegradable in environment. It has been considered as one of the best candidates for oil based polymer substitution due to its low cost, availability and processbility. The main disadvantages of starch based polymers are their high hydrophilic nature therefore; they have poor mechanical properties and are permeable to water vapor. However, these aspects could be considerably reclaimed by shuffling it with nanodimension materials such as itanium dioxide (TiO_2) and Montmorillonite (MMT). The main reason for this improvement in comparison with conventional composites is the large surface area of these nanomaterials which results in high interactions between the nanofillers and starch. The functional behaviors of nanocomposite films have been depended to the compatibility and degree of nanoparticles dispersion in the biopolymer matrix. TiO_2 is a 3D nanosphere has been perused widely because it is inexpensive, chemical inert and, has a high refractive index with visible and UV shielding potential. MMT as a 1D, platelet is the most commonly used layered silicates. The investigation of biodegradable films containing two different nanofillers simultaneously has been rarely done. TiO_2 and MMT as two different inorganic nanofillers have different physical and chemical structures, so simultaneously use of TiO_2 and MMT clearly had a new effect on the nanoparticle distribution and functional properties of starch films. The aim of this study was investigate the synergistic or antagonistic effect of combination of TiO_2 nanoparticles and MMT platelets on the functional properties such as surface hydrophobicity, water vapor permeability (WVP), moisture uptake (MU), Water Solubility (WS) and mechanical properties of plasticized starch-MMT-TiO_2 nanocomposites.

Materials and methods: 100 ml of potato starch solution with a concentration of 4% (w/v) was prepared by dispersion of starch in distilled water. It was gelatinized at 80 ºC for 15 min. Different amount of TiO_2 (0.5, 1 and 2% w/w starch) and MMT (3 and 5% w/w starch) were dissolved in distilled water and added to the gelatinized starch after treatment with ultrasound for 30 min. Glycerol with concentration of 50% (w/w starch) was added to the starch-nanofillers filmogenic solution. Bionanocomposite plasticized starch (PS) films were produced by casting and were dried in an oven at 45 °C for 15 hours. The X-Ray diffraction (XRD) measurements were performed for MMT and TiO_2 powder and starch-MMT and –TiO_2 nanocomposite films. The methodology of WVP measurements was based on the ASTM E96-05 (ASTM, 2005). Mechanical properties of the films were determined according to ASTM standard method D882-10 (ASTM, 2010). With some modifications, the methods described by Tunc et al., (2007) and Rhim et al., (2006) were used to determine MU and WS, respectively. Water contact angle (WCA) measurements were performed by the sessile drop procedure. The statistical analyses on a completely randomized design and were carried out using analysis of variance (ANOVA). Duncan's multiple range test ($p < 0.05$) was used to detect differences among the mean values of the functional properties.

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Results and discussion: XRD demonstrated the change of MMT layers dispersion pattern from exfoliation in binary PS-5%MMT films to exfoliation-intercalation in ternary PS-5MMT-TiO_2 films. These results showed that TiO_2 agglomerates are formed in the starch matrix with MMT level more than 3% wt. This could be due to the interaction between starch and MMT tends to be more favorable than TiO_2. Good dispersion of TiO_2, high miscibility of with clay, and continuous phase can be obtained when the content of MMT discs is low. Due to the strong interfacial interaction between the starch and MMT, the tensile strength (TS) increased considerably from4.86 to 5.24 MPa, while the elongation at break (EB) decreased significantly from 78.23 to 71.93%, As the MMT concentration varied from 3 to 5%. The TS of nanocomposite films were further improved after the incorporation of TiO_2. Suitable dispersal of TiO_2, and creation of new interactions in the PS-MMT network, causes to increase the tensile strength of nanocomposites. The TS and EB values of PS-3MMT-1TiO_2 nanocomposite film was higher than that of the other films. This is indicative of a synergistic effect between TiO_2 and MMT which increases the tensile strength and does not decrease the EB. In the PS-5% MMT films, both mechanical characteristics were reduced. WVP shows more evidences of synergistic effect of combination of 1D MMT and 3D TiO_2 on starch films. WVP reduction by MMT has been attributed to tortuous pathway which created by clay layers in the starch matrix. MMT platelets are water vapor impermeable, thus exfoliation of MMT reduce the voids in starch matrix. The PS-3MMT-2TiO_2 nanocomposite showed the lowest WVP as compared to other PS films. WVP was reduced significantly from 5.84×10^{-7} g/m.h.Pa in the PS-3%MMT binary film to 3.04×10^{-7} g/m.h.Pa in the PS-3%MMT-2%TiO_2 ternary film. TiO_2 have low water solubility and hydrophobicity compared with starch and MMT. Thus, significant decrement of WVP in the prophase of TiO_2 connoted that TiO_2 was obstructing the nano- and micro-pathways in the PS films network. With addition of MMT and TiO_2 content the water solubility and moisture absorption were reduced significantly. Results of water contact angle test confirmed the results of moisture absorption, solubility in water and water vapor permeability and showed that the addition of TiO_2 increased the surface hydrophobicity of starch-MMT films as with addition of 2% titanium dioxide in PS-3% MMT and PS-5% MMT films, the contact angle after 60 seconds increased 4 and 15 degree respectively. As a result, 1% wt TiO_2 nanoparticles (FDA maximum allowable) can be regarded as the optimum concentration and the developed starch based nanocomposite films can enable undertaking applications as appropriate candidates in food packaging systems.

Key words: Packaging, Solubility, Hydrophobicity, Mechanical Properties, Permeability.

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