Evaluation of Starch-PE Multilayers: Processing and Properties

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Adhesion tests performed on various plasticized starch-polyethylene multi-layer systems led to the selection of a suitable combination of polymers compatible with the starch-based layer. The compatibility of starch and polyethylene was better achieved through maleic anhydride functionalized polyethylene (PEg) than chemically modifying starch. The PEg method proved efficient provided that the water content, and the plasticizer nature and contents of the starch layer were carefully chosen. Computed shear viscosity allowed us to select a suitable botanical origin of starch such that the interfacial instabilities of the coextrusion process were minimized. The use of a multilayer structure (PE/PEg/starch/PEg/PE) improved gas barrier properties at high relative humidity. The higher quantity of water sorbed by thermoplastic starch (as compared to EVOH) coupled with starch's specific water sorption isotherm lengthened the water equilibration time in the hydrophilic inner layer significantly. As a result the gas barrier properties of the starch based multiplayer systems were enhanced as compared to existing commercial multiplayer systems (PE/PEg/EVOH/PEg/PE). This specific "water-buffering property" of the starch inner layer should prove useful in packaging applications of perishables with extended shelf life in environments of varying relative humidity. **POLYM. ENG. SCI., 45:217–224, 2005. © 2005 Society of Plastics Engineers**

INTRODUCTION

Multilayer systems are used more and more in packaging. In the last two decades, the technology of complex barrier films has undergone considerable development. The association of different materials with complementary properties has led to high performance films, increasing the lifetime of fresh products. These films generally require good oxygen barrier properties. In addition to protection from oxidation, a high permeation to CO_2 is also highly desirable. A high CO_2/O_2 selectivity is therefore necessary to allow fresh food respiration. Multilayer barriers are usually made up of a polar polymer in the internal layer covered by an apolar polymer, the former acting as a gas barrier and the latter as an hydrophobic skin layer that prevents fast water absorption in the internal layer. Water plasticizes polar polymers. An increase in water content leads to an increase of the molecular mobility and a decrease of the barrier properties. Typical structures are made of polyolefin skin layers covering a polar polymer, e.g., polyethylene-vinyl alcohol copolymer (PE-EVOH).

Transport properties of multilayers are mainly governed by the intrinsic barrier properties of each layer, and it is generally considered that the apparent permeability coefficient can be directly deduced from the permeability of each elementary layer [1]. Since outer layers generally have low gas barrier properties, the permeability of a multilayer is mainly governed by the permeability of the inner layer. Since the inner layer's water content is time dependant (the equilibrium being quickly reached only in thin materials), the difficulty consists in being able to predict the change in permeability behavior in a given environment over time.

The originality of this work consists in replacing the classical rather expensive EVOH inner layer material, by a much cheaper material of similar properties. Starch-based materials are good low cost potential substitutes because of their interesting oxygen barrier properties. Plasticized starch, called "thermoplastic starch" (TPS), is obtained by extrusion of native starch in presence of a plasticizer (water, glycerol, sorbitol,. . .) which controls its glass transition [2, 3]. Recently, TPS has been shown to possess good oxygen barrier properties. The behavior of this system is complex due to the simultaneous evolution of α and β relaxation

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