

Comprehensive Experimental Study of a Starch/Polyesteramide Coextrusion

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Received 21 August 2001; accepted 4 February 2002

ABSTRACT: A comprehensive study of the three-layer film coextrusion was performed. Plasticized wheat starch (PWS) was chosen as the film central layer, and poly(ester amide) (PEA) was used as the surface outer layers. Single-screw extruders and a standard feedblock attached to a flat coat-hanger die were used to prepare the three-layer films. The layer deformation and interfacial instability phenomena, inherent to multilayer flows, were thoroughly investigated. The effect of process variables, such as viscosity ratio, extrusion rate, layer thickness, and die geometry, were studied. Encapsulation of the central layer by the skin layers

readily occurred at the edges of coextruded films. The stability of PEA/PWS/PEA coextrusion flows was closely related to the shear stress at the interface. Increasing global volumetric flow rates and the die gap geometry seemed to promote instabilities. Finally, the existence of instabilities at the interface increased the adhesion strength of multilayered products, due to mechanical interlocking between adjacent layers. © 2002 Wiley Periodicals, Inc. *J Appl Polym Sci* 86: 2586–2600, 2002

Key words: rheology; adhesion

INTRODUCTION

The use of starch in nonfood applications has given rise to a large number of studies. As stated by Lörcks¹ in a recent article, starch may be a good alternative for the substitution of common petroleum-based polymers in specific applications, such as packaging. Native starch, once formulated with glycerol and water, can be transformed by traditional processing techniques, such as extrusion and injection molding. Processing and the properties of the resulting plasticized starch materials have been described in the literature.^{2,3} Over the past decade, our laboratory has contributed significantly to the development of materials based on plasticized wheat starch (PWS), which is inline with the concept of sustainable development. For instance, we have studied the properties of PWS products, as a function of the processing conditions, the plasticizer content, and the storing conditions.⁴ Materials ranging from stiff and brittle to soft and ductile were obtained, according to the plasticizer content. Thus, the properties of PWS products could be tuned, allowing fit with desired materials specifications. However, the high moisture sensitivity and rather low performance properties of PWS have constituted strong restrictions to the development of plasticized starch.

Combining plasticized starch with a hydrophobic polymer allows one to strengthen the properties of starch-based products. The choice of that polymer is made on prerequisites such as biodegradability, barrier properties, market availability, and cost. There may be different strategies used to improve plasticized starch properties, such as melt blending, extrusion coating, and coextrusion. As described in previous articles, melt blending PWS together with various biodegradable polymers, such as poly(ϵ -caprolactone),⁵ poly(lactic acid)⁶ (PLA), and poly(ester amide)⁷ (PEA), has resulted in significant improvement of the physical properties of PWS and a marked decrease of the starch moisture sensitivity, even at low polyester concentrations (10 wt%). Among the various blends tested, a good level of compatibility was found between PWS and the aliphatic PEA. The association of PWS with PLA, although promising because both products are from renewable resources, did not yield satisfactory results in terms of compatibility. We showed that a thin polyester skin layer was formed at the surface of the blends during injection molding,⁸ acting as a potential moisture barrier for starch. However, the starch moisture sensitivity was not fully addressed because of the blend phase distribution (the polyester was the minor component) and the low thickness of that outer skin. The case of extrusion coating, coating extruded films at the exit of die, has only been mentioned in the patent literature.⁹ Its applicability with starch was questioned, and the molecular weight and process temperature of polyesters not appropriate for that process. Finally, the realistic development of moisture-resistant starch-based prod-

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Contract grant sponsor: Europol' Agro.