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Delamination onset and design criteria of multilayer flexible packaging under high pressure treatments

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ABSTRACT

Delamination phenomena frequently occur when multi-layer flexible polymeric films are employed for high pressure treatments of food packaging for pasteurization and sterilization purposes, thus potentially limiting the reliability of this treatment technology. The motivation of delamination is however not yet well understood. For this reason, to catch the key factors leading to interlaminar stress fields which can be prodromal to localized delamination events, the issue of mechanical failure of bi-layer structures under high pressure is thoroughly addressed in the present work, by constructing analytical models and Finite Element based numerical simulations. The theoretical results highlight the crucial role played by the mismatch of Young's moduli and Poisson ratios of the laminated film sheets in promoting delamination. These outcomes are in full agreement with experimental findings obtained by performing tests on high pressure-treated food multilayer packages realized coupling different polymeric materials, that is polypropylene-polyethyleneterephthalate, polypropylene-cast polyamide and polypropylene-bioriented polyamide.

Industrial relevance: High pressure processing is a promising food preservation method endowed with minimal quality loss. To allow a safe use of this technique one cannot address the sole issues related to the effect of treatment on food itself, but also suitability of packaging materials to be used for this technology. In this contribution the attention is focused on multilayer flexible packaging with the aim of supplying guidelines for the proper choice of film structures able to withstand the loads associated with high pressure treatments.

1. Introduction

The mechanical behavior of structures made of polymeric multi-layer thin films has been widely investigated in recent literature works, the main attention being focused on the elastic response of the constituent materials, wrinkling, delamination phenomena and interfacial failure (Basu, Bergstreser, Francis, Scriven, & McCormick, 2005; Chen, Eliot Fried, & Tortorelli, 2012; Davidovitch, Schrolla, Vellab, Bediab, & Cerda, 2011; Feng & Farris, 2002; Gomopoulos, Cheng, Efremov, Nealey, & Fytas, 2009; Okabe, Takeda, Yanaka, & Tsukahara, 1999; Pocivavsek et al., 2008). A technological application where these issues are notably relevant is that of Novel Processing (NP) treatments of packaged food, recently introduced to improve safety, quality and shelf-life of foodstuff. In this framework, mechanical performances and structural integrity of multi-layer flexible polymer films used to package food remain a main concern. Among NPs, High Pressure Processing (HPP) is steadily gaining as a food preservation method that also preserves natural sensory and nutritional attributes of food with minimal quality loss (Fairclough & Cont, 2009; Mensitieri, Scherillo, & Iannace, 2013). Packaged foods, processed by using this technique, maintain most of their original texture and nutritional qualities, additionally exhibiting an extended shelf-life. HPP applies high pressure (typically up to 800 MPa over a time frame of the order of minutes) to packaged foodstuffs in order to significantly reduce the number of microorganisms as well as to deactivate enzymes by mechanically-induced mechanisms (Alpas et al., 1999; Alpas, Kalchayanand, Bozoglu, & Ray, 2000; Cheftel, 1992; Deplace & Mertens, 1992; Farkas & Hoover, 2000; Galotto, Ulloa, Hernández, & Fernández-Martín, 2008). High pressure loads are exerted on packaged foodstuff by means of a pressurized confining fluid imprisoned in a vessel. The process consists of a preliminary heating of both confining fluid and packaging, followed by adiabatic pressurization.

Food treatments are aimed at guaranteeing safety and stability of food eliminating or disabling food pathogens as well as food poisoning and food spoilage micro-organisms, inactivating enzymes and suppressing

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chemical reactions. Thermal pasteurization, conducted below 100 °C (reference temperature is 70 °C), enables only pathogen control in low acid foodstuff while also enables control of spoilage vegetative forms, in the case of high acid foods. Thermal sterilization, conducted above 110 °C (reference temperature is 121 °C), acts effectively instead against pathogenic and heat sensitive spoilage bacteria as well as against spores.

High pressure technology delivers a thermal equivalent of pasteurization or sterilization in a shorter process time, ensuring the safety of the product without appreciable changes in color, flavor and texture. In particular, HP pasteurization is a non-thermal treatment in which the food is typically subjected to pressures of 400 to 600 MPa at ambient or cooled temperature for 1 to 15 min. These conditions inactivate vegetative microorganisms, providing safety and prolonged shelf life to chilled or high-acid foods. Bacterial spores, however, are extremely resistant to commercially attainable pressure levels, and therefore low-acid shelf-stable products can be achieved only by combining high pressure with elevated temperatures in the so-called HP sterilization treatment. A typical high-pressure commercial system for sterilization purposes uses initial chamber temperatures between 60 °C and 90°. Pressures above 600 MPa are applied and actual in-process temperatures can reach 90 °C to 130 °C due to adiabatic heating associated to compression of packaged food and pressure transmitting fluid. In fact, since the adiabatic pressurization process determines a monotonic increase of the initial temperature, the actual treatment temperature depends on the maximum attained pressure, with temperature rise of $2 \div 4$ °C per 100 MPa (Ting, Balasubramaniam, & Raghubeer, 2002). Additionally, the suitable choice of a multi-layer packaging for HPP has to be performed in such a way the treatment process does not affect package integrity as well as its functional properties. As a consequence, the packaging material and design should prevent irreversible deformation phenomena induced by high pressure and severe stress regimes (Caner, Hernandez, & Harte, 2004; Caner, Hernandez, & Pascal, 2000). On the other hand, literature highlights that several types of multilayer films, including bi-oriented polyethyleneterephthalate/polypropylene (PET/PP) bilayer films, aluminium foils and metalized layers, exhibit delamination phenomena (Galotto et al., 2008; Morris, Brody Aaron, & Wicker, 2007).

From the mechanical standpoint, HPP can actually kindle interfacial stresses between the different elements of multilayer structures with selected thermo-mechanical properties as a result of the high pressure loading exerted by the pressure transmitting fluid, thus inviting delamination and extensive detachment phenomena. In general, the mechanical analysis of multilayer systems under severe pressures requires modeling incorporating large displacements, intrinsic and deformation-induced film anisotropy, non linear stress–strain relationships, visco-elastic and plastic behaviors, as well as selected constitutive assumptions for adhesives and bonding conditions at the layer interfaces. However, if the role of each of these aspects in the delamination onset is at the beginning still unclear and thus preliminary sensitivity analyses are needed, the combination of the above mentioned types of nonlinearities as well as the strong difference between the characteristic (in-plane) size of the sheets (generally of the order of centimeters) and film thicknesses (of the order of tens of microns) could not recommend the recourse to onerous in silico Finite Element Method (FEM)-based simulations (Takahashi & Shibuya, 1997), that might obscure the actual contribution of the single geometrical and mechanical factors in promoting the failure. For this reason, simpler mechanical models and ad hoc analytical solutions can be helpfully used to gain specific insights and preliminary qualitative information into the key factors causing delamination onset in multilayer structures, this being not a simple task given the limited amount of scientific works dealing with exact solutions to problems involving the mechanical response of thin multilayer structures (Bufler, 1998; Chen & Ding, 2001; Ding & Chen, 1996; Jiang, Young, & Dickinson, 1996; Lebon, Rizzoni, & Ronel-Idrissi, 2004; Vanimisetti Sampath & Narasimhan, 2006).

Therefore, motivated by the already pointed out scientific and practical interest in the mechanical response of polymeric thin films utilized for food packaging and following the line of reasoning drawn above, the present paper explores the main mechanisms governing the delamination phenomena experimentally observed in polypropylene based bilayer films during HPP (i.e. PP/PET, PP/OPA and PP/PA, OPA and PA denoting oriented polyamide and cast polyamide, respectively), performing both analytical and Finite Element (FE) analyses of pouches containing tap water. In particular, after a detailed discussion of materials and methods (Section 2) and on laboratory findings and on the interpretations of the causes for experimentally observed delamination events occurred in some pouch bi-layer structures under HP treatments (Section 3) in which all the clues suggested to trace the rationale of the structural failure in the development of significant inter-laminar shear stress regimes due to mismatch of both Young's moduli and Poisson ratios of the polymeric sheets, two ad hoc analytical solutions at the beginning of Section 4 have been constructed – named problems A and B – to model through two distinct simpler situations of what happened *near* and *far from* the welded sides of bi-layer food packages under high pressure.

In problem A, a plane-strain solution for a bi-layer film has been found to simulate the effects of high pressure and pressure gradients on the stresses developing inside and at the interface between the materials, near the pouch welded regions. The exact solution B is instead constructed for investigating the response of the pouch far from the welded sides, where the bi-layer structure folds, by idealizing the pouch as a bi-layer hollow cylinder filled by uncompressible water and subject to external pressures. A cascade of events related to different load conditions determined by progressive pressure increase has been then examined, starting from the axis-symmetrical response of the envelope due to the sole action of moderately high pressures, finally evaluating high pressure-induced buckling and wrinkling phenomena accompanied by bending moments which perturb the symmetry of the problem and kindle inter-laminar shear stresses at the films interface.

At the end of Section 4, FE numerical simulations have been performed by faithfully modeling the food package geometry and taking into account nonlinear material behavior, large displacements and fluid–structure interaction in the analyses, obtaining stresses and localized stress gradients prodromal to delamination within the polymeric layers with respect to the three different film couplings actually investigated (i.e. PP/PET, PP/OPA and PP/PA), finally showing the very good agreement among experimental evidences (i.e. film deformation and interfacial failure phenomena found in food packages tested under high pressures) and both theoretical outcomes from the analytical solutions and numerical results.

It is worth to highlight that, actually, the PP cast film exhibits three different layers: these layers, however, cannot be separated since the polymeric structure is produced by coextrusion. As a consequence, the stress–strain experimental tests have been performed for PP cast multilayer film, as well as for the other polymeric films involved in the analyses, considering them as single material units, coherently with the theory of homogenization. Furthermore, given that the thickness of all the different adhesives utilized to join the polymeric films is about 2 μm and the micromechanical behavior of the interface goes beyond the focus of the present work, the polymeric structures will be modeled as bi-layer films.

2. Materials and methods

Capability of multilayer polymer films to withstand high pressure pasteurization and sterilization treatments was assessed by realizing pouches containing a food simulant (tap water and small solid carrots) and submitting them to HP treatment similar to those performed on industrial scale. In particular, bilayer films were obtained by laminating