Journal of Food Engineering 157 (2015) 70–76

Contents lists available at ScienceDirect

Journal of Food Engineering

journal homepage: www.elsevier.com/locate/jfoodeng

Development of a new method to predict the maximum spread factor for shear thinning drops

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ARTICLE INFO

Article history:
Received 21 June 2014
Received in revised form 13 February 2015
Accepted 18 February 2015
Available online 25 February 2015

Keywords:
Drop impact
non-Newtonian fluids
Maximum spread factor
Weber number
Effective viscosity

ABSTRACT

Drop impact on a solid surface has obtained attention for a variety of industrial processes, such as application of edible coatings by spray application method. In the present work, drop impact behavior of edible coating formulations (ECF) based on gelatin, glycerol and cellulose nanofiber on banana and eggplant epicarps was studied. Drops of ECF were generated from a syringe pump system-controlled, for images a high speed camera was used and the maximum spread diameter was determined with ImageJ program. The models reported in the literature do not predict experimental data of maximum spread factor, because the EFC behave as non-Newtonian fluid, so it was necessary to modify these models by introducing an apparent viscosity at average shear rate (effective viscosity). The inclusion of an effective viscosity significantly improves the prediction of the evaluated models.

1. Introduction

Edible coatings are obtained from biological materials such as polysaccharides, proteins and lipids. Proteins are good coating formers and have been used for fruits preservation (Andrade et al., 2012b; Lin and Zhao, 2007). Gelatin has good ability for film forming but its mechanical and water vapor barrier properties are relatively poor. Many attempts have been made to improve these properties. For example, George and Siddaramaiah (2012) reported that addition of bacterial cellulose nanocrystals reduced moisture transport in gelatin-based films.

Moreover, the barrier and mechanical properties of coating films is an important characteristic which is used to predict the stability and shelf life of coated food. Edible film and coating can be applied by different techniques such as casting, panning, fluidized bed, dipping and spraying. The casting method, which is normally used to produce films in laboratory, does not render a reproducible structure and morphology of the coating layer (Perfetti et al., 2010). However, spray coating is the most commonly used technique for applying food coatings (Debeaufort and Voilley, 2009; Zhao, 2012). This technique offers as its main advantages uniform coating, thickness control, and the possibility of multilayer applications (Martín-Belloso et al., 2009; Ustunol, 2009).

To obtain high quality of coated products many factors must be taken into account. These include properties of the material to be coated, properties of the material used for coating, temperature and time of contact between food and coating material. In spray coating, once the coating formulations have been sprayed onto the food surface, the spreading of the drop occurs over the surface in order to produce a dried coating film. Thus, knowing the drop behavior during impact, spreading and dry-ability results to be essential in an attempt of coating process optimization. In particular, drops are impacted and may rebound, splash, or deposit cleanly as it is desirable (Andrade et al., 2012a; Bolleddula et al., 2010; Perfetti et al., 2011; Werner et al., 2007). The solid surface influences the flow behavior of a spreading liquid drop via surface energy caused by the molecular structure seen in the top layer of the surface and surface roughness, e.g. if the substrate is roughened, the drop may splash upon impact (Kannan and Sivakumar, 2008). However, for low numbers of weber (<100), wettability has a negligible effect on the maximum spread factor (Bolleddula et al., 2010).

Most drop impact studies have focused on Newtonian fluids; however, many polymer suspensions used as edible coating exhibit non-Newtonian characteristics. Recently, several researchers have investigated the impact dynamics of non-Newtonian fluids. German and Bertola (2009) and An and Lee (2012b) have found

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http://dx.doi.org/10.1016/j.jfoodeng.2015.02.017
0260-8774/

that, under identical impact conditions, the maximum spread diameters of shear-thinning drops were typically much larger than those of Newtonian drops even though both types of drops showed similar impact morphology. During initial spread, the shear rate experienced by the liquid is high and pseudoplastic fluids behaves as a low-apparent-viscosity liquid (Ravi et al., 2013).

Although viscoelastic properties of gelatin gels have been reported by several authors (Michon et al., 1993; Chandra et al., 2013), this property has not been taken into consideration in this work; according to Bertola (2013), polymer additives cause only a slight reduction of the maximum spread diameter; besides, for drops containing flexible polymers impacting on solid surfaces the retraction velocity reduction is due to the drop-surface interaction (surface wettability) rather than an increased energy dissipation related to the elongational viscosity of the fluid.

Previous studies have shown that the relevant dimensionless parameters governing drop impact on a smooth solid surface are Reynolds ($Re = \frac{\rho U_0 D_0}{\mu}$), Weber ($We = \frac{\rho D_0 U_0^2}{\gamma}$) and Ohnesorge ($Oh = \frac{\mu}{\sqrt{\rho \gamma D_0}}$) numbers, where U_0 is initial impact velocity, D_0 initial droplet diameter, ρ fluid density, γ is surface tension for fluid–air interface and μ is dynamic viscosity (Yarin, 2006). Static contact angle (θ) is also a relevant parameter which depends on surface free energy of the solid (Sikalo et al., 2002). However, It has been shown known that the value of the maximum spread factor is only slightly dependent on the wettability of the substrate if the impact Reynolds and Weber numbers are high (Rioboo et al., 2002).

Theoretically, numerically and experimentally, impact phenomena were characterized with a normalized "spread factor", $\xi(t)$, which is the ratio of droplet spread diameter, $D(t)$, on the solid surface to initial drop diameter, D_0, prior to impact, $\xi(t) = D(t)/D_0$. A common interest in the drop impact research field is to predict the maximum spread factor ($\xi_{\max} = D_{\max}/D_0$), which represents the maximum extent to which a drop can be deformed during spreading (Aytouna et al., 2010; Roisman et al., 2002; Ukiwe et al., 2005; Werner et al., 2009).

During the last few decades many theoretical models have been described for the impact and spreading of droplets on various surfaces. Although, there are many different models, all take the energy balance into account, where system energy at impact is set equal to system energy at maximum spreading (Park et al., 2003; Perelaer et al., 2009). Some models, *e.g.* Asai et al. (1993) model, assumption that the drop volume is conserved and the drop spreads into a cylindrical disk of diameter, *D*, and height, *e*. The cylindrical-disk assumption is reasonable except for low impact velocities, particularly for high contact angles where the surface area of the cylindrical disk is much higher than the actual surface area. According to Moon et al. (2014), the Ukiwe and Kwok (2005) model failed as the Weber number increases, a bigger deviation between the prediction and measurement was clearly observed for the non-Newtonian droplet. This result arises because the original Ukiwe and Kwok model, which uses only zero shear viscosity, cannot consider the change in shear viscosity during the spread.

The aim of this study was to evaluate some models for predicting maximum spread factor of edible coating formulations drop impacting on banana and eggplant epicarps. Finally, a new empirical correlation to represent the maximum spread factor of a shear-thinning drop was proposed.

2. Materials and methods

2.1. Plant material

Banana (*Musa sapientum* var. *paradisiaca*) and eggplant (*Solanum melongena* L.) were purchased as fresh fruit from the local market (Santiago, Chile). Banana and eggplant were carefully selected to ensure uniformity in maturity, size, color, and absence of physical damages on their surface according to visual analysis. Before measurements, they were left at room temperature (20 ± 2 °C) and rectangular samples (1.5 cm × 2.5 cm) were cut. Surface free energies of the banana and eggplant epicarps, obtained by acid-base method, are 39.29 and 33.06 mN m^{-1}, respectively (Andrade et al., 2014).

2.2. Materials

Type B gelatin from bovine peel (180 Bloom) was supplied by Rousselot (Rousselot, Brasil), glycerol was supplied by Sigma (Sigma–Aldrich, Chile). Cellulose nanofibers were produced from agroindustrial residues (pineapple peel juice) and *Gluconacetobacter swingsii* sp. as reported by Castro et al. (2011).

2.3. Preparation of coating formulations

Formulations were prepared with distilled water. Gelatin was hydrated at room temperature (20 ± 2 °C) for 30 min, and then heated at 50 °C for 30 min under continuous stirring until completely dissolved. Glycerol and cellulose nanofibers were added at a certain concentration (based on dry gelatin weight). Then, the mixture was sonicated in a bath type sonicator (Branson Model 2210, USA) during 30 min. Gelatin suspensions were prepared at concentrations of 0.6; 1.3 and 2%w/v, glycerol varied between 10 and 20%w/w based on gelatin, and cellulose nanofibers varied between 1 and 5%w/w based on gelatin.

2.4. Physical properties of coating formulations

Physical properties and droplets initial diameter (D_0) of the coating formulations are shown in Table 1. Densities were measured pycnometrically, surface tension by the pendant drop method as reported in Skurtys et al. (2011), and rheological measurements were performed using a LVDV-II Brookfield rotating viscometer (Brookfield Engineering Laboratories, USA) with concentric cylinder geometry (cup to bob radius ratio of 1.172). Apparent viscosity measurements were made on a shear rate range between 0 and 100 s^{-1} (see Fig. 1). The flow behavior of the coating formulations was adequately described by the power-law model. The drop diameters were determined by averaging the drop mass of each coating formulations over 15 drops using a precision balance. All measurements were performed in quintuplicate. Drop diameter was calculated substituting into $D_0 = \sqrt[3]{\frac{6m}{\pi\rho}}$, where m is the average drop mass and ρ is the coating formulation density.

For each solution (e.g. S7 and S10), rheological behavior of the suspensions were modeled using the Cross model. The time constant λ was 0.1 and 0.12 s, respectively. These values are close to those observed for 1% of carboxymethyl cellulose solutions (Benchabane and Bekkour, 2008). It is well known, for a polymeric solution, that if the time constant, λ, increases, the entanglement – disentanglement process with shear rate is longer whereas if $\lambda \rightarrow 0$ the solution is Newtonian. Moreover, the Weissenberg number (*Wi*) was calculated as described in Jung et al. (2013), $Wi = \dot{\varepsilon}\lambda$, where $\dot{\varepsilon} = (1/D_0)\frac{dD}{dt}$. The dimensionless Weissenberg number represents the ratio of the viscoelastic force to the viscous force. For both solutions (S7 and S10) *Wi* was lower than 0.47 for each time process. Therefore, no significantly viscoelastic effect was found on the drop behavior. Furthermore, Bertola (2013) reported that viscoelasticity phenomenon had no effect on the maximum drop diameter.