SHEETING PROCESS MODELLING AND RHEOLOGICAL ANALYSIS OF AN OLIVE-OIL-EMULSION-BASED PUFF PASTRY

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ABSTRACT

foodstuffs, Different like pastries, are commercialised in the form of laminated sheets; sheeting is, in several cases (i.e. puff pastries), the main process step in which fat/dough rheological matching is achieved. Pastries are baked goods produced to complement the flavour of the fillings and to provide them with a casing. Their ingredients include hard fats hydrogenated with catalytic processes. Unfortunately, these processes lead to the formation of trans fats dangerous for consumers' health. Therefore, hydrogenated fats are nowadays replaced by healthy fats, like olive oil, properly structured to be used as solid fat replacers.

The present work deals with the modelling of the sheeting process of olive-oil-based puff pastries; this allows their final height to be predicted. The rheological parameters necessary to set the model were obtained by the analysis of a pastry produced with water-in-olive-oil emulsions, structured with an organogelator agent, and optimised from a rheological point of view.

Keywords: rheology, puff pastry, W/O emulsions, organogels, sheeting, rolling.

1. INTRODUCTION

Rolling or sheeting between co-rotating rolls is used by baking industry as a dough-forming process for a wide range of products, such as cookies, crackers, pizza, bread and pastry (Mitsoulis E. et al 2009). In many operations rolling is well understood: metal sheeting, rolling of polymer, called calendering, but the roller extrusion of food materials, however, is a broad area of industry activity in which current practice still frequently relies on approximations and expensive pilot plant testing (Peck M. C. et al 2006). This is due to the need to have good experimental values of the material properties (a heterogeneous system), but as well as a constitutive equation capable to fit the material behaviour. Being the material composite and showing time dependency the constitutive equations are often proposed in a differential form, thus a simultaneous integration with the conservation principles is needed

The aim of the current study is to model both from rheological and numerical point of view the puffy pastry dough, to create a tool capable to avoid trial and error approach to the determination of the process variables.

Puff pastry is a dough spread with solid fats and repeatedly folded and sheeted. Thus, it consists of alternating layers of dough and solid-like fat, designed to obtain the proper rheological matching. In fact, the characteristic friable texture is due to the vapour released by either butter or dough during cooking ('puffing' process). Pastry fats (margarine) are W/O emulsions structured in the oil phase. They derive their consistency from a fat crystal network of saturated triglycerides (TAGs) (Cavillot et al 2009).

Traditionally, saturated TAGs can be obtained by hydrogenation, which also produces undesired trans fatty acids (TFAs), considered as the main responsible of heart diseases. Because of this, there is a growing market interest on the design of new pastries produced with a low saturated fat content, and, most of all, without trans fatty acids (Simovic et al 2009; de Cindio B. and Lupi F.R., 2011). It should be noticed that saturated fat substitution needs a different rheological fitting capable to maintain the desired consistency. One of the most promising techniques used to achieve the proper rheology of pastry fats, is the oil phase structuration with organogels (Marangoni 2009). In this work, different structured water-in-olive-oil emulsions were produced. Emulsions optimisation was achieved evaluating the effect of the organogelator amount and the water content on the system rheology, in order to reach properties similar to those of a commercial margarine.

Once the proper fat mechanical characteristics were optimised, a puff pastry was obtained with it. A rheological characterisation was done in order to obtain the setting parameters for the puff pastry sheeting process. The rheological and fluidodynamic modelling of the last step of the sheeting process, is necessary to control the final thickness of the pastry exiting from the rolls because the presence of a significant die swell effect, due to the normal stresses developed into the material during process. The velocity and pressure profiles inside the pastry (considered as an homogeneous material) passing thorough the rolls were evaluated according to a modified Upper Converted Maxwell Model (UCM) constitutive equation.

The sheeting process is traditionally modelled by the simplified Gaskell theory (Tanner R. I., 2000), according to which, the elastic effects of viscoelastic materials can be neglected, and therefore the normal stresses acting on the rolls are ignored. In addition it is assumed that the lubrication theory holds, thus a unique component of the velocity vector is considered. On the contrary according to the viscoelastic material behaviour, when sheeting pastries, the normal stresses must be taken into account. In this work a more general model is presented with the insertion of normal stresses and removing also the other simplification usually considered in the Gaskell theory. Thus experimental determination and numerical simulation are considered to predict the final height of a pastry from which final desired organoleptic properties depend. The model was solved by the finite element method by using the commercial software COMSOL Multiphysics 3.5.

2. MATERIALS AND METHODS

2.1. Emulsions ingredients and manufacture The raw materials used for emulsions preparation were: distilled water and a virgin olive oil (De Santis, Italy) as main constituents of the two phases, cocoa butter (Icam S.P.A., Italy), monoglycerides of fatty acids (Myverol 18-04 K, Kerry Group, Ireland) as organogelator, and NaCl (Panreac, Spain), at 0.1 M aqueous concentration, in order to identify the type of emulsion (W/O or O/W) through electric conductivity measurements.

The structured oil phase was prepared by adding contemporary the cocoa butter and the organogelator to the oil at 70°C, under continuous agitation. Afterwards, a fast cooling process to the gently stirred oil phase was performed, quenching it in a thermostatic cold bath. Different emulsions were produced varying the total organogelator concentration and the aqueous phase content. All samples were prepared leaving the cocoa butter/oil ratio constant. The aqueous phase was obtained by dissolving the NaCl in water, at room temperature, and later mixing it to the cold oil phase using a rotor-stator turbine at 7600 rpm for 30 s (Ultra-Turrax T 50, IKA, Germany).

The composition of each sample and its identification is reported in Table 1.

Sample ID	Oil	Water	Cocoa Butter	Myverol
Emu 1	75	12	10	3
Emu 2	68	20	9.1	2.9
Emu 3	74.12	12	9.88	4
Emu 4	76.76	12	10.24	1

Table 1: Emulsions ID and composition [% wt/wt]

Emulsion– stability was also studied, evaluating the evolution of their rheological properties with time (all samples were studied at 24h, 48h, 72h and 1 week form preparation). An emulsions having a similar rheological behaviour, compared to those of a commercial pastry margarine (i.e. "Vallè", France), was used to prepare a hand-made puff pastry. Moreover, as benchmark, a commercial puff pastry (i.e. Cà Bianca, Italy) was also studied in order to compare the differences between its characteristics and those of the hand-made pastry. For the preparation of the puff pastry feed sheets, the dough was keep from the mixed batch immediately after mixing and rolled out with a wooden cylinder on a kitchen board according to the so-called "Danish method" (Harte 2003). After this step the sheet was reduced to a lower thickness by a lab scale sheeter machine.

2.2. Rheological characterisation

The rheological characterisation of the emulsions (the proposed fat substitute) was performed with a Small Amplitude Oscillation Analysis (SAOTs) at 20°C using a controlled-strain rheometer ARES-RFS (TA Instruments, U.S.A.) equipped with serrated plates (\emptyset =50 mm, gap 2 mm). Frequency sweep tests were performed within the linear viscoelastic region, in the frequency range 0.1-10 Hz.

The characteristics of both commercial and hand-made pastry were analysed with a controlled-stress rheometer DSR500 (Rheometric Scientific, USA), using a serrated plate geometry (\emptyset =25 mm, gap 1.5 mm). The viscous flow behaviour of pastries was determined in the shear rate range 0.0001-1 s⁻¹, and stress relaxation tests were performed at 20°C. Owing to the slippage problems, steady shear viscosity was determined only at very low shear rates with creep tests (at 20°C and in the range of stresses 400-1300 Pa).

This complete characterization was necessary because, while the SAOTs give information about the mechanical characteristics of samples, allowing their rheological optimisation, the flow characterisation and the relaxation time (evaluated from stress relaxation tests) were necessary to obtain the material parameters introduced by constitutive equation discussed below.

By means of these data it was possible to determine an optimum recipe of fat replacer to produce a pastry similar to the commercial margarine.

A Stress Relaxation test of puff pastries was performed with the same rheometer equipped with a serrated plate geometry (\emptyset =25 mm) at 20°C, imposing a constant stress value within the linear viscoelastic region. Thus a relaxation time was determined.

2.3. Rheological data analysis and modelling

Structured emulsions can be described as weak systems, consequence of the development of a three-dimensional network of organogels molecules and cocoa butter crystals with rheological "units" connected by weak bonds. In these cases, the structured oil phase entraps both the oil solvent and the water drops, yielding to a stable water-in-oil emulsion similar to other organogel products reported in the literature (Marangoni 2009).

Owing a linear trend in log-log of both moduli, the rheological data can be interpreted by a "weak-gel model" (Gabriele et al. 2001):

$$G^{*}(\mathcal{T},\omega) = \sqrt{(G')^{2} + (G'')^{2}} = \mathcal{A}(\mathcal{T}) \cdot \omega^{\frac{1}{z(\mathcal{T})}}$$
(1)

where G^* is a complex modulus, ω the oscillation frequency, T the temperature and z the network extension, related to the number of interacting rheological units within the 3-D network, and A is the strength of the interactions. When A increases, the interaction forces within the network increase, whilst a high z value indicates a large number of interacting units cooperating and increasing the network connectivity.

Creep data, reported in terms of compliance versus time, were analysed by using a one-element Kelvin – Voigt viscoelastic model (Steffe, 1996)

$$J(t) = J_0 + J_1 \left(1 - e^{-\frac{t}{\lambda_1}} \right) + \frac{t}{\eta}$$
(2)

where J_0 is the instantaneous compliance, J_1 is the retarded compliance, λ_1 is the retardation time of the Kelvin component, and η is the viscosity. Thus the shear viscosity can be computed as the reciprocal value of the slope of the linear behaviour attained after the retardation time for any applied stress τ_0 , and reported as shear rate function as:

$$\dot{\gamma} = \frac{\tau_0}{\eta} \tag{3}$$

To extend the experimental range, dynamic and shear parameters were related through the so-called Cox– Merz rule applies (Cox and Merz, 1958) stating, as a rule of thumb, that shear and dynamic complex viscosity for polymer systems coincide:

$$\dot{\gamma} = \omega$$
 (4)

where $\dot{\gamma}$ is the shear rate and ω the oscillation frequency. According to the literature (Peressini et al. 2002) for food systems it was found that the equivalence between complex and shear viscosity can holds when assuming

$$\dot{\gamma} = a_{\dot{\gamma}}\omega \tag{5}$$

where $a_{\dot{\gamma}}$ is a shift factor of order of magnitude 0.01. Thus the complex viscosity can be computed as

$$\eta^{*}(\omega) = \frac{G^{*}(\omega)}{\omega} \tag{6}$$

and it can be reported in the same plot together with the steady shear viscosity. Both rheological parameters showed a very similar slope, thus it was possible to overlap those curves by a horizontal shift, estimating $a_{\dot{\gamma}}$. The obtained flow curve exhibited a power-law behaviour in the investigated shear rate range

$$\tau = k \cdot \dot{\gamma}^n \tag{7}$$

in which k can be considered as an index of the material

"consistency" and *n*, the flow index, can be calculated as the slope of the curve and its value is always positive and lower than one.

Finally, stress relaxation data were analysed in order to evaluate the relaxation time λ of the material, according to a single element Maxwell model. The relaxation of stress after the application of a sudden strain can be described by the following equation (Steffe, 1996).

$$\sigma = \sigma_0 \cdot \exp(-\frac{t}{\lambda}) \tag{8}$$

From the initial slope it is possible to find the λ value.

2.4. Sheeting process modelling

The flow is governed by the conservation equation of mass and momentum under isothermal conditions that for an incompressible material read:

$$\rho \cdot \left(\frac{\partial \underline{v}}{\partial t} + \underline{v} \cdot \underline{\nabla} \underline{v}\right) = -\underline{\nabla}P + \underline{\nabla} \cdot \underline{\tau}$$
(9)

$$\overline{\underline{V}} \cdot \underline{\underline{v}} = 0 \tag{10}$$

Where \underline{v} is the velocity vector, P is the pressure, ρ is the density of the pastry and $\underline{\tau}$ is the extra stress tensor and the body forces not considered.

Many complex fluids of industrial interest are assumed to be viscoelastic because they exibit contemporary viscous and elastic behaviour under strain. For these materials the extra stress may be written as an Upper convected Maxwell model (UCM)

$$\underline{\underline{\tau}} + \lambda \underline{\underline{\tau}} = 2\eta \underline{\underline{D}}$$
(12a)

Where $\underline{\underline{D}}$ is the rate of strain tensor and $\underline{\underline{\tau}}$ is the time upper convected derivative defined as:

$$\underbrace{\underline{\vec{\tau}}}_{\underline{\underline{\tau}}}^{\nabla} = \frac{\partial \underline{\vec{\tau}}}{\partial t} + (\underline{\underline{\nu}} \cdot \nabla) \underline{\underline{\tau}} - \left[(\nabla \underline{\underline{\nu}}) \underline{\underline{\tau}} + \underline{\underline{\tau}} (\nabla \underline{\underline{\nu}})^T \right]$$
(13)

Where $\underline{\underline{D}}$ is the rate of strain tensor, which is defined as:

$$\underline{\underline{D}} = \frac{1}{2} \left[\underline{\nabla} \underline{\underline{v}} + \left(\underline{\nabla} \underline{\underline{v}} \right)^T \right]$$
(14)

It should be noticed that the symmetry of the stress tensor implies that the off-diagonal extra stress components are equal, i.e. $\tau_{ij} = \tau_{ji}$ When considering a transient xy planar flow, then the only non zero components of the extra stress are τ_{xy} , τ_{xx} , τ_{yy} and the velocity shows only the two components v_i and v_y .

The values of the two material parameters introduced by eq.12 have been experimental determined as above described. Because the doughs show a shearthinning behaviour, it was necessary to modify the UCM model, by inserting the variation of viscosity with shear rate, as reported in the following? Indeed, the flow curve data were fitted with a power law model in the range between 0.1-100 s⁻¹ (eq. 7), whilst η_{∞} and η_0 were assumed equal to the viscosity value showed at 100 and 0.1 s⁻¹ respectively. The equation (12a) begins:

$$\underline{\underline{\tau}} + \lambda \underline{\underline{\tau}} = 2K \gamma \underline{\underline{D}}$$
(12b)

2.5. Method of solution

The constitutive equation (eq.12b) must be solved together with the conservation equations (eq.s 9-10) and with appropriate boundary conditions.

Figure 1 shows the solution domain. Because of symmetry, only one half of the domain is considered. The boundary conditions at any surfaces are reported in the following for material flowing from left to right, and indicating t and n as tangential and normal versor:

1. along the inlet boundary AF, the only stress acting across the boundary is due to pressure force: $\underline{\tau} \cdot \underline{n} = -p_{atm} \underline{n}$;

2. symmetry along the centerline AB: v_y=0, τ_{xy} =0;

3. along the exit free surface DC, tangential and normal stresses vanish: $(\underline{\tau} \cdot \underline{n}) \cdot \underline{t} = 0, (\underline{\tau} \cdot \underline{n}) \cdot \underline{n} = 0$ and no flow is passing through the surface being $(\underline{v} \cdot \underline{n}) = 0$, where \underline{n} , \underline{t} are the normal and tangential versors to the surface;

4. along the outlet boundary BC, the pressure is the atmospheric pressure: $\underline{\tau} \cdot \underline{n} = -p_{atm} \underline{n}$;

6. along the entry free surface FE, tangential and normal stresses vanish $(\underline{\tau} \cdot \underline{n}) \cdot \underline{t} = 0, (\underline{\tau} \cdot \underline{n}) \cdot \underline{n} = 0$ and no flow is passing through the surface being $(\underline{v} \cdot \underline{n}) = 0$;

7. drag flow along the roll walls from point E to point D (tangential velocity $v_t = \underline{t} \cdot \underline{U}$, normal velocity $v_n = \underline{n} \cdot \underline{U}$) (Mitsoulis E. 2008), assuming no slip.

The reference pressure is set to atmospheric pressure. For the simulations we have used the roll radius is 0.05 m and the velocity is 0.05 m/s. The H_f entry height is set to 0.03m, while the half minimum gap is set to 0.003 m. The numerical solution is obtained with the finite element method (FEM), using the program COMSOL Multiphysics 3.5. The free surfaces at the inlet and outlet are found in a coupled way with the ALE method, implemented in COMSOL too. Because the boundaries of the computational domain are moving in the time, it was necessary a method to deform the mesh. In COMSOL Multiphysics, it is possible to control the movement of the interior nodes by letting the mesh movement determined by physical deformation variables.

The technique for mesh movement is an arbitrary Lagrangian-Eulerian (ALE) method. In the special case

of a Lagrangian method, the mesh movement follows the movement of the physical material. Such a method is often used in solid mechanics, where the displacements often are relatively small.

When the material motion is more complicated, like in a fluid flow model, the Lagrangian method is not appropriate. For such models, an Eulerian method, where the mesh is fixed, is often used—except that this method cannot account for moving boundaries.

The ALE method is intermediate between the Lagrangian and Eulerian methods, and it combines the best features of both—it allows moving boundaries without the need for the mesh movement to follow the materia. The triangular mesh used is shown in-Figure 2 and give 19056 unknown degrees of freedom. More elements have been concentrated near the attachment and detachment points from the roll, where most of the changes are expected. The entering sheet is usually set with a length major then H_f of at least 3 order of magnitude and the exiting sheet with a length of 4H to guarantee a fully developed profile at entry and exit (Mitsoulis E. 2008, Mitsoulis E. 2009).



Figure 1: Flow domain and boundary conditions for the 2-D FEM analysis.



Figure 2: Finite element mesh used in the computation.

3. RESULTS AND DISCUSSION

3.1. Rheological analysis

According to the frequency sweep tests, all the samples showed a typical weak gel behaviour (data not shown); the weak gel model parameters of samples analysed after 72h from the preparation are listed in table 2, and compared to the margarine parameters.

Table 2: Weak Gel Model Parameters (72h from the

preparation)

Sample	A [Pa $s^{1/z}$]	z [-]
Margarine Vallè	43000±200	14±0.6
Emu1	86000±800	9.4±0.4
Emu2	14900±100	9.1±0.4
Emu3	30700±100	10.3±0.2
Emu4	6600±100	7.6±0.6

Samples Emu 1, Emu 3 and Emu 4 were prepared varying the Myverol content in the oil phase. Figure 3 shows the effect of Myverol on the rheological characteristics of the emulsions after 24h, 72h and 8 days (labelled '8d' in figure 1) after the preparation. All the samples underwent a maturation of the network strength until 72h after preparation, (increase of A until a constant value was reached), while the network extension (parameter z) was constant after 48h. Emu 1 (3% of Myverol) showed the highest A value. In fact, a further addition of Myverol (Emu 3) implicated a lower cocoa butter content, and a less consistent emulsion (lower A value), while a lower amount (Emu 4) gave the lowest values of both the weak gel model parameters.

It is worthy noticing that Myverol gives structure to the system (higher crystallisation degree). Nevertheless, despite the little increase of z for Emu 3 with respect to the other samples, higher values of A are preferable for our aims, also considering the comparison with the commercial margarine (Lupi et al. 2011).



Figure 3: Weak Gel model parameters for W/O emulsions at different times from preparation.

Thus, for our scopes, the best emulsion is Emu 1: it is characterised by the higher A value and it results stable (no more variations of both the parameters) after 72h from its production. Considering these results, the formulation of Emu 1 was varied increasing the total amount of water from 12 to 20%. Emu 2 showed a low value of A with respect to Emu 1, but a similar parameter z. It seems that an increase in water content does not affect the crystallisation, but it decreases the strength of the interactions in the network.

From a rheological point of view, the best pastry fat is Emul. This sample was used to prepare an hand-made puff pastry, characterised and compared to a commercial one. Both the SAOTs (table 3) and the flow tests (figure 4) revealed differences between the two pastries, obviously due to the different ingredients and preparation processes. Anyway, the hand-made pastry was cooked (at 200°C), and the consequent 'puffing' process resulted satisfying, giving the characteristic friable texture.

Table 3: Weak Gel Parameters For Pastries

Sample	А	Z
commercial puff pastry	88000±1300	4.6±0.2
hand-made puff pastry	127520±2480	3.1±0.1

The setting parameters used to implement the modelling software for the sheeting process are the relaxing time λ and the power law parameters *k* and *n* (eq. 7):

$$\begin{split} \lambda &= 0.400 \pm 0.005 \ s \\ k &= 5670 \pm 200 \ Pa{\cdot}s^{1/z} \\ n &= 0.27 \pm 0.004 \end{split}$$



Figure 4: Flow Curves Of Commercial And Hand-Made Puff Pastry (η K.V. is referred to eq. 2)

3.2. Sheeting modelling

We present results from the two-dimensional isothermal analysis of rolling of puff pastry. The velocity profiles and the value of Pressure and Normal stresses along the coordinate x is reported in the Figure 5 and 6.



pastry.

The velocity profile along the x-coordinate shows variations of concavity due to the pressure profile. In

fact, the speed has a reversal point of the concavity at the attachment point. This change is due to the complete adherence of the sheet to the roll and then to the drag force that pushes forward the dough. As the material passes through the nip, it accelerates. The magnitude of the velocity at the nip is almost 3 times higher than the entry.

Then, after the maximum pressure value, the velocity increases due to the pressure gradient to begins constant, showing a typical plug velocity profile due to the existence of the free surfaces (Mitsoulis E. 2008, Mitsoulis E. 2009). Its speed at the exit is higher its speed at the entry, because of the mass conservation, namely, as the thickness decreases, the speed increases.

Looking at the pressure profile (Figure 6), it is possible to observe that the maximum pressure value is at 0.11 m, then before the minimum gap, and have a maximum value of 0.11 MPa. The pressure profile is slightly asymmetric, in agreement with the literature, due to the non-Newtonian material (Mitsoulis E. 2008, Mitsoulis E. 2009) and as expected, the biggest changes occur in the nip region.



Figura 6: Pressure Profile in rolling of puff pastry.

Finally, it is clear that the points of attachment and detachment give the difficulty in the calculation with FEM.

The swelling of the puffy pastry is found at the exit of the nip of the roll, due to the material memory that completely relaxes the normal stresses because is not any longer confined. The point of detachment was obtained at a value of about 0,1455 m. The value of swelling, calculated as H/H_0 is equal to 1.8.

The normal stress τ_{xx} varies along the calendaring exhibiting its maximum value at the attachment point and a minimum value at the maximum of the pressure (Figure 6).

The model validation was not performed, but it is currently under experimental investigation.

4. CONCLUSIONS

Rheological modelling allowed, through the optimization of the formulation of water-in-olive-oil emulsions, structured with an organogelator agent, to characterize and obtain a puff pastry with rheological characteristics similar to the commercial one. The rheological parameter were used to model a rolling process. The rolling model, using an UCM modified constitutive equation, was implemented in Comsol Multiphysics 3.5.The velocity, pressure and normal stress profile were found and the swell and detachment point were predicted by the model.

The present results are a first step towards a better understanding of rolling of viscoelastic materials. This analysis coupled with rheological data for a UCM modified model, and operating data for the process, can be useful to predict the final sheet height and swell.

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