

Chapter 2

Expansion of a single bubble in an infinite viscoelastic medium

2.1 Introduction

Dough may be considered as a foam since air bubbles incorporated during mixing are separate and surrounded by a continuous phase. It is generally accepted that the rheological properties of dough are extremely important for the final quality of every baked product and that it is very important to control the latter from the mixing stage onwards.

Biscuits are commonly prepared from flour, water, sugar, butter, eggs and additional ingredients. On adding water, flour particles are wetted and slowly hydrated, but the mass still remains in a lumpy incoherent form until mixing is started, during which the mixture develops into a coherent homogeneous-appearing dough. Starch granules, yeast cells and air bubbles are dispersed in a continuous phase.

Gas bubbles and then aeration during mixing are critical to baked loaf quality in the mechanical dough development process and, moreover, this stage plays a crucial role for all other bakery products. The amount of air and the bubble-size distribution incorporated during mixing critically affect the structure, texture and appearance and for this reason the occlusion of air during mixing is a vital step. The behaviour of the bubbles in a rheological complex medium plays a crucial role in many industrial fields. One of the industrial sectors interested in these phenomena is the alimentary sector since many goods produced, are constituted of a liquid matrix that englobes some gas. Dough being a dense system, with high consistency, and definable from a rheological point of view as solid-like material, involves rather limited bubble movement, however negligible.

As the gluten layers are being sheared and folded by the mixing blades, air gets entrapped within these layers. The air bubbles are incorporated into the dough during the mixing stages and these bubbles form the nuclei for the gas cells. These gas cells will expand due to the production of carbon dioxide and other products of fermentation reactions that diffuse into the gas cells during the leavening stage. It should also be emphasized that no new cells can be produced beyond these stages, but the existing cells may be subdivided during the moulding and sheeting operations [Elmehdi H. M., 2001]. Therefore, the gas bubble structure and the volume potential of the final product are largely determined by the end of

the mixing process. Among the important features of the matrix is its ability to retain gas and its ability to accommodate the expansion of the gases during the fermentation process.

Then, it is possible to say that the bubbles incorporated during mixing act as nucleation sites for bubble growth during proving; without these sites, the CO₂ produced by the yeast is largely lost to the atmosphere, resulting in a coarse, unattractive loaf or biscuit of low volume [Campbell G. M. et al., 1999]. Unfortunately the mechanisms controlling the bubble formation and its fate are not well understood, what is evident is that the viscosity of the dough and the carbon dioxide produced during fermentation play a fundamental role in the formation of the cellular structure of leavened bread [Bloksma AH, (1990)] and baked biscuits. The nature of the materials forming the thin bubble walls is unknown although gluten is often assumed to be involved for foam-like dough. The ability of gas bubbles in the dough to expand, suggests that the walls have considerable elasticity, while bubble expansion and reduction for products like whipped cream is strictly related to both rheological and interfacial properties; indeed, single cell growth is due to the internal pressure (higher than the external one) but it is limited by bulk properties of the viscoelastic medium and by interfacial properties of the emulsion layer.

This chapter describes the bubble growth in the continuous viscoelastic medium that surrounds the single cell. The growth of spherically symmetric bubbles in liquids has been the object of continual investigation and the source of much stimulation for scientists and engineers interested in the interaction between the transport of mass, energy, and momentum. Practical applications are apparent in the study of boiling, cavitation, and evaporative transport. Here we turn our attention to the growth of spherical bubbles in a non-Newtonian viscoelastic medium to help the phenomenological study of baking.

To describe the dimensional change it is, therefore, necessary to solve a mechanical problem by writing the momentum balance containing a constitutive equation describing in the proper way the medium rheological behaviour.

2.2 Bubble Growth

It was stated that the bubbles inserted at the beginning into the dense cereal paste, grow as a consequence of the combined actions due to raising agent decomposition, water evaporation, and temperature increase. All these factors directly and indirectly influence gas cell composition and consequently their volume. Owing to their initial dimensions, at the start the bubbles may be

considered as submerged in an infinite medium of paste. According to the suggestion of dividing the problem of bubble growth into kinetics, thermodynamic and mechanical equilibrium, the system was assumed to be constituted of a single closed spherical gas bubble surrounded by an infinite mass of a viscoelastic paste. At any instant this system is subjected to both mechanical and thermodynamic equilibrium. In such a way, once a finite integration time interval is fixed, from the reaction kinetics the amount of either new products or components present at any time in the gas bubble is determined by a thermodynamic flash, while the bubble volume is computed by the mechanical equilibrium. Thus, from a theoretical point of view, the process has been considered to change as a quasi-static transformation, allowing passing through equilibrium stages.

According to [de Cindio and Corra \(1995\)](#), the bubble expansion modelling is developed, considering the mechanical behaviour of a single bubble expanding in a viscoelastic fluid assumed as infinite medium (figure 2.1).

It can be considered as a reasonably homogeneously mixed medium and it can be assumed that the bubbles of air formed during the mixing phase are uniformly distributed and all have a radius equal to R_0 [[CARLSON And BOHLIN, 1978](#)]. Moreover, it can be considered that the system is not concentrated and that the bubbles inside the medium are in such number as not to interact among themselves. Then the spherical bubbles of radius R_0 are considered and a system of spherical coordinates is defined with the origin in the centre (figure 2.1).

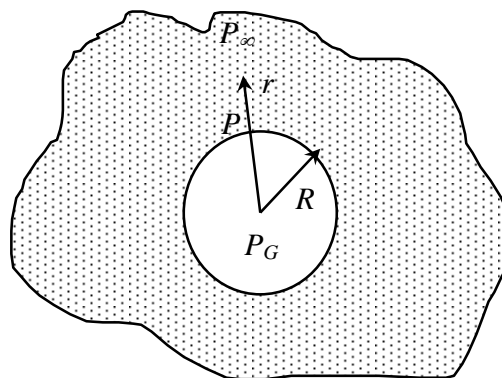


Figure 2.1 – The bubble-paste system.

The volumetric gas fraction, can be expressed as simply as the ratio between the volume of gas and the total volume of the system:

$$\varphi = \text{frazione volumetrica} = \frac{V_{gas}}{V_{Tot}} \quad (2.1)$$

where the total volume of the bubbles is:

$$V_{gas} = n_{bubble} * V_{bubble} \quad (2.2)$$

It is supposed that the bubbles contain only air, the pressure p_0 agent initially on the inside surface in the bubble will be given by:

$$p_0 = p_g(t=0) = p_{atm} + \frac{2\gamma}{R_0} \quad (2.3)$$

Where γ represents the interfacial tension air/paste and R_0 is the initial radius of a single bubble.

Bubble unity is defined as the air bubble and the part of continuous phase that interacts with it. This mass will be given by:

$$m_{ub} = \rho_p \left(\frac{1-\varphi}{\varphi} * \frac{4}{3} \pi R_0^3 \right) \quad (2.4)$$

where ρ_p is the density of the continuous phase. It is possible reasonably to assume for the air an ideal gas behaviour, therefore supposing that is present n_a^0 moles of air, we will have:

$$p_0 * V_{bubble}^0 = n_a * R_g * T_0 \quad (2.5)$$

$$\left(p_{atm} + \frac{2\gamma}{R_0} \right) * \frac{4}{3} \pi R_0^3 = (n_a^0) R_g * T_0 \quad (2.6)$$

The kinematics analysis of the bubble growth was performed separately considering the continuous side and the phase gas. With reference to a differential element of matrix spherical inclusive between r and $r+dr$, it is had:

- Paste Size:

equation of continuity

$$\frac{D\rho_p}{Dt} = -\rho_p \nabla \cdot \underline{v} \quad (2.7)$$

\underline{v} = Velocity vector, in the hypothesis of incompressibility of the continuous medium and spherical symmetry. The speed \underline{v} in such hypotheses has the only radial component V different from zero and that is only a function of the radius. The continuity equation reduces it therefore to:

$$\frac{\partial \rho}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} (V(r) * r^2) = 0 \quad (2.8)$$

In the case of $\frac{\partial \rho}{\partial t} = 0$, integrating between R and $r > R$, we have:

$$V(r) * r^2 = V(R) * R^2 \quad (2.9)$$

- Bubble Side

The mass balance relative to the interval of time dt , during which the bubble grows to dR , requires that the variation of mass of the bubble during this time proves equal to the gas flow that enters inside it during the same interval. The gas flow is equal to the difference between the total mass of continuous phase in the spherical shell of thickness dr and the mass of paste that moves toward the outside in the time dt .

The mass balance can be written then as following:

$$d\left(\frac{4}{3}\pi R^3 * \rho_g\right) = 4\pi R^2 \rho_i dR - 4\pi R^2 \rho_p V(R) dt \quad (2.10)$$

From this last relationship the value of the radial behaviour of the velocity $V(R)$ can be as following:

$$-V(R) = \frac{\rho_g - \rho_i}{\rho_p} * \frac{dR}{dt} \quad (2.11)$$

Moreover, it can be held that ρ_i is around equal to ρ_p being the contribution of gas dissolved negligible to the effects of the calculation of the total density. We then have:

$$V(R) = \frac{\rho_p - \rho_g}{\rho_p} * \dot{R} \approx \dot{R} \quad (2.12)$$

being $\rho_p \gg \rho_g$ and considering the substitution of the 2.12 in the 2.9, we get:

$$V(r) = \dot{R} * \frac{R^2}{r^2} \quad (2.13)$$

If we point out with $\dot{\varepsilon}$ the mono-dimensional elongational velocity of the bubble that expands, it has:

$$\dot{\varepsilon} = \frac{\partial V(r)}{\partial r} = \frac{\partial}{\partial r} \left(\dot{R} * \frac{R^2}{r^2} \right) = -2 * \frac{R^2 \dot{R}}{r^3} \quad (2.14)$$

For which the kinematics realized during this phase can be expressed, in general terms, by the following tensor of deformation velocity:

$$\underline{\underline{D}} = \dot{\varepsilon} \begin{bmatrix} 2 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{bmatrix} \quad (2.15)$$

It is possible now to make an analysis of the dynamic process from the side of the continuous phase. The momentum balance, in general terms, can be written as:

$$\frac{D\underline{v}}{Dt} = -\underline{\nabla} p - \underline{\nabla} \underline{\tau} + \rho_p * \underline{g} \quad (2.16)$$

Where $\underline{\tau}$ represents the stress tensor or equivalently the flow of motion quantity and \underline{g} the vector acceleration of the gravitational forces.

Neglecting the contribution of the gravity forces, assuming the incompressibility of the fluid and the spherical symmetry, the radial component of 2.15 becomes:

$$\rho_p \left(\frac{\partial v}{\partial t} + v \cdot \frac{\partial v}{\partial r} \right) = -\frac{\partial p}{\partial r} - \left(\frac{\partial \tau}{\partial r} + 3 \frac{\tau}{r} \right) \quad (2.17)$$

τ represents the component of the deviatoric tensor of the stresses that acts long r on the surface of normal r .

Replacing in equation 2.16 the $V(r)$ and its derivative, gets:

$$\rho_p \left(2 \frac{\dot{R}^2 R}{r^2} + \frac{R^2}{r^2} * \ddot{R} - 2 \frac{R^2 \dot{R}^4}{r^5} \right) = -\frac{\partial p}{\partial r} - \left(\frac{\partial \tau}{\partial r} + 3 \frac{\tau}{r} \right) \quad (2.18)$$

Neglecting the inertial forces, represented by the first member of the 2.15 and integrating between the value $R(t)$ and the infinite:

$$0 = -\frac{\partial p}{\partial r} - \left(\frac{\partial \tau}{\partial r} + 3 \frac{\tau}{r} \right) \quad (2.19)$$

$$\int_{R(t)}^{R(\infty)} dp = -\int_{R(t)}^{\infty} \left(\frac{\partial \tau}{\partial r} + 3 \frac{\tau}{r} \right) dr \quad (2.20)$$

$$p(\infty) - p(R) = -\int_R^{\infty} \left(\frac{\partial \tau}{\partial r} + 3 \frac{\tau}{r} \right) dr \quad (2.21)$$

Then:

$$p(R) - p(\infty) - \int_R^{\infty} \left(\frac{\partial \tau}{\partial r} + 3 \frac{\tau}{r} \right) dr = 0 \quad (2.22)$$

The momentum balance to the interface paste/bubble, in the hypothesis that the stress on the gas side is negligible:

$$p_g = p(R) + \tau(R) + \frac{2\gamma}{R} \quad (2.23)$$

replacing in 2.22 the 2.21 and assuming at infinite distance p_{∞} equal to p_{atm} , we have:

$$p_g = p_{am} - \int_R^\infty \left(\frac{\partial \tau}{\partial r} + 3 \frac{\tau}{r} \right) dr + \tau(R) + \frac{2\gamma}{R} \quad (2.24)$$

$$p_g = p_{am} + \frac{2\gamma}{R} - 3 \int_R^\infty \frac{\tau}{r} dr \quad (2.25)$$

This relationship expresses the functionality between the pressure, that is realized inside the gas bubble, the interfacial tension, the radius of the bubble and the stress conditions that are realized in the shell of continuous phase that envelops the bubble.

The following quantity:

$$R_h = -3 \int_R^\infty \frac{\tau}{r} dr \quad (2.26)$$

it represents the rheological contribution and ties the value of the stress component to the kinematics and therefore to the deformation. Obviously this contribution will be different according to the models assumed for describing the material behaviour.

It is necessary to specify the term R_h , or τ , and this is possible postulating a constitutive equation like:

$$\underline{\underline{\tau}} = h(\underline{\underline{D}}) \quad (2.27)$$

which through the functionality $h(\underline{\underline{D}})$ models the rheological behaviour of the material.

The food materials have some properties that initially can be approximately well-interpreted by rheological models like viscoelastic models, that is, by models for which the stress is not only a function of $\underline{\underline{D}}$, but also a function of the deformation history to which the material is subjected. Specifically, the almost rheological linear equation of Goddard and Miller was used that, in the hypothesis of small deformations, describes the viscoelastic linear fluid. According to equation 2.14:

$$\tau(r, t) = - \int_{-\infty}^t G(t-t') \dot{\epsilon}(r', t') Dt' \quad (2.28)$$

where $G(t-t')$ is the relaxation modulus and the integral must be solved in a substantial way. If we impose $s=t-t'$, we get:

$$Rh = \int_0^t 12G(s, T) \frac{\dot{R}[s] \cdot [R(s)]^2}{[R(s)]^3 - [R]^3} \ln \frac{R(s)}{R} ds \quad (2.29)$$

This quantity must be inserted in the equation 2.25. Weakly structured systems, such as foods, emulsions and others, show a characteristic enough common behaviour: if they are subjected to deformation they behave like a solid, while for high deformations the weak bonds present in their structure break and they begin to flow as a liquid, but first the functionality of the relaxation modulus

with time and temperature must be specified. This structure is typical of weakly structured systems that are indicated as "weak gel". It seems, therefore, reasonable to start from the already existing theories in the sector of the gels, to build a unique model to describe food behaviour, which takes into account the weakness of the interactions. This structure is typical of weakly structured systems that are indicated as "weak gel"[Gabriele et al., 2001].

One of the more-used theories for the study of gels is the theory of Winter [WINTER, 1989], according to which the relaxation modulus of a critical gel, which is in correspondence to the transition sol-gel, follows a power law like the following:

$$G(t) = S \cdot t^{-n} \quad \lambda_0 < t < \infty \quad (2.30)$$

where S is defined as "the gel strength", a function of the mobility of the segments of the chain, n is the relaxation exponent, λ_0 represents the shortest relaxation time, which depends on the original material, below which other dynamics of the polymer are prevailing. Applying the equation 2.30 to the equation 2.29, is obtained:

$$Rh = 12 \cdot S \int_0^t (t-t')^{-n} \frac{\dot{R}[s] \cdot [R(s)]^2}{[R(s)]^3 - [R]^3} \ln \frac{R(s)}{R} ds \quad (2.31)$$

The latter equation completely closes the problem under examination. Effectively the model of Winter was modified applying it to a system that is solid-like in a limited range of relaxation times [Gabriele et al., 2001]. This allows the original theory of Winter to be applied to the foods.

Therefore, if the initial bubble radius is known and the paste material parameter G(t) is measured, it is possible to calculate the bubble radius as a solution to the equations 2.25.

The proposed approach needs the solution to the dynamics of the paste embedding the bubble as a function of external conditions. These are, in turn, local temperature profiles (changing according to baking conditions and local position of the bubble inside the biscuit) and the internal pressure P_G assumed to follow the well-known constitutive equation of an ideal mixture of ideal gases:

$$P_G = \sum_i P_i = \frac{R_g T}{V} \cdot \sum_i n_i \quad (2.32)$$

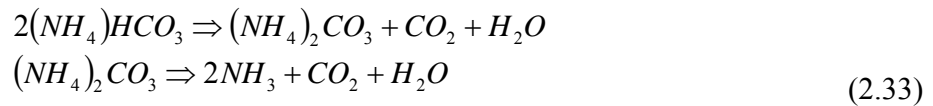
where R_g is the gas constant, n_i are the moles of any i-component in the gas bubble.

2.3 Internal gas pressure calculation

The gas pressure inside the bubble changes with time as the effect of two main factors: the production of carbon dioxide and ammonia as the effect of raising agent decomposition and the changes in water vapour, assumed to be in equilibrium with the liquid phase inside the dough.

2.3.1 Raising agent decomposition kinetic

Raising agents used in biscuit production, such as ammonium bicarbonate in the present work, are responsible for gas production as a result of temperature-driven decomposition. The chemical compound is dissolved in water partially bonded inside the gluten network and the following stoichiometric decomposition equations must be taken into account:



The two reactions take place only in the liquid phase and reaction products can be assumed to be soluble only in water and not in the paste. In addition CO_2 exhibits a rather low solubility described by Henry's Law. These conditions suggest that it is reasonable to assume that the two reactions are irreversible, and according to the literature (Nowak and Skrzypek, 1989) the following kinetic equations for the second-order irreversible reaction was used:

$$\begin{aligned}-r_A &= k_1 c_A^2 \\-r_B &= k_2 c_B^2\end{aligned}\quad (2.34)$$

where A and B stand respectively for NH_4HCO_3 and $(NH_4)_2CO_3$. If the rates are expressed in $mmol/(kg\ dough \cdot s)$, the kinetic constants of 2.34, expressed as $kg\ dough / (mmol \cdot s)$, are:

$$\begin{aligned}k_1 &= 3.5 \cdot 10^6 \cdot e^{\frac{-7700}{T}} \\k_2 &= 1.3 \cdot 10^{11} \cdot e^{\frac{-11900}{T}}\end{aligned}\quad (2.35)$$

Reaction products are still dissolved in the liquid phase but, according to the thermodynamic conditions, tend to go into the gas phase, according to the liquid/vapour equilibrium.

2.3.2 Thermodynamic equilibrium

The modelled system is a single bubble surrounded by a fixed amount of dough that can be determined by the mass balance if the bubble concentration is known (number per unit of volume). The reactions take place only in the partially immiscible liquid-like phase and the product is assumed to be at

thermodynamic equilibrium with a gas phase (Figure 2.1). The water is assumed to be partially bonded to the flour paste and the raising agent remains in the liquid phase because of its low volatility. In addition the products coming from the ammonium bicarbonate irreversible decomposition, carbon dioxide and ammonia, are assumed to be soluble only in water. The phase equilibrium phenomenon and the chemical production of gases take places simultaneously but, assuming a fast decomposition rate, a “quasi-static” equilibrium transformation can be considered. If a finite time interval is fixed, from the reaction kinetics it is possible to compute the amount of new products and, with respect to the mass balance, a thermodynamic flash allows computing the amount of components present at any time in the gas bubble and the paste, under equilibrium conditions. The following thermodynamic equilibrium equations apply:

$$\begin{aligned}
 P_{H_2O} &= a_w \cdot P_{H_2O}^0 = P_G \frac{n_{H_2O}}{n_{tot}} \\
 P_{CO_2} &= H_{CO_2} \cdot x_{CO_2} = P_G \frac{n_{CO_2}}{n_{tot}} \\
 P_{NH_3} &= P_{NH_3}^0 \cdot x_{NH_3} = P_G \frac{n_{NH_3}}{n_{tot}} \\
 P_G &= P_{H_2O} + P_{CO_2} + P_{NH_3} + P_{Air}
 \end{aligned} \tag{2.36}$$

It is worth noticing that because CO_2 and NH_3 are soluble in water, but completely immiscible in the cereal paste, their molar fraction x in the liquid phase must be referred only to water. For CO_2 a classic Henry’s theory holds, whilst for NH_3 the ideal Raoult’s law was applied. On the contrary H_2O , coming from both the amount initially added to flour and that produced by the reactions, is governed by the water activity bonding relationship inside the dough. In addition, the air present from the beginning in the bubbles is assumed to be completely insoluble in the liquid phase.

The thermodynamic computation is performed according to an equivalent mass flash at constant temperature governed the set of 2.36 (Smith and Van Ness, 1987). At the beginning the old equilibrium (or the initial condition) is known, which is changing because both temperature changes and raising agent decomposition proceeds. Then it was assumed that the new conditions can be calculated solving the equilibrium stage where the inlet feed F is represented by the sum of total moles both in the dense phase and in the newly generated gas phase, and the outlet vapour V is given by the moles going into the bubble, and the outlet liquid L includes the moles remaining in the paste. According to this model the following total and partial mass balances hold:

$$\begin{cases} F = L + V \\ F \cdot z_i = L \cdot x_i + V \cdot y_i \end{cases} \quad (2.37)$$

According to Eqs.12, the following expressions may be written in terms of gas and liquid molar fractions:

$$\begin{aligned} y_{H_2O} &= \frac{a_w \cdot P_{H_2O}^0}{\pi} \\ y_{CO_2} &= K_{CO_2} \cdot x_{CO_2} \\ y_{NH_3} &= K_{NH_3} \cdot x_{NH_3} \\ K_{(NH_4)HCO_3} &= \infty \\ K_{(NH_4)_2CO_3} &= \infty \end{aligned} \quad (2.38)$$

where K_i are the partition coefficients representing the split between vapour and liquid. They are fixed if temperature and pressure are known and for the raising agents are assumed coherently to the non-volatility of the compounds. Since in the considered system water may deviate from ideal behaviour two different computing procedures were adopted, according to a_w values. When $a_w=1$ the ideal condition is used in 2.38, this condition is realized when the temperature is above $100^\circ C$. On the contrary, at lowest temperatures, the current value of the water activity has to be used. In order to solve the set of equations 2.36 was adjusted obtaining 2.38 where the unknown values are V and K_i .

$$\sum_i y_i = \sum_i \frac{F \cdot z_i}{V \cdot \left(1 - \frac{1}{K_i}\right) + \frac{F}{K_i}} = 1 \quad (2.39)$$

Using 2.37 and by applying an iterative method, V can be calculated and equilibrium conditions can be calculated as the solution of the overall set of non-linear equations.

2.4 Conclusions

Thus given an initial bubble radius and the paste material parameters, it is possible to solve the previous equations that are given as the bubble evolution with time in a non-Newtonian-viscoelastic medium. Pressure inside the bubble, P_G , depends on the total amount of gas, which has to be estimated at any time to take into account raising agent development and water evaporation. It should be remembered that raising agent products, such as carbon dioxide and ammonia, are produced by chemical reaction in the paste phase but they tend to go, together with water, into the gas phase,

producing an increase of the total amount of moles. Assuming that thermodynamic equilibrium, between bubble and surrounding paste, is instantaneously obtained, the amount of volatile components can be calculated at any time. The amount of raising agent produced, at any time, can be determined by using proper kinetic equations. The equation system composed of mechanical and thermodynamic equilibrium equations, can be solved to calculate at any time the bubble radius (and, therefore, a local void fraction) and CO₂, NH₃, H₂O mass distribution between gas and paste phase. However, no limitation is imposed on bubble growth, whilst experimental evidence shows that a maximum bubble dimension cannot be overcome, therefore a gas cell stabilization or coalescence criterion must be inserted into the model.

Generally speaking, the dough being dense, few definable fluids from a rheological point of view as solid-like materials, involve limited bubble movement, however negligible. It is possible to conclude then with the belief that the bubbles initially grow in an infinite medium, to then start to interact among themselves, reaching coalescence conditions or, following the approach proposed by van [Vliet et al. \(1992\)](#), at a determined instant of the process, the stabilization of the gas cells inside the dough is obtained, which can substantially happen either thanks to the superficial properties or owing to strain hardening. In the next chapter the criterion to use for the biscuit baking model will be discussed.

Moreover, this chapter has allowed highlighting of how the use of rheology is important with the purpose of defining the conditions that are realized during baking and, certainly, how to write and to use an appropriate constitutive equation is essential for a good writing and matching of the model. For this reason it will be essential to complete bubble growth simulations, a rheological fundamental characterization able to furnish the essential material parameters to the model as well as an opportune criterion of coalescence or stabilization of the gas cells.

2.5 References

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