A Gas Supply and Gas Diffusion

A.1 Generalized Johnson-Mehl-Avrami Approach

Aim of this section is to describe a theoretical approach to apply JMA theory (see Sect. 2.5.2.2), which is generally used at constant temperature, to experiments with constant heating rate. The following description is based on the PhD thesis by Kempen [135].

A.1.1 Theory

The JMA approach, Eq. (2.5), has to be generalized in order to apply it to the experimental situation of a constant heating rate $\dot{T}$ with

$$T(t) = T_0 + \dot{T}\,t, \tag{A.1}$$

where T_0 denotes the initial temperature.

It is assumed that the thermal history, i. e. the path in the time-temperature-diagram, determines the degree of transformation (of the blowing agent MgH_2). A path variable β, which is a function of the thermal history describes the degree of transformation [135, 180]:

$$f(t) = \exp\left(-\beta^n\right). \tag{A.2}$$

The dependence of the path variable β on the thermal history can be described as a time integral of a constant $k(T(t))$, where the time dependence is only implicit through the temperature:

$$\beta(t) := k_0 \int_0^t \exp\left(-\frac{E_\mathrm{A}}{R\,T(\tilde{t})}\right)\mathrm{d}\tilde{t} \quad \overset{\text{Eq. (A.1)}}{=} \quad k_0 \int_0^t \exp\left(-\frac{E_\mathrm{A}}{R(T_0 + \dot{T}\tilde{t})}\right)\mathrm{d}\tilde{t}. \tag{A.3}$$

Equation Eq. (A.3) is based on additivity, which is only valid if the transformation mechanism does not change throughout the temperature range of interest. We follow the approach in [135] and substitute the time variable t by

$$\hat{t} = t + T_0/\dot{T} \;=\; T/\dot{T}. \tag{A.4}$$

The time variable $\hat{t}$ is the time which would be necessary to heat the sample from $T = 0$ K to the temperature T with constant heating rate $\dot{T}$. With this new time

variable, Eq. (A.3) reads [135]:

$$\begin{aligned}\beta(\hat{t}\,) &= k_0 \int_{T_0/\dot{T}}^{\hat{t}} \exp\Big(-\frac{E_\mathrm{A}}{R\,\dot{T}\,\tilde{t}}\Big)\mathrm{d}\tilde{t} \;\approx\; k_0 \int_0^{\hat{t}} \exp\Big(-\frac{E_\mathrm{A}}{R\,\dot{T}\,\tilde{t}}\Big)\mathrm{d}\tilde{t} \\ &\approx \frac{k_0\,\dot{T}\,R\,\hat{t}^2}{E_\mathrm{A}} \exp\Big(-\frac{E_\mathrm{A}}{R\,\dot{T}\,\hat{t}}\Big). \end{aligned} \tag{A.5}$$

Back substitution of $\hat{t}$ by $T/\dot{T}$ results in

$$\beta(T) = \frac{k_0\,R\,T^2}{E_\mathrm{A}\,\dot{T}}\;\exp\Big(-\frac{E_\mathrm{A}}{R\,T}\Big). \tag{A.6}$$

A.1.2 Material Constants for MgH_2

There are three constants which have to be experimentally determined, E_A, n and k_0, compare Eq. (2.5) and Eq. (k-for-JMA). The apparent activation energy was found by Andreasen et al. [3] with the help of isothermal experiments: $E_\mathrm{A} = 160$ kJ/mol. The Avrami exponent n and the constant k_0 are determined by fitting the theoretical decomposition data, Eq. (A.6), to own decomposition experiments[1], see Fig. A.1.

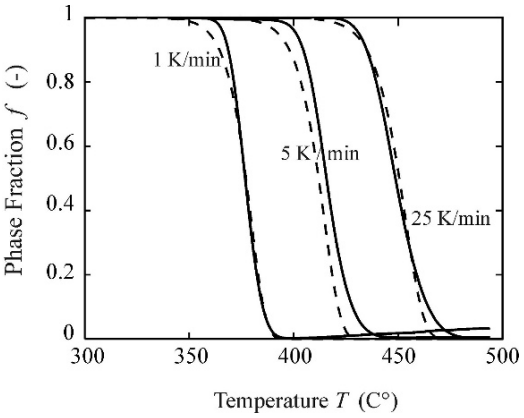

Figure A.1: Decomposition of MgH_2 at heating rates of 1, 5 and 25 K/min. *Full lines*: Experimental data (thermo-gravimetry). *Dashed lines*: Theoretical data fitted by applying Eq. (A.2), Eq. (A.6) and Table A.1.

[1]The decomposition of MgH_2 was measured by thermo-gravimetry: NETZSCH STA 409C, temperature range: 20–500 °C, argon gas.

The temperature at the onset of decomposition is determined by k_0 whereas the steepness of the decline is given by n.[2] The JMA theory is in fact able to describe the decomposition and its dependence on the heating rate. The physical parameters are listed in Table A.1.

Table A.1: Parameters for the decomposition of MgH_2 on basis of JMA theory.

E_A	n	k_0
160 kJ/mol	3	$4.63 \cdot 10^9\,s^{-1}$

A.2 Gas Diffusion

There are several situations where the exact description of gas diffusion is essential for foam evolution:

- Very low density of blowing agent particles.
- Ostwald ripening. The different capillary pressures of bubbles with different diameter lead to gas currents from smaller bubbles to larger ones. Although very important for aqueous foams, Ostwald ripening has been demonstrated to be irrelevant for metal foams [15, 146].

During IFM, gas is provided by quite large (10–100 µm) MgH_2 particles whose role is to act as bubble nucleation sites and to release gas directly into bubbles, see Fig. A.2, left. The role of gas diffusion appears to be quite moderate due to the direct contact with the bubbles and the small diffusion length calculated from the diffusion constant. The most exact way to describe this situation would be to consider each blowing agent particle as individual blowing agent source. Such a procedure would strongly exceed the numerical capacities.

In order to mimic the real situation the individual blowing agent particles are considered as homogeneous volume source, see Fig. A.2, right. In addition, the diffusion constant is strongly increased to make the gas diffusing very quickly to the bubble nuclei. This procedure has a similar effect as individual blowing agent particles releasing their gas directly into bubbles. During foam expansion, the number of blowing agent particles present at bubble interfaces increases due to coalescence events. Thus, differences between the treatment of individual blowing agent particles and the homogeneous volume source more and more diminish with increasing

[2] The Avrami exponent obtained in literature varies between $n = 2$ and $n = 4$ [69, 70].

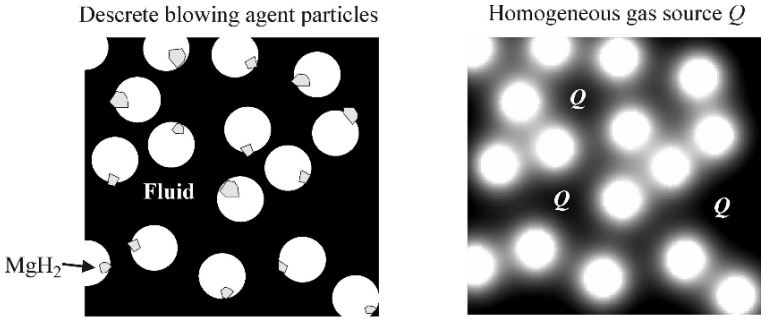

Figure A.2: Gas release and diffusion (schematic). *Left*: Real process. Blowing agent particles acting as bubble nuclei and releasing their gas directly into bubbles. *Right*: Modeling. Homogeneous volume source and high diffusion constant.

porosity. In order to imitate the real situation, the physical material parameters[3] have to be adapted. We demand:

1. $Q^\star \cdot n_{\text{steps}} \gg S^\star$ where n_{steps} denotes the number of time steps,
2. $Q^\star \ll S^\star$ and
3. $\sqrt{4\,D^\star \cdot n_{\text{steps}}} \gg 1$.

Condition 1 guarantees that the total amount of released gas is much larger than the amount of gas which is dissolved in the fluid. Condition 2 and 3 ensure that the gas does not accumulate in the fluid but diffuses rapidly to the bubbles.

[3]Dimensionless parameters are marked with $\star$, see also Appendix C.

B Miscellany

B.1 Foaming Pressure

The aim of this section is to estimate the foaming pressure during IFM of aluminum. In a first step, the amount and volume of hydrogen gas evolving during IFM is calculated. The amount of hydrogen gas n_{H_2} contained in 1 g MgH_2 follows from

$$n_{H_2} = \frac{m_{MgH_2}}{M_{MgH_2}} = \frac{1\,\mathrm{g}}{26.32\,\mathrm{g/mol}} = 0.038\,\mathrm{mol}, \tag{B.1}$$

where m_{MgH_2} and M_{MgH_2} denote the mass and the molar mass of MgH_2, respectively.

The volume of hydrogen gas V_{H_2} contained in 1 g MgH_2 is given by the ideal gas law,

$$V_{H_2} = n_{H_2} \cdot \frac{R\,T}{p} = \frac{2750\,\mathrm{cm^3\,bar}}{p}, \tag{B.2}$$

where $T = 600°\mathrm{C}$ is assumed.

Only part of the total blowing agent decomposes due to the chilling effect of the walls. In a simple model, the casting is divided into a compact skin where no decomposition of the blowing agent takes place and a cellular core where complete decomposition is assumed. The total mass of decomposed MgH_2 is given by the mass of the cellular core m_{core} and the mass fraction ϵ_m of the blowing agent,

$$m_{MgH_2} = \epsilon_m \cdot m_{core} = \epsilon_m \cdot \rho_{rel,core} \cdot \rho_{Al} \cdot V_{core} \tag{B.3}$$

with V_{core}: volume of the cellular core, ρ_{Al}: density of aluminum and the relative density of the core,

$$\rho_{rel,core} = 1 - (1 - \rho_{rel}) \cdot \frac{d}{d_{core}}, \tag{B.4}$$

where d and d_{core} denote the thickness of the casting and the core, respectively.

The gas volume V_{H_2} resulting from the blowing agent is

$$V_{H_2} = m_{MgH_2} \cdot \frac{2750}{p}\,\frac{\mathrm{cm^3\,bar}}{\mathrm{g}} = \epsilon_m \cdot \rho_{rel,core} \cdot \rho_{Al} \cdot V_{core} \cdot \frac{2750}{p}\,\frac{\mathrm{cm^3\,bar}}{\mathrm{g}}. \tag{B.5}$$

On the other hand, V_{H_2} is determined by the relative density of the cellular core,

$$V_{H_2} = V_{core} \cdot (1 - \rho_{rel,core}). \tag{B.6}$$

The resulting **foaming pressure** p follows from Eqs. (B.5) and (B.6):

$$\begin{aligned} p &= \epsilon_{\mathrm{m}} \cdot \frac{\rho_{\mathrm{rel,core}}}{(1-\rho_{\mathrm{rel,core}})} \cdot \rho_{\mathrm{Al}} \cdot 2750 \, \frac{\mathrm{cm}^3 \ \mathrm{bar}}{\mathrm{g}} \\ &= \epsilon_{\mathrm{m}} \cdot \frac{\left[1-(1-\rho_{\mathrm{rel}}) \cdot \frac{d}{d_{\mathrm{core}}}\right]}{(1-\rho_{\mathrm{rel}}) \cdot \frac{d}{d_{\mathrm{core}}}} \cdot \rho_{\mathrm{Al}} \cdot 2750 \ \frac{\mathrm{cm}^3 \, \mathrm{bar}}{\mathrm{g}}. \end{aligned} \tag{B.7}$$

B.2 Mean Cell Diameter

In the following, the 2D and 3D mean cell diameter are referred to as D_{2D} and D_{3D}, respectively. In order to obtain local information about the cell diameter, the 3D cell diameter is estimated from 2D μCT data by determining the 2D mean cell diameter and weighting it with an appropriate factor to account for the 3D effect.

B.2.1 2D Mean Cell Diameter

D_{2D} is determined by exploring 2D μCT information. It is supposed that the cell structure may be described by a mean cell diameter. In addition, the cells are assumed to show a perfect round shape. With these assumptions and the 2D μCT information, which comprises the total area A_{tot}, the total material area A_{mat} contained in A_{tot} and the internal gas-material interface length L_{tot} within A_{tot}, it is possible to estimate the mean cell diameter D_{2D} as follows:

$$D_{2D} = 4\,\frac{A_{tot} - A_{mat}}{L_{tot} - 4\,\sqrt{A_{tot}}\,\frac{A_{mat}}{A_{tot}}}. \tag{B.8}$$

The second term in the denominator takes into account that the boundaries of the 2D area are also included in L_{tot} and have to be subtracted. The factor $\frac{A_{mat}}{A_{tot}} = \rho_{rel}$ accounts for the fact that the boundary length decreases with decreasing relative density.

B.2.2 3D Mean Cell Diameter

The mean cell diameter in 2D is a mean value, see Fig. B.1:

$$D_{2D} = \frac{2}{D_{3D}} \int_0^{D_{3D}/2} D(x)dx. \tag{B.9}$$

With

$$x = \frac{D_{3D}}{2} \cdot \cos\alpha \qquad \text{and} \qquad D = D_{3D} \cdot \sin\alpha \tag{B.10}$$

and

$$dx = -\frac{D_{3D}}{2} \cdot \sin\alpha \; d\alpha, \tag{B.11}$$

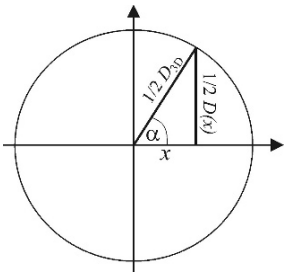

Figure B.1: Correlation between 2D and 3D mean cell diameter. The 2D diameter is a mean value due to different intersection planes.

the integration with respect to x is replaced by an integration over the angle α:

$$\begin{aligned} D_{2D} &= -\frac{2}{D_{3D}} \int_{\frac{\pi}{2}}^{0} \sin\alpha \cdot \sin\alpha \cdot \frac{D_{3D}^2}{2}\, d\alpha = D_{3D} \int_{0}^{\frac{\pi}{2}} \sin^2\alpha d\alpha \\ &= D_{3D} \left[\frac{1}{2}\alpha - \frac{1}{4}\sin 2\alpha\right]_{0}^{\pi/2} = \frac{\pi}{4} \cdot D_{3D} \end{aligned} \tag{B.12}$$

B.3 Curvature Calculation in 2D: Template Sphere Model

The template sphere model is based on the observation that the curvature can be expressed in leading order by the portion of fluid volume enclosed by a template sphere centered on the interfacial points, see Fig. B.2 [223]:

$$\kappa = \frac{1}{R} = \frac{3\left(\frac{\pi r^2}{2} - A\right)}{r^3}. \tag{B.13}$$

where A is the area in the sphere covered with fluid.

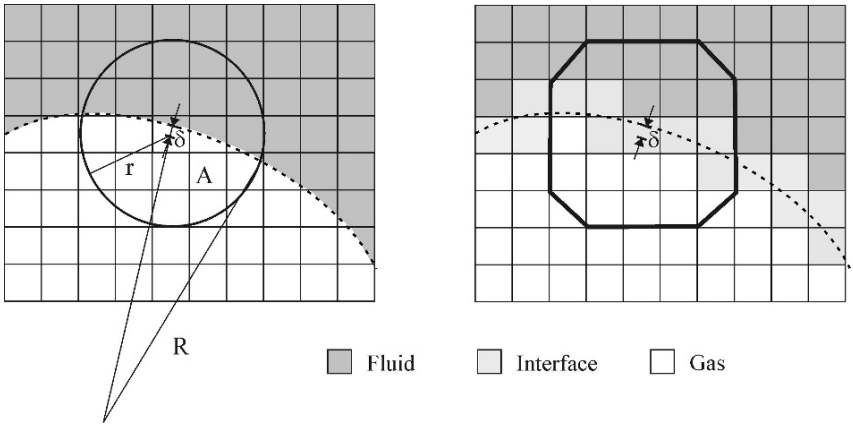

Figure B.2: Template sphere model. *Left*: Continuum situation. *Right*: Discrete lattice with 25-neighborhood. The distance between the center of the template sphere and the actual interface is referred to as δ.

On a discrete LB lattice, only neighborhoods which are similar to spheres, e. g. 25- or 36-neighborhood, can be defined. In order to reproduce the curvature as accurate as possible, Eq. (B.13) is modified in order to take into account that the interfacial point does generally not coincide with the center of the interface cell. With the distance between the center of the template sphere and the actual interface denoted with δ, the modification of Eq. (B.13) reads [223],

$$\kappa = \frac{1}{R} = \frac{3\left(\frac{\pi r^2}{2} - 2\,r\,\delta - A\right)}{r^3}. \tag{B.14}$$

A 25-neighborhood where the outermost cells are weighted by a factor 1/2 has shown to represent the best trade-off between accuracy and computational work.

Generally, the mean curvature of a bubble is quite well reproduced. However, there might be strong fluctuations between neighboring interface cells especially in diagonal direction or before or after cell state transformation.

C Material Parameters

For the numerical simulation the material parameters, ν, σ, ρ, g, S, D, have to be expressed in dimensionless form. A time scale Δt, a length scale Δx, a mass scale Δm, and a scale for the amount of substance Δn have to be defined. The dimensionless quantities are marked with $\star$. The material parameters follow by multiplying the dimensionless quantities with the relevant scales:

$$\nu^\star = \nu \frac{\Delta t}{\Delta x^2} \quad \text{and} \quad \tau^\star = \frac{6\ \nu^\star + 1}{2} \tag{C.1}$$

$$\sigma^\star = \sigma \frac{\Delta t^2}{\Delta m} \tag{C.2}$$

$$\rho^\star = \rho \frac{\Delta x^3}{\Delta m} \tag{C.3}$$

$$g^\star = g \frac{\Delta t^2}{\Delta x} \tag{C.4}$$

$$S^\star = S \frac{\Delta x^3}{\Delta n} \sqrt{\frac{\Delta x\, \Delta t^2}{\Delta m}} \tag{C.5}$$

$$Q^\star = Q \frac{\Delta x^3}{\Delta n} \tag{C.6}$$

$$D^\star = D \frac{\Delta t}{\Delta x^2} \quad \text{and} \quad \tau_{\mathrm{D}}^\star = \frac{4\ D^\star + 1}{2} \tag{C.7}$$

In addition, pressure and density are correlated by

$$p = \frac{1}{3}\ \rho\ \frac{\Delta x^2}{\Delta t^2}. \tag{C.8}$$

For a given length scale Δx, the time scale Δt is determined by the pressure p and the density ρ. Time scale and length scale may not be varied independently!

Table C.1: Physical properties of liquid aluminum and magnesium [123].

	ρ (Mg/m^3)	η (mPa· s)	ν (m^2/s)	σ (N/m)	v_{ca} (m/s)
Al	2.38	1.20	$0.50{\cdot}10^{-6}$	0.91	777
Mg	1.59	1.25	$0.79{\cdot}10^{-6}$	0.56	448

Table C.2: Numerical experiment Sim1:
Global parameters: $\Delta x = 10$ μm, $\Delta t = 0.3 \cdot 10^{-6}$ s, $\Delta m = 2.4 \cdot 10^{-9}$g, $p = 9$ bar, $\rho = 2.38$ g/cm^3, $g = 0.$, $S^\star = 0.1$, $Q^\star = 10^{-5}$, $D^\star = 0.025$, expansion velocity $v = 0.005$ m/s, 200×200 cells, 75 bubble nuclei, n_{steps}: $8 \cdot 10^4$, periodic boundary conditions in x-direction.

Experiment	$\tau^\star$	$\sigma^\star$	$\nu^\star/\sigma^\star$	v_{ca} (m/s)	Ca
Sim1	0.55	0.03	0.55	61.7	$0.82 \cdot 10^{-3}$

Table C.3: Numerical experiments Sim2 - Sim6:
Global parameters: $\Delta x = 25$ μm, $\Delta t = 2.25 \cdot 10^{-6}$ s, $\Delta m = 3.75 \cdot 10^{-8}$ g, $p = 1$ bar, $\rho = 2.38$ g/cm^3, $g = 0.$, $S^\star = 0.1$, $Q^\star = 10^{-5}$, $D^\star = 0.025$, $d_\pi^\star = 3\Delta x$, expansion velocity $v = 0.001$ m/s, 400×400 cells, 300 bubble nuclei, n_{steps}: $1.4 \cdot 10^5$, periodic boundary conditions in x-direction.

Experiment	$\tau^\star$	$\sigma^\star$	$\nu^\star/\sigma^\star$	v_{ca} (m/s)	Ca
Sim2	0.6	0.003	10.79	1.04	0.0097
Sim3	0.7	0.0018	36.00	0.31	0.0322
Sim4	2.5	0.003	171.37	0.066	0.1533
Sim5	5.5	0.0006	2699.99	0.0042	2.42
Sim6	1.7	0.00006	5555.55	0.002	4.98

Table C.4: Numerical experiments Sim7 - Sim8:
Global parameters: $\Delta x = 25\ \mu$m, $\Delta t = 2.25 \cdot 10^{-6}$ s, $p = 1$ bar, $\rho = 2.38$ g/cm^3, $g = 0.$, $S^\star = 0.1$, $Q^\star = 10^{-5}$, $D^\star = 0.025$, $d_\pi^\star = 3\Delta x$, 400×600 cells, n_{steps}: $5 \cdot 10^5$, periodic boundary conditions in x-direction.

Experiment	$\tau^\star$	$\sigma^\star$	$\nu^\star/\sigma^\star$	v_{ca} (m/s)	Ca	$\frac{\Pi_{\max}}{\sigma/\Delta x}$
Sim7	0.7	0.0018	36.00	0.31	0.0322	0.45
Sim8	0.7	0.0018	36.00	0.31	0.0322	1.35

Table C.5: Numerical experiments Sim9 - Sim10:
Global parameters: $\Delta x = 25\ \mu$m, $\Delta t = 2.25 \cdot 10^{-6}$ s, $p = 1$ bar, $\rho = 2.38$ g/cm^3, $g = 0.$, $S^\star = 0.1$, $Q^\star = 10^{-5}$, $D^\star = 0.025$, 400×500 cells, $n_0 = 25/\text{mm}^2$, periodic boundary conditions in x-direction.

Experiment	$\tau^\star$	$\sigma^\star$	$\nu^\star/\sigma^\star$	v_{ca} (m/s)	Ca	$\frac{\Pi_{\max}}{\sigma/\Delta x}$
Sim9	0.7	0.0018	36.00	0.31	0.0322	0.45
Sim10	5.5	0.0006	2699.99	0.0042	2.42	0.9

Table C.6: Numerical experiments Endo1 + Endo2:
Global parameters: $\Delta x = 2\ \mu$m, $\Delta t = 2.25 \cdot 10^{-6}$ s, $\Delta m = 1.9 \cdot 10^{-11}$ g, $p = 64$ mbar, $\rho = 2.38$ g/cm^3, $\Pi = 0.$, $g = 0.$, expansion velocity $v = 0.001$ m/s, 800×700 cells, $n_0 = 156/\text{mm}^2$, periodic boundary conditions in x-direction.

Experiment	$\tau^\star$	$\sigma^\star$	$\nu^\star/\sigma^\star$	v_{ca} (m/s)	Ca	V_{rel}
Endo1	0.65	0.01	65.00	0.014	0.071	0.34
Endo2	0.65	0.01	65.00	0.014	0.071	0.68

Symbols

Abbreviations

CA	Cellular automata
CFD	Computational fluid dynamics
IFM	Integral foam molding
HP-IFM	High pressure integral foam molding
JMA	Johnson-Mehl-Avrami
LBE	Lattice Boltzmann equation
LBM	Lattice Boltzmann method
LGA	Lattice gas automata
LP-IFM	Low pressure integral foam molding
NSE	Navier-Stokes equations
PM foam	Foam produced by powder compacts
μCT	Micro computer tomography

Mathematic Symbols

∂	Partial derivative
δ_{ij}	Kronecker delta function
∇	Nabla Operator
$\delta^{(3)}$	Delta function in three dimensions

Greek Symbols

δ	Mean material thickness
δ^{diff}	Diffusion length
Δm	Mass scale
Δn	Scale for the amount of substance
Δx	Lattice spacing = length scale
Δt	Time step = time scale
ϵ	Volume fraction of fluid within a cell
Γ	Liquid-gas interface
Π	Disjoining pressure
α	Spatial index
β	Spatial index
η	Dynamic viscosity
κ	Curvature
ν	Kinematic viscosity
ϕ	Phase ratio
ρ	Density
ρ_{rel}	Relative density
ρ_{F}	Foam density
σ	Surface tension
τ	Relaxation time
τ_{D}	Relaxation time for diffusion

Latin Symbols

B_{F}	Beam stiffness
Ca	Capillary number
D	Mean cell diameter
D	Gas diffusion constant
D_{p}	Particle diameter
E	Elastic modulus
I	Moment of Inertia
Q^{-1}	Internal friction
R	Gas constant
Re	Reynolds number
R_{Pl}	Curvature radius of the Plateau border
R_{p}	Particle radius
S	Sieverts' constant
T	Temperature
V_i	Volume of cell (bubble) i
V_{rel}	Relative particle volume fraction

Latin Symbols

c	Gas concentration
c_Π	Disjoining pressure constant
c_s	Speed of sound
d	Cell wall thickness
d_{crit}	Critical cell wall thickness
d_π	Range of the disjoining pressure
f	Phase fraction
$\boldsymbol{g}$	Gravitational acceleration
k_{B}	Boltzmann constant
m	Mass
m	Spacial dimension, 2D: $m = 2$, 3D: $m = 3$
n	Amount of substance
n	Evolution exponent
n_0	Nuclei density
n_{steps}	Total number of time steps
p	Pressure
p_0	Atmospheric or external pressure
p_i	Pressure of cell (bubble) i
p_{cast}	Casting pressure
p_{dwell}	Dwell pressure
t	Time
$\mathbf{v}$	Velocity
v_{ca}	Capillary velocity
$\mathbf{x}$	Spatial coordinate

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