Effect of Surface Tension in the Gas-Dough Interface on the Rheological Behavior of Dough

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ABSTRACT

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Physical and geometric objections are raised to a recently proposed model for estimating the contribution of surface tension in the gas-dough interface to the elastic resistance of dough. An alternative model is proposed, in which the deformation of gas cells is equal to that of the dough. According to this model, the contribution of surface tension to the modulus of dough is much smaller than in the original model, particularly in doughs with little occluded gas. The alternative model predicts that this

contribution is of little importance, possibly with the exception of doughs in an advanced stage of fermentation. Surface tension does not contribute to the viscous resistance of dough to an external load. Its contribution to the excess pressure in gas cells may, however, be larger than is required in a fermenting dough to overcome the viscous resistance of the liquid dough phase.

The behavior of disperse systems is affected by the properties of the interface in them. Because a fermenting dough is a dispersion of gas in a continuous dough phase, its behavior is affected by the number and size of gas cells and by the surface tension in the gas-dough interface. This article contains some theoretical considerations on the relations between these quantities and dough behavior. It is a critical reaction to and an expansion of a recent communication by Carlson and Bohlin (1978) on the same subject.

This article disregards the fact that the continuous dough phase is a disperse system in itself, namely of starch granules in a continuous protein-lipid-water phase.

The discussion is divided into two parts, dealing with elasticity and viscosity, for the following reasons. If an elastoviscous material is loaded, its deformation is partly recoverable or elastic and partly irrecoverable or viscous; the distinction between the two parts can be made by removal of the load. The viscous part of the deformation increases with increasing duration of the load; the elastic part does not increase or hardly does so. As a consequence, after loading for a short time, the deformation is predominantly elastic, whereas after loading for a long time, it is predominantly viscous. Whether a time interval is to be considered short or long depends on its length in comparison to the relaxation time of the material examined. The relaxation time of dough is of the order of 10 s (Bloksma 1971). Therefore, brief experiments can be designed, in which the dough behaves approximately as an elastic solid, and other ones of long duration, in which the dough behaves approximately as a viscous liquid. Fermentation in a bakery belongs to the latter category.

In the two parts of this article, therefore, the gas-free part of the dough is termed "solid dough phase" and "liquid dough phase," respectively. These different terms refer to the same material under different experimental conditions. Its volume fraction is indicated by v_s and v_l. Different indexes are used to make various equations clearer; however, v_s and v_l have the same meaning.

The part on elasticity discusses the model of Carlson and Bohlin (1978) as well as an alternative model, which, in the author's opinion, is more probable. The part on viscosity will discuss the contribution of surface tension to the viscous resistance to external loads, as well as its contribution to the excess pressure in the gas cells in a fermenting dough.

In this article, I assume that the surface tension in the gas-dough interface is not affected by changes in the area of the interface. Surface tension and (Gibbs or Helmholtz free) interfacial energy per unit interface area are considered equal (Davies and Rideal 1963); the symbol γ is used for both of them.

Other symbols are explained in Table I.

ELASTICITY

Model of Carlson and Bohlin

An essential assumption in the model of Carlson and Bohlin (1978) is that the gas phase and the solid phase are arranged in

series. This means that the stresses in both phases are equal and that the strain in the solid phase is negligible. Consequently, the strain in the gas phase is larger than the average strain of the complete composite material, the more so as the relative volume of the gas phase is smaller. This is expressed in equation 5 of Carlson and Bohlin, which can be written as

$$(\exp \epsilon_g) - 1 = \frac{(\exp \epsilon) - 1}{v_g} \tag{1}$$

TABLE I
Explanation of Symbols

Symbol	Definition
A	Surface area of one gas cell (L^2)[a]
a	Distance between centers of gas cells (L)
B	Surface area of bottom of tin and of piston in Fig. 4 (L^2)
b	$[1 - \exp(-3\epsilon_g)]^{1/2}$
c	$(2^{1/2}3/\pi)^{1/3} = 1.105$
E	Apparent Young modulus of dough with gas cells $(M \cdot L^{-1} \cdot T^{-2})$
E_i	Contribution of surface energy to the apparent Young modulus of dough $(M \cdot L^{-1} \cdot T^{-2})$
E_s	Young modulus of solid dough phase $(M \cdot L^{-1} \cdot T^{-2})$
h	Height of piston in model of Fig. 4 (L)
h_0	Height of piston in model of Fig. 4, if dough does not contain any gas (L)
l	Length after extension (L)
l_0	Original length (L)
n	Number of gas cells per unit volume of solid or liquid dough phase (L^{-3})
P	Pressure in gas cells $(M \cdot L^{-1} \cdot T^{-2})$
P_0	Atmospheric pressure $(M \cdot L^{-1} \cdot T^{-2})$
p	h/h_0 at zero time
q	Increase of dough volume during proof per unit time and per unit volume of gas-free dough (T^{-1})
r	Radius of spherical or cylindrical gas cells (L)
t	Time (T)
v_g	Gas-total volume ratio
v_l	Liquid-total volume ratio
v_s	Solid-total volume ratio
W	Stored energy per unit volume of dough $(M \cdot L^{-1} \cdot T^{-2})$
W_i	Surface energy per unit volume of dough $(M \cdot L^{-1} \cdot T^{-2})$
W_s	Energy stored in solid phase per unit volume of dough $(M \cdot L^{-1} \cdot T^{-2})$
γ	Surface tension or surface energy per unit area $(M \cdot T^{-2})$
ϵ	Extension of the composite material
ϵ_g	Extension of gas cells
ϵ_s	Extension of solid phase
$\dot{\epsilon}_z$	Rate of extension $d\epsilon/dt$ in the z-direction (T^{-1})
μ_i	Contribution of surface tension to the apparent coefficient of viscous traction in the equation for the the excess pressure in gas cells $(M \cdot L^{-1} \cdot T^{-1})$
μ_l	(Trouton) coefficient of viscous traction of liquid dough phase $(M \cdot L^{-1} \cdot T^{-1})$
σ_j	Tensile stress in j-direction in liquid dough phase $(M \cdot L^{-1} \cdot T^{-2})$

[a] Dimension of mass (M), length (L), or time (T).

in which $v_g < 1$. The difference in notation between the article by Carlson and Bohlin and this one is explained in Appendix I. The model of Carlson and Bohlin is illustrated by Fig. 1. Spherical gas cells in the unstretched dough are converted into highly elongated ellipsoids.

A number of objections to this model can be raised.

Equation 1 of Carlson and Bohlin and, consequently, their equation 5 imply that the gas phase transfers a tensile stress, whereas, as a result of their negligible shear modulus, gases can only transfer hydrostatic pressure.

In the derivation of equation 5, the authors ignore the fact that, at constant volume, the highly extended gas parts of their rectangular section must contract laterally; this is in contrast to the nonextended solid parts. If so, a rectangular shape can be maintained only if solid material is displaced from the solid parts to the sides of the gaseous parts. This corresponds to a displacement of solid material from position A to position B in Fig. 1. What could be the driving force for such a displacement?
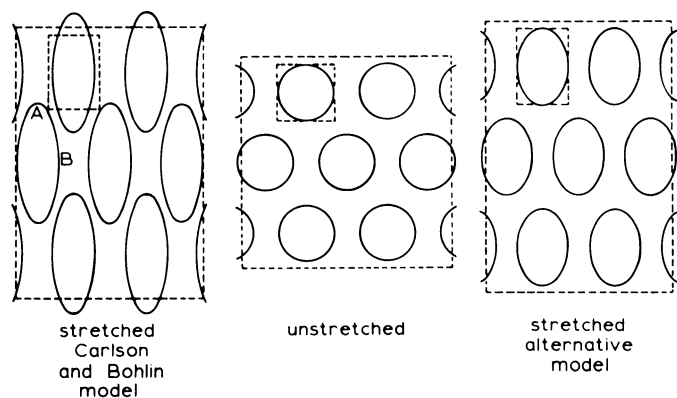

Fig. 1. Deformation of spherical gas cells upon uniaxial stretching in the model of Carlson and Bohlin (1978) and in an alternative model. The drawings are on scale for $v_g = 0.28$ and $\epsilon = 0.25$.
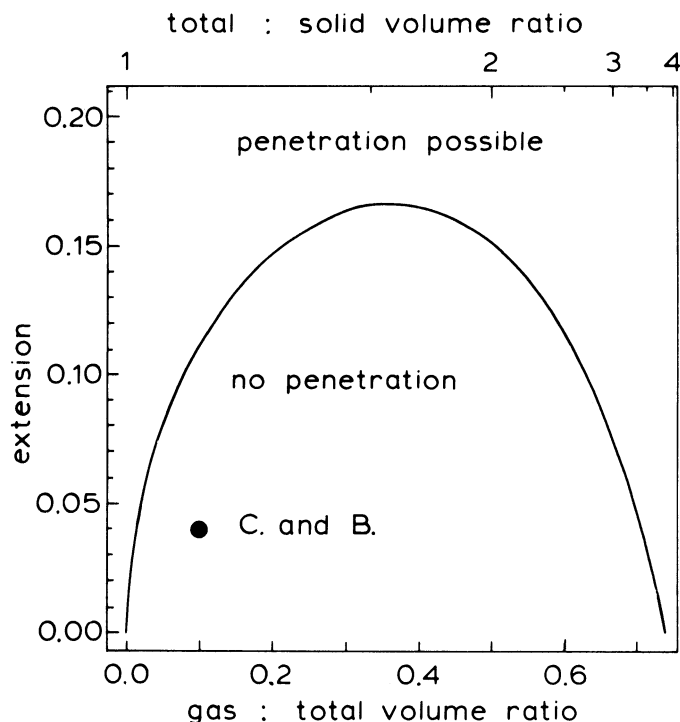

Fig. 2. Combination of relative volume of occluded gas and extension that, in the model of Carlson and Bohlin, do not or can lead to mutual penetration of gas cells. The dot marked "C. and B." represents the example for which Carlson and Bohlin (1978) calculated the contribution of surface energy to dough elasticity.

The left part of Fig. 1 shows that the relatively large extension of the gas cells greatly reduces the shortest distances between them. If, in this figure, the same extension had been applied in a direction making an angle of 30° with the vertical, ie, along a line connecting centers of adjacent spheres, the resulting ellipsoids would have penetrated one another. The combinations of relative gas volume and extension that may lead to such a mutual penetration can be derived as follows. The assumption is made that, in the undeformed state, spherical gas cells with a given radius (r) are arranged hexagonally or cubically with a given distance (a) between the centers of adjacent cells. In the deformed state, half of the long axis of the ellipsoid gas cells is $r \cdot \exp \epsilon_g$. If the direction of stretching is parallel to lines connecting centers of adjacent cells, the distance between the centers of adjacent cells has become $a \cdot \exp \epsilon$. If

$$2r \cdot \exp \epsilon_g > a \cdot \exp \epsilon \qquad (2)$$

penetration is possible, dependent on the direction of stretching. By substitution of equations 1 and 30 (Appendix II), equation 2 can be transformed into

$$\epsilon > \ln \frac{1 - v_g}{1 - v_g^{2/3}/c} \qquad (3)$$

The area in which this inequality holds is shown in the top part of Fig. 2. Only in the restricted bottom part can the model be applied without the risk of geometric accidents. The shape of the curve in Fig. 2, with $\epsilon = 0$ both at $v_g = 0$ and $v_g = 0.74$, can be explained as follows. If $v_g = c^{-3} = 0.74$, the spherical gas cells are already in contact one with another in the undeformed state (Appendix II); any deformation, however small, may cause mutual penetration. If v_g approaches zero, the deformation of the gas cells becomes excessively large even at a moderate overall deformation; this is a consequence of equation 1. Then, the ellipsoidal gas cells that result from the deformation are excessively long. This is a justification for Carlson and Bohlin's restriction that the gas-total volume ratio should not be too low.

In addition to mutual penetration, gas cells near the boundaries may, upon extension, also protrude from the test-piece. This is also illustrated by the left part of Fig. 1.

Alternative Model

Carlson and Bohlin have arbitrarily considered a section of material in the direction of stretching. If, however, one considers a section perpendicular to the direction of stretching, a more logical model is that the two phases are arranged in parallel, that their strains are equal, and that the deviatory stress in the gas phase is negligible. With this assumption,

$$\epsilon_g = \epsilon_s = \epsilon \qquad (4)$$

instead of equation 1. The stretched material is shown in the right part of Fig. 1. The objections to the model of Carlson and Bohlin raised in the preceding section do not apply to the alternative model.

A model that in some positions corresponds with that of Carlson and Bohlin and in other positions with the alternative model might approach reality more closely than either of these models alone. Its elaboration is, however, beyond the mathematical ability of the author. Therefore only the behavior of the alternative model is investigated.

For an estimation of the contribution of surface energy to the elastic resistance, the relation between the extension, the Young modulus (E) of a linear material, and the stored energy (W) per unit volume

$$W = \frac{1}{2} E \epsilon^2 \qquad (5)$$

will be used. In a linear material, the deformation is proportional to the stress, and the Young modulus is independent of stress and deformation.

The energy is composed of two parts, a part required to deform the solid phase and a part required to increase the surface area of the gas cells.

$$W = W_s + W_i \qquad (6)$$

The energy required to deform the gas phase at constant volume is considered negligible.

The energy to deform the solid phase refers to a volume (v_s). By analogy to equation 5:

$$W_s = v_s \cdot \tfrac{1}{2} E_s \epsilon_s^2 \qquad (7)$$

The number of gas cells per unit volume of dough is $v_s \cdot n$. Upon stretching, the surface area of each cell increases from $4\pi r^2$ to $A(\epsilon_g)$; that is the surface area of an ellipsoid with one axis $r \cdot \exp(\epsilon_g)$ and two axes $r \cdot \exp(-\epsilon_g/2)$. The energy required per unit volume of dough is

$$W_i = v_s n[A(\epsilon_g) - 4\pi r^2]\gamma \qquad (8)$$

Substitution of equation 39 (Appendix III) into equation 8 and combination with equations 5–7 lead to

$$E = v_s\left[(\epsilon_s/\epsilon)^2 E_s + \frac{16\pi}{5} \cdot (\epsilon_g/\epsilon)^2 n r^2 \gamma \left(1 - \frac{5}{21} \cdot \epsilon_g \ldots\right)\right] \qquad (9)$$

Equation 9 reveals the extent to which the model of Carlson and Bohlin assigns greater importance to the surface energy than does the alternative model. In their model, $\epsilon_g/\epsilon \simeq 1/v_g$ (equation 29, Appendix I), and in the alternative model, $\epsilon_g/\epsilon = 1$ (equation 4). In the Carlson and Bohlin model, the contribution of surface energy to the modulus is larger than in the alternative model by a factor of approximately $1/v_g^2$ ($v_g < 1$); with 10% occluded gas, this is a factor of 100.

If the alternative model is correct, equation 4 can be substituted into equation 9 with the result

$$E = v_s\left[E_s + \frac{16\pi}{5} \cdot n r^2 \gamma \left(1 - \frac{5}{21} \cdot \epsilon \ldots\right)\right] \qquad (10)$$

The factor v_s accounts for the fact that the contribution of the solid phase to the elastic resistance of the dough is proportional to its volume fraction; it is reduced as the dough contains more gas.

The right side of equation 10 still contains ϵ. This means that, even if the elasticity of the solid phase is linear, the elasticity of the dough with gas cells is nonlinear. For small deformations, ie, if $\epsilon \ll 21/5$, the nonlinear terms with ϵ and higher powers of ϵ can be neglected.

$$E = v_s\left[E_s + \frac{16\pi}{5} \cdot n r^2 \gamma\right] \qquad (11)$$

For an estimate of the relative importance of the contribution of surface energy, elimination of one more variable from equation 11 is useful. During fermentation, the gas cells grow but their number remains constant (Baker and Mize 1941). Therefore, by means of equation 32 (Appendix II), we write r as a function of n and v_g; in this operation, the gas cells are assumed to be of uniform size. Substitution of the result into equation 11 and combination with equation 26 (Appendix I) lead to

$$E = (1 - v_g)(E_s + E_i) \qquad (12)$$

in which
$$E_i \equiv \frac{(2^8 3^2 \pi)^{1/3}}{5} \cdot \left[\frac{v_g}{1 - v_g}\right]^{2/3} \cdot n^{1/3} \gamma \qquad (13)$$

In Fig. 3, the ratio E_i/E_s is plotted versus v_g for various values of the parameter $n^{1/3}\gamma/E_s$. Logically, the relative contribution of surface energy to the elastic resistance increases as the dough contains more gas (at a given number of gas cells per unit volume of solid phase),

as the number of gas cells is larger, as the surface energy is larger, and as the modulus of the solid phase is smaller.

The result has been expressed in terms of a dimensionless variable parameter $n^{1/3}\gamma/E_s$ because information on the various relative quantities is yet insufficient. Nevertheless, an estimate of the parameter $n^{1/3}\gamma/E_s$ can be attempted. The Young modulus can be estimated fairly reliably. The Young modulus of an incompressible material is three times its shear modulus. The latter is approximately 3×10^3 kg·m^{-1}·s^{-2} (Bloksma and Nieman 1975); consequently, $E_s = 9 \times 10^3$ kg·m^{-1}·s^{-2}. From data of Carlson and Bohlin (1978), the number of gas cells is estimated to be $n = 1.5 \times 10^{11}$ m^{-3} (Appendix II); this value may depend very much on the mixer used. Because of the presence of surface-active proteins and lipids, surface energy will most probably be smaller than that of pure water; the latter is 7×10^{-2} kg·s^{-2} (Weast 1971). As a result, the parameter $n^{1/3}\gamma/E_s$ is probably smaller than 4×10^{-2}. Therefore the ratio E_i/E_s is expected to be smaller than 0.04 if $v_g = 0.1$ and smaller than 0.3 at the end of tin proof when $v_g \simeq 0.7$. If the estimates used are correct, the contribution of surface energy to the elastic resistance is of little importance, possibly with the exception of doughs in an advanced stage of fermentation. Why this conclusion is different from that of Carlson and Bohlin has been explained before.

The conclusion in the preceding paragraph is only apparently in contradiction to the opinion that doughs with a large volume fraction of occluded gas are more elastic than doughs with little gas. This opinion is based upon the observation of volume elasticity, that is, the phenomenon that the volume of a body is reversibly reduced by hydrostatic pressure. Occluded gas imparts volume elasticity to a dough; dough with gas has this property in common with a football.

Thus far we have only considered uniaxial extension. If the alternative model is applied to shear, a sphere is transformed into an ellipsoid with three different axes, one being longer and one being shorter than the original radius. No exact equation is available for the surface area of such a body. For small deformations, however, the area can be expanded in a series such as in equation 39 (Appendix III). By this procedure, the author has satisfied himself that the relative contribution of surface energy to the shear modulus is exactly the same as to the Young modulus.
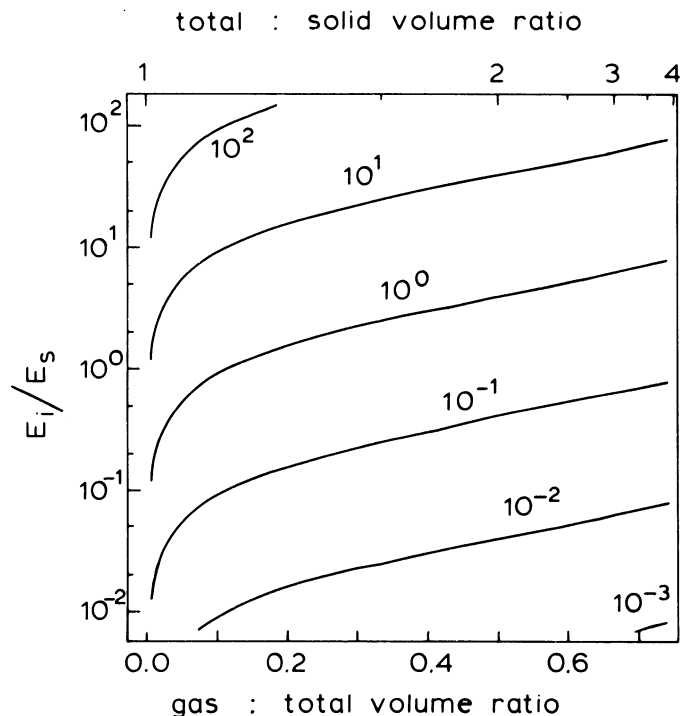

Fig. 3. Relative contribution of surface energy to the Young modulus of dough as a function of the quantity of occluded gas. The labels at the curves are values of the parameter $n^{1/3}\gamma/E_s$.

VISCOSITY

Effect on Viscosity Measurements

In a resting dough, surface energy or interfacial tension causes occluded gas cells to assume a spherical shape. Interfacial tension also restores their original spherical shape to gas cells that have become nonspherical by stretching, as in Fig. 1. This process only occurs if the continuous phase between the gas cells can flow; it does not occur in purely elastic materials. In this process, stored surface energy is dissipated in flow in the continuous phase; it is a mechanism of stress relaxation. If the rate of the external deformation is low in comparison to the rate of the relaxation process, the deviation from the spherical shape of the gas cells will be negligible. Then the presence of gas cells affects the viscosity in the same way as does the presence of solid particles to which the continuous liquid phase does not adhere. Surface energy does not contribute directly to the viscous resistance. At intermediate rates of external deformation, the presence of gas cells transforms a purely viscous liquid into an elastoviscous liquid. At high rates of deformation, the elastic resistance will dominate; then the considerations of the preceding sections apply.

Effect on Pressure in Gas Cells During Fermentation

Although surface energy does not contribute to the viscous resistance to an external load, it does contribute to the excess pressure in the gas cells. This contribution can be compared with the excess pressure that is the driving force of proof during fermentation.

For this purpose the model in Fig. 4 is used. In this model, the dough is enclosed in a prismatic tin under a frictionless piston. The gas is collected in a number of vertical cylinders with a uniform radius, each of them running from the bottom of the tin to the piston; the distribution of the gas makes this model different from that in an earlier publication (Bloksma 1962, Fig. 11). If the surface area of the bottom of the tin is B, the surface area of the cross sections of the gas cells is $v_g B$, and the total circumference of their cross sections $2v_g B / r$.

Forces acting on the piston are due to atmospheric pressure, tensile stress in the liquid dough phase, gas pressure in the cylindrical gas cells, and surface tension in the walls of these cells. Equilibrium of forces on the piston requires

$$BP_0 + v_l B \sigma_z - v_g BP + 2v_g B\gamma/r = 0 \qquad (14)$$

If no interfacial tension existed, the tensile stress in the liquid phase in the x-direction and y-direction would correspond to the pressure in the gas cells. The effect of gas pressure is reduced by surface tension. For cylindrical gas cells

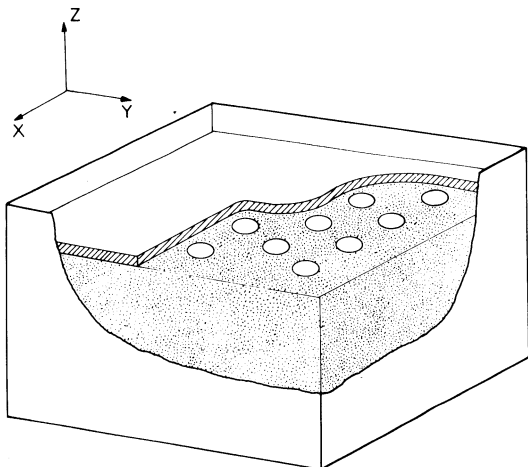$$\sigma_x = \sigma_y = -P + \gamma/r \qquad (15)$$

Fig. 4. Model for a fermenting dough.

The stress in the liquid dough phase can be considered a superposition of simple tension by a stress $(\sigma_z - \sigma_x)$ and hydrostatic pressure (σ_x). If the liquid phase is a Newtonian liquid with a (Trouton) coefficient of viscous traction μ_1,

$$\sigma_z - \sigma_x = \mu_1 \dot\epsilon_z \qquad (16)$$

in which $\dot\epsilon_z$ is the rate of stretching in the z-direction (Reiner 1956).

From equations 14–16 and the equation $v_l + v_g \equiv 1$ (equivalent to equation 26), it follows that

$$P - P_0 = v_l \mu_1 \dot\epsilon_z + (1 + v_g)\gamma/r \qquad (17)$$

The first term of the right side is also obtained if the earlier model (Bloksma 1962) is used, in which surface tension is ignored. The second term, describing the contribution of surface energy, has the factor γ/r in common with the equation for the excess pressure in a spherical gas cell in a resting liquid (Davies and Rideal 1963). The latter equation, however, contains a factor 2 instead of $(1 + v_g)$ in equation 17. This difference may be a consequence of the use of cylindrical gas cells in the present model. The first term of the right side of equation 17 is an estimate of hydrostatic pressure in the liquid phase near the gas-liquid interface. Hydrostatic pressure at the other side of a spherical interface will be $2\gamma/r$ higher. Therefore,

$$P - P_0 = v_l \mu_1 \dot\epsilon_z + 2\gamma/r \qquad (18)$$

is used for spherical gas cells.

For the application of equation 18, expression of the rate of extension in the rate of proof and in the volume ratio of occluded gas is necessary. The volume of the dough is assumed to increase at a constant rate. Then the height of the piston in the model in Fig. 4 can be written as

$$h/h_0 = p + qt \qquad (19)$$

in which h_0 is the height of the piston if the dough does not contain any gas. Therefore, $h/h_0 = 1/v_l$, and

$$\dot\epsilon_z = \frac{d \ln h}{dt} = qv_l \qquad (20)$$

Substitution of equations 20 and 32 (Appendix II) into equation 18 leads to

$$P - P_0 = (1 - v_g)^2 q(\mu_1 + \mu_i) \qquad (21)$$

in which $\qquad \mu_i \equiv \left[\dfrac{2^5 \pi}{3}\right]^{1/3} v_g^{-1/3} (1 - v_g)^{-5/3} \cdot \dfrac{n^{1/3}\gamma}{q} \qquad (22)$

In Fig. 5 the ratio μ_i/μ_1 is plotted versus v_g for various values of the parameter $n^{1/3}\gamma/q\mu_1$. At a given number of gas cells per unit volume of liquid phase, the quantity of occluded gas is much less important than if dough is considered an elastic material (Fig. 3); this is a result of the factors $v_g^{-1/3}$ and $(1 - v_g)^{-5/3}$ working in opposite directions. The first factor dominates only if v_g approaches zero, resulting in a sharp increase of μ_i/μ_1; then $1/r$ goes to infinity, as does the second term of equation 18. Logically, the relative contribution of surface energy increases as the number of gas cells is larger, as the surface tension is larger, as the rate of proof (q) is smaller, and as the coefficient of viscous traction (μ_1) is smaller.

For the same reason as with elasticity, the result has been expressed in terms of a dimensionless variable parameter, in this case $n^{1/3}\gamma/q\mu_1$. An estimate of this parameter can be attempted. The values of n and γ have already been discussed for the elastic resistance. During tin proof, the dough volume increases at a rate of about $3L/L$ of liquid dough phase per hour; this corresponds with $q = 8 \times 10^{-4}$ s^{-1}. The coefficient of viscous traction of an incompressible material is three times its coefficient of viscosity. The latter is approximately 1.6×10^5 kg·m^{-1}·s^{-1} at a rate of shear of

10^{-3} s^{-1} (Bloksma and Nieman 1975); consequently, $\mu_l = 5 \times 10^5$ kg·m^{-1}·s^{-1}. Combination of the various estimates leads to the conclusion that $n^{1/3}\gamma/q\mu_l$ is probably smaller than 0.9. Therefore μ_i/μ_l is expected to be smaller than 7 if $v_g = 0.15$ and smaller than 16 if $v_g = 0.6$. If the estimates used are correct, the contribution of surface tension to the excess pressure in the gas cells may be important.

Even though the contribution of surface energy to the excess gas pressure may be larger than that of the viscous resistance of the liquid dough phase, it is still small with respect to atmospheric pressure (1.0×10^5 Pa) or the total pressure in the gas cells. This is illustrated by Table II, which has been calculated by means of equations 21 and 22, the values of q and μ_l mentioned above, and with the assumption that $n^{1/3}\gamma/q\mu_l = 0.9$; most probably it therefore overestimates the effect of surface tension.

For a test of the theory developed in this section and for experimental studies on the role of surface tension, measurements of the excess pressure in gas cells would be an invaluable tool. Experiments with pressure gauges at the bottom of a vessel with a fermenting dough (Baker 1939; Matsumoto et al 1971, 1973) are, in the author's opinion, not suitable for this purpose; they measure hydrostatic pressure in the dough phase between the gas cells rather than in the gas cells. The procedure of Bailey (1955), which measures the increase in gas pressure in a closed vessel in which a dough is punched, appears more reliable; its result, 3×10^3 Pa with $v_g = 0.72$, is about a factor 3 higher than the (overestimated) value in Table II. The low ratio of excess to total pressure makes measurements of this type difficult.

APPENDIX I. DEFINITION OF EXTENSION

Whereas Carlson and Bohlin (1978) used the Cauchy definition of extension

$$\epsilon \equiv (1 - l_0)l_0 \tag{23}$$

in this paper the Hencky definition

$$\epsilon \equiv \ln l/l_0 \tag{24}$$

is used. The latter is more suitable for large viscous deformations. For small deformations, the difference between the two definitions is negligible.

With the Hencky definition, equation 4 of Carlson and Bohlin has the form

$$\exp \epsilon = v_g \cdot \exp \epsilon_g + v_s \cdot \exp \epsilon_s \tag{25}$$

By definition

$$v_g + v_s \equiv 1 \tag{26}$$

If the extension of the solid part is relatively small, ie, if

$$\epsilon_s \ll \frac{1 + \epsilon}{1 - v_g} \tag{27}$$

equations 25 and 26 can be simplified to

$$(\exp \epsilon_g) - 1 = \frac{(\exp \epsilon) - 1}{v_g} \tag{28}$$

If $\epsilon_g \ll 1$, equation 28 can be simplified to

$$\epsilon_g = \frac{\epsilon}{v_g} \tag{29}$$

in accordance with equation 5 of Carlson and Bohlin.

TABLE II
Excess Pressure in Gas Cells, Predicted by Theory[a]

Gas-Total Volume Ratio	Excess Gas Pressure (Pa)		
	Due to Viscous Resistance of Liquid Phase	Due to Surface Tension	Total
0.1	3.2×10^2	2.4×10^3	2.7×10^3
0.4	1.4×10^2	1.3×10^3	1.5×10^3
0.7	3.6×10^1	8.8×10^2	9.1×10^2

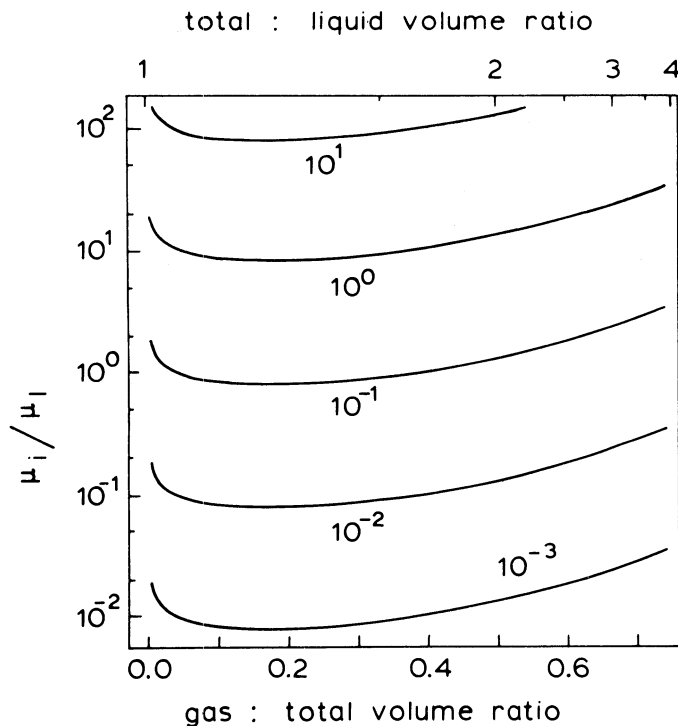[a]Numerical values used are given in the text.

Fig. 5. Relative contribution of surface tension to the excess pressure in gas cells as a function of the quantity of occluded gas. The labels at the curves are values of the parameter $n^{1/3}\gamma/q\mu_l$.
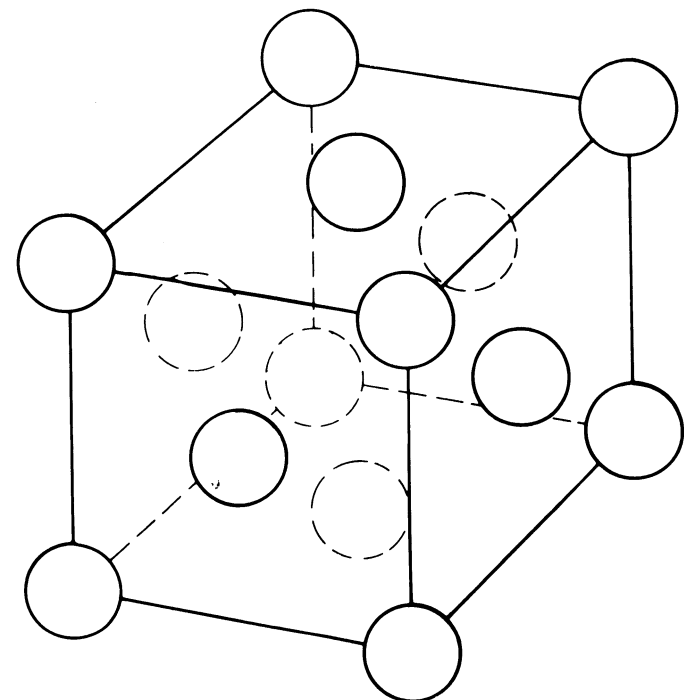

Fig. 6. Cubical array of spheres.

APPENDIX II. GEOMETRIC RELATIONS

Suppose that spherical gas cells with uniform radius are arranged cubically, that is, at the corners and in the centers of the faces of cubes (Fig. 6). If the distance between the centers of adjacent gas cells is a, the edges of the cube have a length $2^{1/2} \cdot a$. The volume of the cube is $2^{3/2} \cdot a^3$. Each cube shares the eight gas cells at the corners with eight other cubes and the six gas cells in the centers of faces with two other cubes. Therefore, the volume of one cube contains $8/8 + 6/2 = 4$ gas cells; their volume is $4 \cdot (4\pi/3) \cdot r^3$. Consequently

$$v_g = \frac{4 \cdot (4\pi/3) \cdot r^3}{2^{3/2} \cdot a^3} = c^{-3} \left[\frac{2r}{a}\right]^3 \quad (30)$$

in which $c \equiv (2^{1/2} \cdot 3/\pi)^{1/3}$ \quad (31)

For a hexagonal array, the same result is obtained. Close packing occurs if $2r/a = 1$, that is, if $v_g = c^{-3} = 0.74$.

If n spherical gas cells with radius r are present per unit volume of solid or liquid phase, then

$$n \cdot (4\pi/3) \cdot r^3 = \frac{v_g}{v_s} = \frac{v_g}{1 - v_g} \quad (32)$$

The order of magnitude of n can be estimated on the basis of data of Carlson and Bohlin (1978); with $v_g = 0.10$, they found an average gas cell radius of 56 μm. Substitution of these values into equation 32 results in $n = 1.5 \times 10^{11}$ m^{-3} or 150 per mm^3.

APPENDIX III. THE SURFACE AREA OF AN ELLIPSOID

The surface area of a prolate ellipsoid with one long axis $r \cdot \exp(\epsilon_g)$ and two short axes $r \cdot \exp(-\epsilon_g/2)$ is (Witting 1940)

$$A(\epsilon_g) = 2\pi r^2 \left[\exp(-\epsilon_g) + \exp(\epsilon_g/2) \cdot \frac{\arcsin b}{b}\right] \quad (33)$$

in which $b \equiv \left[1 - \exp(-3\epsilon_g)\right]^{1/2}$ \quad (34)

Note that b is only real if $\epsilon_g > 0$. Equation 33 is equivalent to the equation in Carlson and Bohlin's (1978) Appendix II.

Various parts of equation 33 can be written in the form of MacLaurin expansions, the first terms of which are

$$\exp(-\epsilon_g) = 1 - \epsilon_g + \frac{1}{2} \cdot \epsilon_g^2 - \frac{1}{6} \cdot \epsilon_g^3 \ldots \quad (35)$$

$$\exp(\epsilon_g/2) = 1 + \frac{1}{2} \cdot \epsilon_g + \frac{1}{8} \cdot \epsilon_g^2 + \frac{1}{48} \cdot \epsilon_g^3 \ldots \quad (36)$$

$$b^2 = 3\epsilon_g - \frac{9}{2} \cdot \epsilon_g^2 + \frac{9}{2} \cdot \epsilon_g^3 \ldots \quad (37)$$

$$\frac{\arcsin b}{b} = 1 + \frac{1}{2} \cdot \epsilon_g - \frac{3}{40} \cdot \epsilon_g^2 - \frac{39}{560} \cdot \epsilon_g^3 \ldots \quad (38)$$

Substitution into equation 33 results in

$$A(\epsilon_g) - 4\pi r^2 = 4\pi r^2 \left[\frac{2}{5} \cdot \epsilon_g^2 - \frac{2}{21} \cdot \epsilon_g^3 \ldots\right] \quad (39)$$

In the case of an oblate ellipsoid ($\epsilon_g < 0$), equation 33 has a different appearance, containing an inverse hyperbolic sine; equation 34 has to be replaced by another definition of its argument. If these equations are written in the form of MacLaurin expansions, the result is identical with equation 39. Therefore, this equation is valid irrespective of the sign of ϵ_g.

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