

The Topology of the Dispersed Phase in Gas Concrete

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Abstract: The process of bulk gas pore formation in cellular concrete mixtures is studied as well as the most probable pore size distribution in non-autoclave cellular concrete is obtained.

Key words: Cellular concrete • Pore formation • Pore distribution

INTRODUCTION

The size distribution of gas bubbles in the gas concrete has the same character as in the foam concrete, but with greater heterogeneity. At the phase interface, one of which is polar fluid at least, at the surface of the gas bubbles is formed an interfacial shell containing an excess amount of cations in comparison with the liquid phase. In the liquid phase bubbles surrounded counter ions and polar water molecules. Emergence of the double electric layer is result of necessity to reduce the surface energy and surface (interphase) tension. The local heterogeneity of gas agent distribution lead to heterogeneous pore size distribution in gas concrete and in any case, to collapse of bubbles and formation of bubbles torn porosity. The process of gas pore formation in gas concrete can be considered from the point of view of the formation of the double electric layer, the main principles of which are similar to the foam formation in foam concrete. The both pore formation processes are described by the same thermodynamic laws. Therefore, we consider the general pore formation process in cellular concrete mixtures and pore size distribution in foam and gas concrete.

When cellular concrete porization the following experimental and theoretical statutory provisions occur [1–6]:

- Poroused cement paste and hardened cellular concrete with standard strength class and average density of 300... 450 kg/m³ contain large air involved pores with dimension of 1... 2 mm and the micropores and submicropores;
- Cement paste also contains some of the gas phase adsorbed and clamped between surface defects in the solid phase, as well as air filled gas babbles introduced with mixing water;
- When adding with water into cement the saturated steam bubbles are formed on the cement particles surface in the center of hydration. It is initiated by reducing the volume of the system "cement–water" forming the contraction porosity. But the formation of gel and cement solidification submicropores are formed, which are not overgrown by new formations and produce gel porosity;
- In the cement paste or cement suspension the water gas bubbles is closed with water shells forming a films on the surface thereof of low surface tension

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and containing an excess amount of cations in comparison with the liquid phase as well as polar part of the molecules of surface-active constituent (SAC). In result of one-sided orientation of SAC-molecules, the polar part is blocked in the shell and the non-polar part is blocked in the gas phase bubbles. By reducing the surface tension in the interphase layer, its film is saturated with excess amount of cations and the anions in the liquid phase as well as the polar water molecules form a counter ion extended layer to generate a double electric layer;

- Reducing of high surface tension in the films compared with the surface tension of the water solution leads to an increasing of air bubbles in the cement paste up to a certain equilibrium sizes. The smaller gas bubbles initiate more air pressure and water vapor therein and the greater the surface tension of the films. Stability of steam-air bubbles is due to the presence of the electrostatic field formed by the same charges their shells surface like oil-in-water emulsion. The saturation concentration of the interfacial layer with cation or SAC depends on many factors, in particular, on the surface area of the bubbles and the area of surface for hydrophilic molecule zone;
- Deformation of the large bubbles shells leads to a redistribution of the liquid phase in the films and possibility of their merger similar large mercury droplets. Fusion of small-size gas bubbles is difficult due to the increased pressure inside of them and the elastic nature of their collision when stirring of cellular concrete mixtures;
- In the plane of the cross section and on the surface of fracture of the samples of porous concrete the distribution is observed in certain sizes of large and small pore, the shape of which ones is close to spherical.

Main Part: The problem of the pore size distribution (fractionation) in the cellular concretes refers to capillary phenomena in which there is a change in the chemical potential and the pressure of deformed surface in interfacial membranes of steam-gas and steam-air bubbles.

In the article two bulk phases L and G are studied. These ones are divided by spherical surface being in equilibrium at $T=\text{const}$, for example, a gas bubble (G) in the liquid medium (L) or a drop of liquid (L) in the vapor

phase (G). At equilibrium state, variations of changes in the surface dS and bubble volume dV without transferring substance from one phase to another one is possible, i.e. the molar composition of the phases does not change, $dn_i=0$. If the volume of the air bubble V is increased by dV and the surface of its shell S is increased to dS than the general equation for the change in the Helmholtz free energy for the total system is following [7]:

$$dF = -P_L \cdot dV_L - P_G \cdot dV_G + \sigma \cdot dS \quad (1)$$

At constancy of total volume of the system as well as n_i and T , $dF = 0$, while from the equation (1) considering that $dV_L = -dV_G$, we can define

$$P_L - P_G = \sigma \cdot d \cdot S / dV_L \quad (2)$$

Thus: $P_L > P_G$. Taking into account that $V_L = 4/3(\pi R^3)$ and $S = 4\pi R^2$, where R is radius of the bubble surface tension of (drop), we get following:

$$dS / dV_L = 2 / R \quad (3)$$

As $R = r / \cos\theta$, where r is radius of the bubble (drops), θ (theta) = 0° is wetting angle of the surface tension of the bubble curvature, then $R=r$.

The substitution of (2) and (3) into (1) gives the Laplace–Jung equation:

$$P_L - P_G = 2 \cdot \sigma / r \quad (4)$$

This equation indicates the pressure difference between the bulk phase of bubbles increases with increasing of σ and decreasing of r .

Excess (capillary) pressure $\Delta P = P_L - P_G$ is a result of a force of a surface “film” holding down the phase disposed on the concave side of the interface. Since this force equal and opposite reaction, the contraction force causes an increase in pressure in phase at the concave side. Regardless of the state of phase aggregation concave pressure surface side is always greater than the convex side in equilibrium state [7].

Next, the change of the vapor pressure of the gas bubble in the liquid phase, or a low pressure under drop in the gas phase is considered. As the pressure in the bulk phase with the deformed surface is changed, the change to be expected as the chemical potential μ and therefore the vapor pressure p , ($d\mu / dp \neq 0$).

During the formation of the curvature of the surface bubbles or droplets in a monocomponent ($i=1$) two-phase (L, G) system at T, S and $n_i = const$:

$$d\mu_i = v_i \cdot dp = v_i \cdot d(2 \cdot \sigma / r) \quad (5)$$

Take the molecular weight of the gas-vapor mixture v_i in first approximation as a constant. Thus, for water at $r = 10^{-7}$ m and $\sigma = 73 \cdot 10^{-3}$ N/m (73 erg/cm²) in equation (3) we define that: $p = 1,5$ MPa (15 atm). This value is low compared to the inner water pressure ($\approx 10^3$ MPa). In this case, the integration of a flat surface ($r = \infty$) to deformed (r) can be taken v_i outside the integral.

$$\int_{\mu_i^0}^{\mu_i^r} d\mu_i = v_i \cdot \int_{r=\infty}^r d(2 \cdot \sigma / r) \text{ hence } \mu_i^r - \mu_i^0 = 2 \cdot v_i \cdot \sigma / r, \quad (6)$$

where μ^0 is value of μ substance in a flat surface.

From (6) it follows that μ -value in the droplet (bubble) is higher than in a flat surface but in an equilibrium state $\mu_L = \mu_G$. Then, referring to (6) to the left side of equation to vapor and the right part to the fluid, the equation can be written for an ideal system:

$$\mu_i^{rL} = \mu_i^{rV} = \mu_i(T) + R \cdot T \cdot \ln P^r;$$

Substituting μ_i^r and μ_i^0 in the expression (6) gives the desired ratio:

$$R \cdot T \cdot \ln(p^r / p^0) = 2v_i \cdot \sigma / r \quad (7)$$

Over the concave liquid meniscus when $p^r < p^0$ the radius of curvature changes index and for a spherical meniscus the similar expression is obtained:

$$R \cdot T \cdot \ln(p^0 / p^r) = 2v_i \cdot \sigma / r \quad (8)$$

Equation (7), called as Thomson equation shows the vapor pressure of the saturated vapor over a droplet (or steam-gas within the bubbles) will be the greater, the greater σ and the smaller the droplet radius (bubble radius) r .

For example, for water drop with a radius of $r = 10^{-6}$ cm ($\sigma = 73$, $v=18$) the calculation gives a value of $p^0 / p^r = 1,11$. This is a consequence of the equation Thomson-Kelvin that predicts the observed phenomenon of isothermal distillation, which consists of small droplets of evaporation and condensation of the vapor on the larger drops, as well as a flat surface.

Mechanical mixing of water- cement suspensions where the fragmentation of porizing large bubbles is accomplished leads to artificial bubbles polydispersity rise in pressure inside of small bubbles filled with gas or air and saturated steam. It leads to a gas and air diffusion through the film from small to large bubbles till equilibrium sizes as well as diffusion of small bubbles in a matrix substrate and their adsorption on the surface of the solid phase. This process that is similar to the isothermal distillation, as consequence should have an increase in uniformity, reduction of dispersity and the establishment of the equilibrium bubble size classes defined by scale (dimensional) factor [8].

The equation of equilibrium of the system with the external pressure would be following:

$$P + \rho \cdot g \cdot h + 2 \cdot \sigma / r = p_V + p_{LG} + \frac{3}{4} \pi \rho g r^3 / S \quad (9)$$

where S is $1/2$ of the surface area of the gas bubble; P is air pressure at the free surface of the cement suspension, kgf/m²; ρ is density of the suspension, $\rho \geq 1$ kgf·s²/m⁴; g is free fall acceleration of bodies, $g = 9,81$ m/s²; h is depth of the bubble plunge, m; σ is surface tension of the solution, for water $\sigma = 0,073$ N/m at $T = 293$ K, P_V is partial pressure of water vapor inside the bubble, $P_V = 2,34$ kPa, $T = 293$ K; P_L is the partial pressure of air or gas inside the bubble.

Neglecting the air pressure inside the bubble and assuming that $h = 0$ (the force $F = \rho \cdot g \cdot h \cdot s$ is less or equal to the buoyant Archimedes force $F = \frac{3}{4} \pi \rho g r^3$), from (9) we obtain following:

$$P + 2 \cdot \sigma / r = p_V \quad (10)$$

The resulting formula allows determination the radius of the bubble, depending on the external pressure

$$r = 2 \cdot \sigma / (p_V - P) \quad (11)$$

Since $p_V < P$, then (11) gives a negative value of r , which has no physical meaning, but shows the need of presence for the initial nucleation centers, such as gel or contraction pores, from which the growth of bubbles in the liquid phase begins.

With the pressure in the gas or air bubbles from the equation (9) we obtain:

$$r = 2 \cdot \sigma / (p_V + p_{LG} - P) \quad (12)$$

This raises the problem of determining the initial value of p_{L0} (at $P = p_{L0} = 760 \text{ MmHg} = 101.3 \text{ kPa}$ and determination of dependence of $p_{LG}(P)$ or $p_{LG}(r)$.

One of the possible solutions for this problem is the use of Boyle's law: $p_{L0} V_{L0} = p_L V_L$ where $p_{L0} V_{L0} = p_0$ is initial condition, V_{L0}, V_L is volume of the bubble at the initial moment (in normal conditions $P = p_0$) and when change of P , respectively.

Then, as $V_L \approx r^3$, we get the following: $p_L = p_{L0} r_0^3 / r^3$.

Taking into account these assumptions and further solution of the problem on influence of external pressure on the radius of the bubble an expression (12) can be demonstrated as following:

$$r = 2 \cdot \sigma / (p_V \cdot p_{L0} r_0^3 / r^3 - P) \quad (13)$$

After substituting in this expression the $\alpha = r / r_0$ is relative radius of the bubble and its transformations, we obtain following:

$$(P - p_V) \cdot \alpha^3 - p_V / \alpha^2 - p_0 = 0$$

Further transformations of this expression with dividing of introduced the notation $\beta = p_V / p_0$, $\gamma = P / p_0$ by p_0 , analysis of the resulting equation

$$(\beta - \gamma) \cdot \alpha^3 + \gamma \cdot \alpha^2 - 1 = 0$$

may be required in the case of external pressure changes, for example, by evacuating of the cellular mixture. The result of this analysis demonstrates for steam at a temperature of 20°C $r = (6,58 \dots 11) \cdot r_0$ at equality of the external pressure (vacuum) to vapor pressure in the bubble and at the limit (critical) bubble radius.

Considering that relatively large bubbles in the initial period ($T = 20^\circ\text{C}$) of their formation have $p_{VG} \approx P$, from (12) we get following:

$$r_0 \leq 2 \cdot \sigma / r_V \quad (14)$$

From the expression (14) with the assumptions of $\sigma = 0,073 \text{ N/m}$ (for water at 20°C) and $p_V = 2.34 \text{ kPa}$ (water vapor at 20°C), we get following:

$$r_{02} = (2 \cdot 0,073 \text{ N/m}) / 2,34 \cdot 10^3 \text{ N/m}^2 = 0,0624 \cdot 10^{-3} \text{ m} = 6 \cdot 10^{-5} \text{ m}.$$

In order of magnitude ($r_{02} = 0.06 \text{ mm}$) this corresponds to the really observed sizes of small-size pores (bubbles) in the cement paste and cellular concrete.

According to one of the principles of structural topology on size (scale) factor in the structure stability [8], we get the size of a large and middle bubble class the on the base of the law of voids distribution in the random compact packing $\eta_i = 0,65$ and regular one $\eta_i = 0,7405$, respectively:

$$d_V / d_1 = 1 / \left(\frac{2,549}{10\eta_1} \right)^{m(n-1)/3} \quad (15)$$

where $\eta_i = 0,65$ is theoretical packing density of spherical pores.

So for each class of $m = 3; 6; 9; 12$ for distribution systems the full range of largest pore sizes the ($n = 2$) in the packages will be equal:

$$r_1 = 3,92 \cdot \eta_1 \cdot r_{02} = 3,92(0,65 \dots 0,74)0,06 \text{ MM} = 0,15 \dots 0,17 \text{ MM} \quad (16)$$

$$r_1 = 15,39 \cdot \eta_1^2 \cdot r_{02} = 15,39(0,65 \dots 0,74)^2 \cdot 0,06 \text{ MM} = 0,38 \dots 0,50 \text{ MM} \quad (17)$$

$$r_1 = 60,38 \cdot \eta_1^3 \cdot r_{02} = 60,38(0,65 \dots 0,74)^3 \cdot 0,06 \text{ MM} = 0,95 \dots 1,5 \text{ MM} \quad (18)$$

In fact, the observed radius of large pores in the cellular concrete is $0.5 \dots 1.5 \text{ mm}$, which is in good agreement with the calculated data. While the size of small bubbles according to (18), taking $\eta_1 \leq 0,64$ for real disperse (discrete) systems is equal following:

$$\begin{aligned} r_3 = r_{02} / 60,38 (0,64 \dots 0,74)^3 &= 0,06 \text{ mm} / 60,38 (0,64 \dots 0,74)^3 \\ &= 0,004 \dots 0,0025 \text{ mm} (4 \dots 3 \text{ } \mu\text{m}). \\ r_4 = r_3 / 60,38 (0,64 \dots 0,74)^3 &= (0,004 \dots 0,0025) / \\ &60,38 (0,64 \dots 0,74)^3 = \\ &= 0,25 \dots 0,1 \text{ } \mu\text{m}. \end{aligned}$$

These dimensions are also typical for the cellular concrete.

For standard density of loose random packing of spherical bodies ($\eta_i = 0,60$) we have following:

$$\begin{aligned} r_1 &= 60,38 \cdot \eta_1^3 \cdot d_2 = 60,38 \cdot 0,60^3 \cdot 0,06 \text{ MM} = 0,65 \text{ mm}. \\ r_3 = r_{02} / 60,38 \cdot 0,60^3 &= 0,06 \text{ mm} / 60,38 \cdot 0,60^3 = 0,0046 \text{ mm} \\ &(4,6 \text{ } \mu\text{m}). \\ r_4 = r_3 / (60,38 \cdot 0,60^3) &= 0,0046 / 60,38 \cdot 0,60^3 = 0,35 \text{ } \mu\text{m}. \end{aligned}$$

Table 1: The distribution of steam-air bubbles in water-cement aerated concrete and foam concrete mixture

Bubbles diameter in cellular concrete	1,3...1,9 mm	0,06...0,062 mm	8...9,2 μm	0,5...0,7 μm
Volume concentration of bubbles in cellular concrete with D300	0,28549	0,24796	0,20280	0,12089
Volume concentration of bubbles in cellular concrete with D450	0,26202	0,22752	0,18600	0,11018
Volume concentration of bubbles in cellular concrete with D300	0,40170	-	0,28535	0,1700
Volume concentration of bubbles in cellular concrete with D300	0,37400	0,32480	-	0,15836
Volume concentration of bubbles in cellular concrete with D300	-	0,37179	0,30408	0,18127
Volume concentration of bubbles in cellular concrete with D300	0,60216	-	-	0,25498
Volume concentration of bubbles in cellular concrete with D300	-	0,57621	-	0,28093
Volume concentration of bubbles in cellular concrete with D300	-	-	0,53702	0,32012

This size classes form limits for the previous ones only.

The bulk pores content of each size in the cement cellular concrete with average density of 300 and 450 kg/m³ determines its porosity.

$$P(D300) = 1 - \gamma/\rho = 1 - 300/2100 = 0,857143,$$

$$P(D450) = 1 - \gamma/\rho = 1 - 450/2100 = 0,785714,$$

where the 2100 kg/m³ is average of cement stone density, according to T. Powers [9] $\rho = (2000...2200)$ kg/m³.

Distribution of polydisperse porosity in cellular concrete on the base of the packing density of the non-interacting bubbles in the water-cement system for the recurrence equation of topological transformations [8]:

$$\eta = \eta_1 \cdot \left[1 - \frac{1}{3 \cdot \ln(120,754\eta_1^5)} \right] \quad (19)$$

$$0,857143 \rightarrow 0,7861 \rightarrow 0,7131 \rightarrow 0,63654 \rightarrow 0,55285 \rightarrow 0,45217 \rightarrow 0,26954,$$

$$0,785714 \rightarrow 0,7127 \rightarrow 0,63609 \rightarrow 0,55234 \rightarrow 0,45150 \rightarrow 0,267489.$$

The third and subsequent values in these rows are quasi-rigid and quasi-solid → quasi-liquid → critical state of dispersed (discrete) systems.

Taking the sum of the ranks of the last four values of random packing bubbles density for four sizes, we get the coefficient of the introduction of the subsequent fraction of bubbles in the cement paste contained between the bubbles of the previous fraction:

$$K_B = \sum \eta_i = 0,63654 + 0,55285 + 0,45217 + 0,26954 = 1,9111,$$

$$K_B = \sum \eta_i = 0,63609 + 0,55234 + 0,45150 + 0,267489 = 1,9074.$$

Thus the volume concentration of each bubble fraction in the cement paste is calculated by following formula:

$$\varphi_i = \left(\gamma / \sum \eta_i \right) \eta_i, \quad (20)$$

where η_i is random packing density of monodispersed pore corresponding to this pseudophase; for highly polydispersed systems with following porosity:

$$\sum \eta_i = \Pi = 0,8571 (0,7131) \text{ and } \sum \eta_i = \Pi = 0,7857 (0,7127).$$

The results of calculations by formula (20) for random and the most probable distribution of pores in the porous polydispersed concrete suspensions with a porosity of P(D300) = 0,85714 and P(D450) = 0,7857 are shown below in Table 1.

According to data of the Table it follows that porosity distribution curves for cellular concrete demonstrate from one to three maxima. At the critical state of the system the maxima are directed to smoothing up to plateau-like curve depending on the bubbles size tending to zero as a result of reducing the number and size of small bubbles. A similar dependence has also curves of particle size distribution of products of fine grinding of mineral raw.

When the critical state of bubbles in interporous partitions the porosity (P=0.11...0.12) is detected where difficulty removed water during foam concrete drying is approximately 11% by volume [9]. When the size of the large bubbles increases the volume fraction of ones in the cement paste of aerated concrete increase to limited random packing density ($\eta=0.64$), or to a regular closest packing ($\eta=0.74$). The bubbles polydispersity reduces these values as a result of their separation by cement paste, but increases their total content in the system.

In our opinion, the redistribution of large and small bubbles during mixing of the “cement–water” system has influence of a weak electrostatic field of a surface bubbles film, formed by the same cationic charge or polar part of the molecules of gas or foam agent. The charge density of the large bubbles surface is higher than small ones due to its lesser curvature, but the surface tension of the interface is smaller than the small bubbles.

Therefore, the probability of large bubbles merge to reduce surface tension is greater than the small ones, while greater mobility in the cement systems have small bubbles which are in the cellular concrete at a much greater distance from them due to the increased mobility of the cement slurry when stirring as well as the elastic collision. Based on this assumption, we found it possible to equate the resultant force of the surface tension of large and small bubbles force resistance to the bodies' mobility in liquid media to the expression of Stokes' law [9].

The resulting force of mobility of small bubbles is determined. For this the equilibrium equations for small (s) and large (l) of bubbles in the system can be written:

$$P + \rho \cdot g \cdot h + 2 \cdot \sigma_s / r_s = p_{Vs} + p_{Ls} \quad (21)$$

$$P + \rho \cdot g \cdot h + 2 \cdot \sigma_l / r_l = p_{Vl} + p_{Ll} \quad (22)$$

Subtracting the second equation from the first one we get following:

$$2 \cdot \sigma_s / r_s - 2 \cdot \sigma_l / r_l = (p_{Vs} + p_{Ls}) - (p_{Vl} + p_{Ll}) \quad (23)$$

Differential of pressure for gas or air and saturated steam in the small and large bubbles is greater than zero. This allows the left side of (23) to equate the expression for the Stokes' law:

$$2 \cdot \sigma_s / r_s - 2 \cdot \sigma_l / r_l = 6 \cdot \pi \cdot \eta \cdot r_s \cdot v, \quad (24)$$

where η is viscosity of cement slurry (cement system); v is the rate of displacement and diffusion of small bubbles when mechanical stirring of cement slurry (mass), r_s, r_l are radius of small and large bubbles respectively, σ_s, σ_l are surface tension of interfacial film for small and large bubbles, respectively.

Since $\sigma_s / r_s > \sigma_l / r_l$, then (24) holds.

From (24) we find following:

$$v = \frac{2 \cdot \sigma_m \left(1 - \frac{\sigma_m \cdot r_m}{\sigma_m \cdot r_m} \right)}{6 \cdot \pi \cdot \eta \cdot r_m^2} \quad (25)$$

CONCLUSION

Analysis of the results obtained and the expression (25), allows conclude the following:

- Exceeding of Archimedes force $\frac{4}{3}(\rho \cdot g \cdot \pi \cdot r^3)$ above hydrostatic pressure force $\rho \cdot g \cdot h \cdot s$ causes the bubbles rise to the exposed surface of the cement suspension and saving and shape of bubbles (drops) a surface shape tension provides;
- Rate of movement (diffusion) of small bubbles in the cement mixture (slurry) when mechanical stirring is higher than for large ones and it is directly proportional to the surface tension and inversely proportional to the square of its radius;
- Intensity of mixing the cement mixture up to a certain limit leads to a compact filling of the surface area of the interfacial film by landing sites of the cation in gas concrete or polar part of SAC molecules in the foam concrete as a result of overflow of the liquid phase and vapor diffusion of saturated steam molecules inside the bubble to establish thermodynamic equilibrium and topology;
- These conditions lead to a decreasing in surface tension and small bubbles rise up to a certain size, also to increasing of surface tension and reducing of radius of the largest ones to provide with geometrical (topological) correlation of complete displacement in interfacial layer of cation or polar part of SAC molecules, which leads to their size fractionation.

Output: Thus, the topological correlation is performed under certain equilibrium radius of the bubble in the cement system.

Since $\sigma_l / \sigma_s < 1$ and $r_s / r_l < 1$, then the mutual repulsion of large bubbles in the cement system leads to a homogeneous distribution of them and the high mobility of small bubbles e.g. displacement of them to internodal elements Plato, disposed in spaces between the large bubbles and at a high concentration of large bubbles in the high porous systems with of polydisperse porosity – in inter pore partitions of weight “water-cement” system of gas or foam concrete.

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