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Research Article

Elastic Repulsion-Not-Attraction Between Pressurized Holes/Pores

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Key Words

Materials Behavior in Service; Elasticity; Micro-Mechanics; Pore Interactions; Thermodynamics; Modeling

Abstract

Porosity plays a crucial role in materials' behavior as regards swelling, bubble/pore lattice formation, micro-structural changes-e.g. due to bubble/pore migration (especially at high temperatures) plus coalescence or 'collisions' - and, response to complex loading conditions, structural stability (including in geo-mechanics), mechanical-electrical-thermal properties' variations, etc.; through a complex dependence upon the numbers, sizes, positions, internal pressures of the pores, in addition to their strain fields' (elastic) interactions and total volumetric occupation. Hence the pivotal question about the nature, repulsive or attractive, of the elastic interaction among the pores; tackled since the 1950s by pioneers in the micro-mechanics of materials science (e.g. Eshelby). Herein it is proven, in a physically and mathematically rigorous way, that matrix-atoms are diffusing around each pore, practically leaving from one side of the pore and depositing on the opposite (from low-stress regions to high-stress ones), thus forcing the interacting pores to 'repel': In line with experimental evidence which strongly suggest repulsion; e.g. the existence of gas bubble lattices in solids (difficult to justify in a regime of mutual bubble attraction), or, of moving bubbles in solids (due to thermal gradients), which when converging towards 'collision', rotate around each other rather than coalesce. Contrary to opposite results of previous calculations - i.e. 'repulsion-not-attraction' -which, in order to circumvent physico-mathematical complexity, rely upon algebraic manipulations and idealized models (and/or boundary conditions) that are physically unattainable.

Introduction

The existence of pores within materials has been identified, since very long ago, as having a pivotal role w.r.t. porous materials' behavior, in a great variety of phenomena: Swelling, bubble/pore lattice formation, micro-structural changes - e.g. due to bubble/pore migration (especially at high temperatures) plus coalescence or 'collisions' - and, response to complex loading conditions, structural stability (including in geo-mechanics), mechanical-electrical-thermal properties' variations, etc. [1-5]; with all these being greatly influenced by the specific characteristics of the porosity, i.e. the numbers, sizes, positions, internal pressures of the pores, and their strain fields' (=elastic) interactions and total volumetric occupation. Hence, the strong research/industrial interest in the aforementioned interactions, both theoretical and experimental, since more than half a century ago, e.g. [6]. Mechanical forces on bodies lead to displacements (deformation, motion/movement). For instance, a bubble in a liquid is 'pushed upwards' due to buoyancy the same way as a balloon in the sky. Pores (also called cavities or bubbles) inside solids - e.g. elastic and isotropic - do move [6,7] but via a different mechanism: Matrix atoms from one side of the pore surface 'travel' to the opposite side, by diffusion, and thus the pore 'moves'. So, for a pair of pressurized holes/pores within infinite matrix, their motion closer or farther from each other is dictated by a 'generalized force' causing a 'generalized displacement' of the holes/pores [6-9]. For mathematical simplicity, a pair of 3-D equipressurized/equisized pores (of radius R, and internal pressure P, e.g. due to entrapped gas), in an 'infinite' solid, is routinely modeled as a pair of infinitely long (parallel) cylinders, i.e. 2-D equipressurized/equisized holes [2-5,10-12]; as per Figure 1. The matrix is taken, initially, as elastic and isotropic. The scope is to use plane strain elasticity in order to determine the interaction (repulsion or attraction) between this pair of holes. However, even with the simplification of 2-D geometry, the problem remains very complicated mathematically, thus requiring some further degree of approximation. Consequently, the methodology of approximation, and the used assumptions, are crucial elements for obtaining the correct result. Herein, a methodology is established, based on first principles, in order to go through the various proposed solutions appearing in the literature; for the purpose of clarifying/identifying the route to obtaining sound results, backed by experimental evidence.

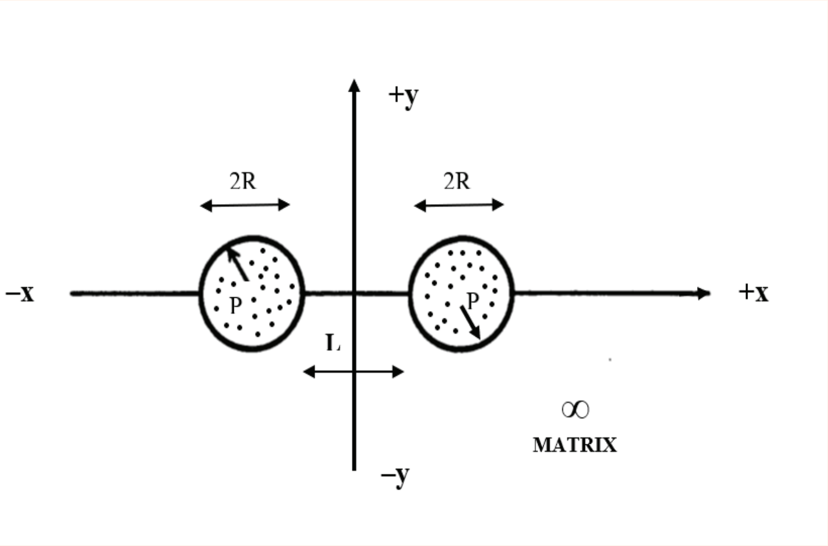

Figure 1: Two equisized pores of radius R under internal pressure P at tip-to-tip distance/separation L – within an 'infinite' matrix where the (2-D) pores are depicted as 'infinitely' long cylinders along the 'paper depth dimension

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Methods

A twofold approach is taken herein in order to address the problem (of inter-hole/pore elastic interaction): Energetic and Thermodynamic Methodologies. From the Energetic Methodology's point of view, one focuses on the energy needed for the neighboring and interacting (elastically) pores to move, either towards to, or away from, each other. The primary target is not so much to find the exact amount of energy necessary for such motion(s), but rather to identify the possible source(s) that can supply the aforementioned energy. In addition to identifying which way/direction this energy will 'incline' particular chunks of matrix-atoms to move, so that the relevant pore motion can be revealed. From the Thermodynamic Methodology's point of view, one focuses on the variation of the total Gibbs energy as the holes approach to, or move away from, each other; i.e. hole/pore motion will be dictated by the minimization of the total Gibbs energy [6]. For the total Gibbs energy, one must account for both the matrix and the gas (which fills the holes/pores producing the pressure). Regarding the hole pressures, some authors assume [2] that they are constant (and equal for all holes) throughout the process of hole/pore motion, even when the holes' volumes are assumed to be changing (during motion, under isothermal conditions); this being a violation of the gas law (not accepted herein).

Results

Original case – two equipressurized & equisized holes

For two equi-sized/equi-pressurized holes in an elastic and isotropic matrix, the question is whether their interaction is attractive or repulsive; i.e. whether diffusion (as described previously) will lead their in-between initial separation $L = L_I$ to become (subsequently) $L_{II}<L_I$, or, $L_{II}>L_I$, respectively (under isothermal conditions).

The matrix surface (gas/matrix interface) area has extra strain energy w.r.t. the bulk of the matrix:

$$\{\text{Surface Energy}\} = \gamma \cdot 2\pi R \cdot 1 \quad (1)$$

where γ is the surface energy per unit area of the matrix (and '1' is the unit length).

During 'I' → 'II', by taking the boundary condition P = constant at radius = R (also within the pore) regardless of pore motion - as done in almost all previous calculations (e.g. see review in [6], as well as [2-5,10,12-14]) - one is obliged to take both the volume and the surface area of the pores as constant as well (due to the gas law, given the isothermal conditions). Thus, the total surface energy of the pores remains also constant. In order for the (gas-filled i.e. pressurized) pores to either approach or distance themselves, a number of matrix atoms need to move (by diffusion from one pore side to the opposite), and this requires energy. Within the (isolated) system 'matrix + gas', there are three repositories that can supply the needed energy to such pores during/for their motion towards to, or away from, each other: {1st} the (matrix/gas interface) surface energy; {2nd} the strain energy of the matrix; {3rd} the kinetic energy of the gas atoms (i.e. the internal energy of the pore gas, which depends only on temperature). As explained above, the 1st is inaccessible, since it stays constant; the 3rd is unavailable in an isothermal process as herein; inevitably, the 2nd has to diminish.

Hence, the pores will take the 'direction' (move closer or farther) of diminishing strain energy.

In [3,4] exact analytical expressions have been derived regarding the said strain energy:

$$E_{matrix}(L) = 2 \cdot \left[\frac{\pi(R^2 \cdot 1)P^2}{(2\hat{G})}\right] \cdot \left[1 + \frac{R}{L}\right] = 2 \cdot E_0 \cdot \left[1 + \frac{R}{L}\right] \quad (2)$$

where Gˆis the shear modulus, and E_0 is the strain energy of an isolated pressurized hole within an 'infinite' isotropic matrix [15]. Therefore, matrix strain energy is inversely proportional to pore separation (⇒ repulsion); QED. The above result, is fully consistent with matrix atom diffusion directivity (from one pore side to the opposite): Matrix atoms diffuse towards relaxing stress concentrations (from low stress regions to high stress ones) -e.g. [16] (Eq. (4) pg. 369 etc.), and the phenomenon of diffusional creep [17] - i.e. their motion is compatible with repulsion, since high stresses are more pronounced between the holes/pores rather than on the opposite side(s) [10]. This result has significant repercussions in a variety of phenomena; e.g. regarding material swelling: If (or when) there are no pore-to-pore repulsions (more so if/when there are pore-to-pore attractions), pore coalescence is enhanced and individual pores tend to get larger, although total volumetric porosity stays the same; but larger pores grow faster either by diffusion (and/)or creep of the matrix (which surrounds them) [17,18]; thus, swelling is enhanced as well (while fracture life is likely shortened).

Generalization – unequally pressurized pores of unequal sizes

The results of the previous Section-3.1 are by no means limited to the symmetries of equal sizes and internal pressures of the holes/pores. The rationale elucidated

above is valid for all possible pressure-size inequalities; thus, repulsion or attraction depends on the functional form of the strain energy for every case. Hence, following [3], three more cases - other than Eq. (2) taken as 'Case (i)'- can be shown for comparison: 'Case (ii)' for an equilibrium hole (internal pressure = surface tension) vs. a pressurized one, 'Case (iii)' for a pressurized hole vs. an equilibrium one of a much larger size, 'Case (iv)' for an under-pressurized hole (internal pressure < surface tension) vs. an equally pressurized one; E_{tot} being the total (matrix) strain energy

($E_{tot} \equiv E_{matrix}$) in Figure 2 below [3]:

Comparative graph for curve/case

{'i'} of Eq. (2) $\left(E_{matrix} \equiv E_{tot} = 2E_0 + 2E_0 \cdot \frac{R}{L}\right)$,

and the analogous curves,

{'ii'} $\left(E_{tot} = E_0 + \frac{1}{2} \cdot E_0 \cdot \frac{R}{L}\right)$,

{'iii'} $\left(E_{tot} = E_0 + \frac{1}{4} \cdot E_0 \cdot \sqrt{\frac{R}{L}}\right)$

{'iv'} $\left(E_{tot} = 2E_0 + \frac{1}{6} \cdot 2E_0 \cdot \sqrt{\frac{R}{L}}\right)$

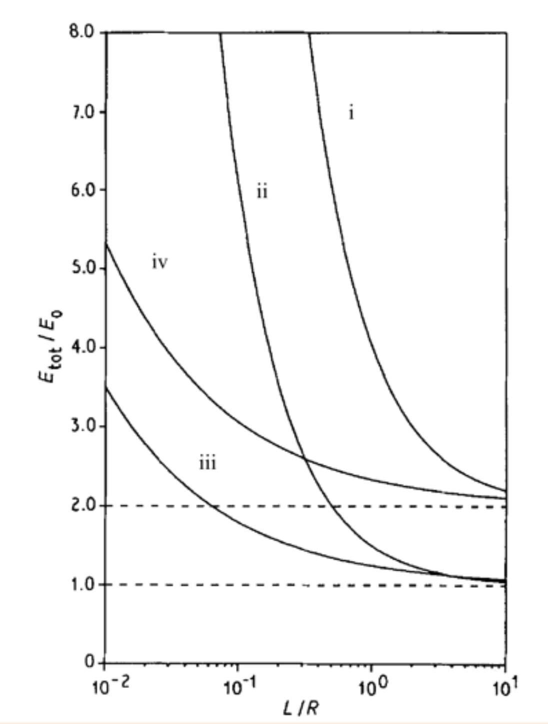

Figure 2: Total strain energy over single-pore strain energy for two pores … (i) one pressurized the other equally pressurized both of same size & (ii) one equilibrium the other pressurized both of same size & (iii) one pressurized the other equilibrium of much larger size & (iv) one pressurized the other equally under-pressurized both of same size [3] – L and R as per Figure 1.

Since the strain energy is always inversely proportional to inter-pore separation, repulsion happens in all cases. The same is true, regardless of the magnitude of the pressure of each individual hole in the interacting pair [4]. Interestingly enough, repulsion is also obtained when a pair of equilibrium pores (holes), within 'infinite' matrix, is subjected to a remote (uniform) tensile stress perpendicular to the plane of the holes' (infinite cylinders') axes; again, this is because the strain energy is inversely proportional to inter-pore separation [5]; see Figure 3. The common characteristic in all cases (Figures 2-3) is the stress concentrations within the region in-between the holes [10,11]: As elucidated previously, atomic fluxes drive matrix atoms to this region, i.e. fluxes initiating from the region of the opposite side of the pores (opposite to their in-between region), thus 'making' the pores move away from each other (repulsion); in absolute consistency with the mechanism already described (about the stored strain energy 'fueling' the pore motion).

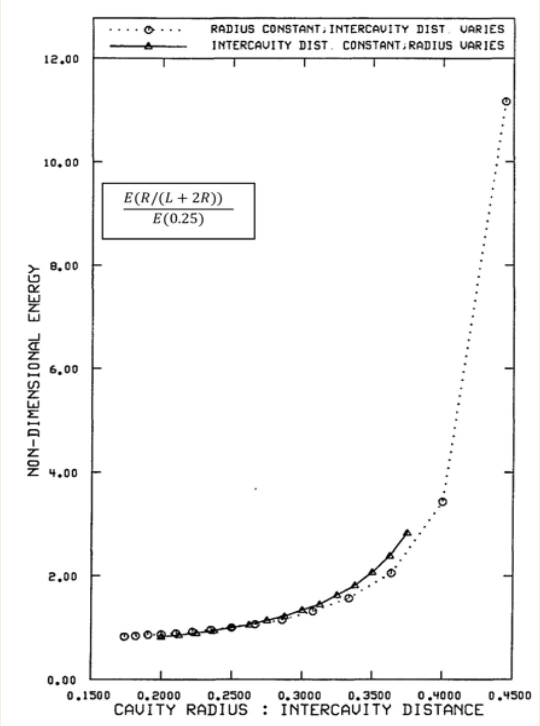

Figure 3: Repulsion between two equi-sized equilibrium pores within 'infinite' matrix under remote & uniform tensile stress perpendicular to the plane of the cylindrical holes' axes as per E(R/(2R+L)):E(0.25)∝L^{-3} [5] – 'interpore separation' ≡ L and 'intercavity distance' ≡ (L+2R) where L & R as per Figure 1 (i.e. for L→0 ⇒ R/(L+2R)→0.5).

Thermodynamics of elastic repulsion

Coming back to the 'original case' (two elastically interacting equi-pressurized/equi-sized holes embedded in 'infinite' matrix), it is shown in the following that one can obtain the same result (repulsion) as per Eq. (2), via a thermodynamic calculation by incorporating the P = constant assumption of

$$P_{II,gas} = P_{I,gas} = P \ . \ (3)$$

Then, under isothermal conditions, and due to the ideal gas law,

$$V_{II,pore} = V_{I,pore} \ (4)$$

where $V_{II,pore}$, $V_{I,pore}$ are the deformed volumes of the holes (i.e. after the application of pressure).

Pore movement in a solid can be treated via the Gibbs energy,

$$G = U + PV - TS \ (5)$$

where G, U, P, V, T, S have their standard/usual meaning, i.e. Gibbs energy, internal energy, pressure, volume, temperature, entropy, respectively; therefore, taking a priori that $L_{II}>L_I$, nature will 'choose' repulsion if

$G_{II}<G_I$, and attraction if $G_{II}>G_I$ [6].

Following [6] (page 151),

$$\Delta G = \Delta G_{system} = G_{II} - G_I = \Delta G_{matrix} + \Delta G_{gas} + \Delta G_{surface} \ (6)$$

where the subscript 'surface' refers to the change in the gas/matrix surface energy.

For the gas (Q=heat, W=work, c_V = specific heat capacity under constant volume, n= # of the gas particles in a pore, k=Boltzmann constant),

$$\Delta G_{gas} = \Delta U_{gas} + \Delta(PV)_{gas} - \Delta(TS)_{gas} =$$
$$= 2nc_v\Delta T_{gas} + 2\left(P_{II,gas}V_{II,pore} - P_{I,gas}V_{I,pore}\right) - T\cdot\Delta S_{gas} \Rightarrow$$
$$\Delta G_{gas} = 0 + 0 - Q_{gas} = -Q_{gas} \ (7)$$

No work (W_{gas}) is done, because pressures and volumes of all (gas-filled) pores remain constant, thus, due to the first law of thermodynamics:

$$-Q_{gas} = W_{gas} - \Delta U_{gas} = 0 = \Delta G_{gas} \ (8)$$

For the 'infinite' matrix [6] (page 151),

$$\Delta G_{matrix} = \Delta E_{matrix} + \Delta\{ExternalForces'PotentialEnergy\} - Q_{matrix} =$$
$$= \Delta E_{matrix} + 0 + Q_{gas} = \Delta E_{matrix} \ (9)$$

since the {External Forces' Potential Energy} does not change, because gas pressures and volumes are identical in 'I' and 'II'.

As previously explained,

$$\Delta G_{surface} = 0 \ (10)$$

which is a tautology anyway, since each hole's (pore's) volume/dimensions is/are constant, so, its surface area remains constant as well (⇒ its surface energy too).

From Eqs. (3-10), under isothermal conditions,

$$\Delta G \equiv \Delta G_{system} = \Delta E_{matrix} \ (11)$$

From Eq. (2) it is seen that the difference in the internal (=strain) energy of the matrix, for pores 'moving' apart ($L_{II}>L_I$), is negative (⇒ repulsion); QED.

Discussion and Conclusion

By employing first principles, it is confirmed that equi-pressurized and equi-sized holes/pores in an isotropic and linear elastic matrix, are 'inclined' to 'move' apart (=repulsion), as per the Davanas [3-5] analytic solutions, and the Teltow [14] approximation; due to the actions of 'generalized forces' resulting in 'generalized displacements', e.g. as per Ref. [6] (pages 151 & 152 about 'effective forces'). The natural mechanism appears to be as follows: In order for the pores to move farther from each other, by the process described earlier (diffusion of atoms from one pore side to the opposite, towards relaxing stress concentrations and high stress regions in-between the pores), the only energy source capable of fueling such a movement is the stored strain energy of the matrix; consequently, the latter (E_{matrix}) diminishes as per Section-3.2; consistently with the thermodynamic analysis (minimization of the Gibbs energy) as per Section-3.3. These two proofs (energetic & thermodynamic) are the verifying clarifications, as regards the Davanas' works [3-5], which re-affirm that the gradient (w.r.t. separation) of the strain energy alone is what decides the nature (repulsive-not-

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attractive) of pore-to-pore interactions (negative gradient w.r.t. separation ⇒ repulsion and vice versa).

Thus, among other phenomena, taking as an example material swelling, one sees that pore-to-pore repulsion can play a major role: By/when hindering pore coalescence, it does not allow pores to get fewer but larger at the same volumetric porosity; hence, since larger pores grow faster than smaller ones (both by diffusion and matrix-creep[17,18]), swelling can be hindered as well (moreover, fracture life can be extended). The opposing views [2,13,19,20] - i.e. attraction instead of (the correct) repulsion - may be due to a variety of reasons: Utilization of multiple assumptions which can lead to results hesitantly applicable and only at extreme cases; e.g. approximations by Eshelby [19], commensurate only for holes very far away from each other, as indicated by Nichols [6] & Willis and Bullough [20]]; i.e. when the interaction is expected to be minimal anyway, if not non-existent altogether. Or, employment of assumptions subsequently proven unsound; e.g. assuming uniform displacements at the hole surfaces [13,19-21]; analytically refuted by a later calculation [3]. Or, usage of the P = constant assumption - i.e. the internal pressures of the pores being equal and constant at all times [2] - even when the pores' volumes are assumed changing during pore motion (under isothermal conditions, in violation of the gas law); unknown when applicable, if ever (as quoted from Ref. [2] … "A constant pressure setting, however, would not be applicable to problems where the pressure changes with expansion, such as expansion of a cavity containing gas, where the pressure and volume are coupled together by gas law").

The latter paper, i.e. Saeedi & Kothari [2], as most recent/modern, deserves a few more words: It appears dubious, also, w.r.t. the so-called 'potential energy method' (to calculate forces at various places in the 'matrix + gas' system), which it uses like if there exists a 'pushing-closer' mechanical force, analogous to the case of a buoyancy-driven 'upward-push' of a bubble in a liquid or a balloon in the sky. But such a mechanical force would have a point (or surface or volume) of application, not obvious at all where, and not mentioned at all in the said paper. In addition, the 'potential energy' stemming from the pore-gas pressure (force), is treated in a questionable manner: E.g. for the case of an isolated hole embedded in 'infinite' matrix, the potential energy (per unit length) due to the pore pressure, is taken as equal to $\{-P\cdot\Delta A^*\}$; where ΔA^* is supposed to be the change in area of the hole due to deformation, computed as $\Delta A^* = \pi\cdot(\alpha^2 - A^2)$, where α is the deformed radius of the hole (i.e. after the application of pressure), and, A is the un-deformed radius of the hole; although, as seen from Figure 1, it is rather $\Delta A^* = 2\cdot\pi\cdot(\alpha - A)\cdot 1$, with '1' being the unit length. Hence, the misconception about the nature (mechanical versus 'generalized') of the pore-to-pore force (even more so if coupled with numerical/algebraic errors of unknown/fundamental gravity), can certainly be a source of unsound results; especially in the light of strong experimental evidence: The data from an old but classic/focal experimental paper definitely attest to the above conclusions (repulsion); see Barnes & Mazey's [7] page-52 quotation, regarding diffusional motion of pores/bubbles, due to thermal gradients, in a matrix: "…that a short range repulsion does exist between these bubbles … there is repulsion due to a strain field … the small bubble is seen to rotate about the larger bubble like a satellite during the heating pulses and yet it has not been assimilated by the larger bubble…". The same holds, when elasticity applies, for the phenomenon of gas bubble lattices in solids [22-27]; where it is difficult to reconcile the fact that these bubbles do not collapse/coalesce into one another with the claim of mutual attraction among them.

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