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Modelling of swelling and deformations of homogeneous hydrogel

J. Štorkán[a], T. Vampola[a], M. Dušková-Smrčková[b], K. Dušek[b]

[a] Faculty of Mechanical Engineering, Czech Technical University in Prague, Technická 4, 160 00 Praha, Czech Republic
[b] Institute of Macromolecular Chemistry, Czech Academy of Sciences, Heyrovského nám. 2, 162 06 Praha, Czech Republic

1. Introduction

The work deals with the analysis of mechanical behavior of swelling materials and numerical problems that arise in solving them. Any material capable of absorbing or dissolving a fluid can swell. Most often, osmosis enters liquid into the mass, increasing the body volume. Elastic forces try to maintain volume, and create pressure in the liquid. The pressure from the elastic forces must be in equilibrium with the osmotic pressure. On larger scales, the osmotic pressure can be replaced by the surface tension of the liquid that holds the liquid in the pores of the material. The liquid pressure causes a volume increase. If the pore structure is sufficiently fine (compared to the overall model size), the swelling material can be modeled as a continuum. In the case of osmotic pressure swelling, the conditions for the continuum are always met.

Our team develops materials for biological use. A suitable medium is required for the cultivation of cells, fungi or other materials. The environment must be humid. In the environment there must be solid matter on which the cells grow and multiply. Polymer hydrogels are used as this medium. They are materials made up of long polymer chains. Swells on contact with solvent [2]. Both homogeneous and porous can be produced. The size, amount and topology of the pores can be well influenced during manufacture. One method of pore formation is to mix crystals into the gel, which are then allowed to dissolve. Representative of such gels is silica gel, which is used as a moisture absorber.

2. Mechanical properties

Modeling of material swelling using the finite element method goes in several ways. Volume change can be forced by boundary conditions, by analogy to thermal expansion, or by inclusion in the material description. The last option is closest to reality. Material experts describe swelling by changing Gibbs' free energy. Neglecting the difference between Gibbs and Helmholtz free energy (hereinafter referred to as potential), this energy can be used directly as a material description. Most large FEM packages allow you to use any hyperelastic description of material behavior.

The potential for swelling materials has 2 parts. One part describes the specific work performed by osmotic pressure Δg_{mix}, see [4]. The second part presents the normal hyperelastic material description $\Delta g_{el,n}$, see [3]. These energies are additive.

$$\Delta g = \Delta g_{mix} + \Delta g_{el,n}\,, \tag{1}$$

$$\Delta g_{mix} = RT\frac{\phi_2^0}{V_{1mol}}\frac{\phi_1}{\phi_2}(\ln\phi_1 + g(\phi_2)\phi_2) - RT\frac{f_e-2}{f_e}v_e\phi_2^0\ln\phi_2\,, \tag{2}$$

$$g(\phi_2) = g_0 + g_1\phi_2 + g_2{\phi_2}^2, \tag{3}$$

$$\Delta g_{el,n} = \Delta\psi_{el}(\boldsymbol{\lambda}) - \upsilon_e RT\phi_2^0 \ln\frac{V}{V_0}, \tag{4}$$

$$\Delta\psi_{el}(\boldsymbol{\lambda}) = -RT\upsilon_e\phi_2^0\frac{n-1}{2}\left(\ln\left(1-\frac{\lambda_1^2}{n-1}\right)+\ln\left(1-\frac{\lambda_2^2}{n-1}\right)+\ln\left(1-\frac{\lambda_3^2}{n-1}\right)-3\ln\left(1-\frac{1}{n-1}\right)\right), \tag{5}$$

$$\phi_1 + \phi_2 = 1\,, \tag{6}$$

$$\frac{\phi_2^0}{\phi_2} = \frac{V}{V_0} = \lambda_1\lambda_2\lambda_3\,. \tag{7}$$

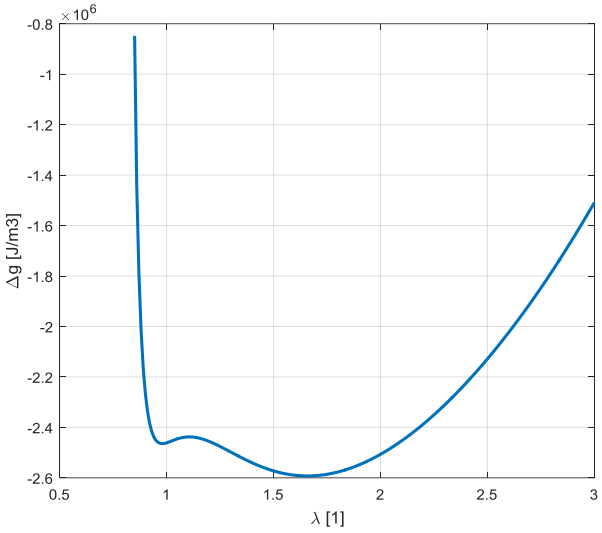

Fig. 1. Potential for isotropic stretch

Parameters λ_i are the main encounters, parameters ϕ_1, ϕ_2 are volume fractions of gel and solvent. The other parameters are constants of different physical meaning. Only the constants g_0-g_2 have no direct physical meaning. In substitution, the potential of functions is only deformation and is therefore well applicable to finite element calculations.

Swelling modeling can be a problem for the stability of the finite element calculation. FEM is a numerical method that requires an initial estimate to find a solution. This estimate is zero shifts. If the actual solution is far from the estimate used, the calculation may not converge. The solution is simple parameters that enforce the magnitude of feeds (typically boundary conditions) do not set straight to the final magnitude, but increase gradually. The calculation is incremental. After each increment, the calculation iterates, finds the solution, and uses it as a new estimate. The incremental scheme ensures small changes to the solution from the estimate and thus ensures convergence. If the material description itself induces the shifts, FEM does not have parameters that it can incrementally control. Therefore, the entire swelling process takes place in a single increment. If the displacements since swelling are large, the calculation may not converge.

The second problem of the material description used is the uniqueness of the solution. Depending on the constants used, the potential may not be convex. The potential may show more extremes. The only way to choose one of many solutions is to initially estimate the displacements. This choice is very problematic. It can be done on simple tasks, but on complicated tasks it may not be realistic.

Our solution procedure allows to solve both problems simultaneously. The whole principle is to apply the incremental scheme not only to boundary conditions and other external influences, but also to the material model itself. It is necessary to find a parameter in the material description that controls the amount of swelling. From non-swelling material to fully swelling. There is no such parameter. But it is possible to create it artificially. We can use non-swelling material description in addition to swelling. This sum may be supplemented by a weighting

coefficient. This coefficient folds as a switch between material descriptions. Switching the description can be both step and continuous. If the weight coefficient is incrementally controlled, the swelling will be controlled to ensure convergence.

It still does not solve the case of non-convex potential. The choice of solution is not determined. We use three potentials instead of two. The first description is still non-swelling, the second can be swelling convex, where the minimum is equal to the minimum of the non-convex potential to which the calculation will converge. The last third description is the material description of the equation (1).

$$\Delta g = (1-\alpha)\Delta g_1 + \alpha\big((1-\beta)\Delta g_2 + \beta\Delta g_3\big)\,. \tag{8}$$

Equation (8) shows the composition of the total potential from three parts coefficient α serves as a switch between potential 1 and 2, coefficient β switches between potential 2 and 3. The calculation is divided into two steps in the first is β = 0 and α is incrementally controlled from 0 to 1. In the second step, α = 1 and β is controlled from 0 to 1. In such a case, the calculation should reliably converge and it is possible to choose the solution to which the calculation converges.

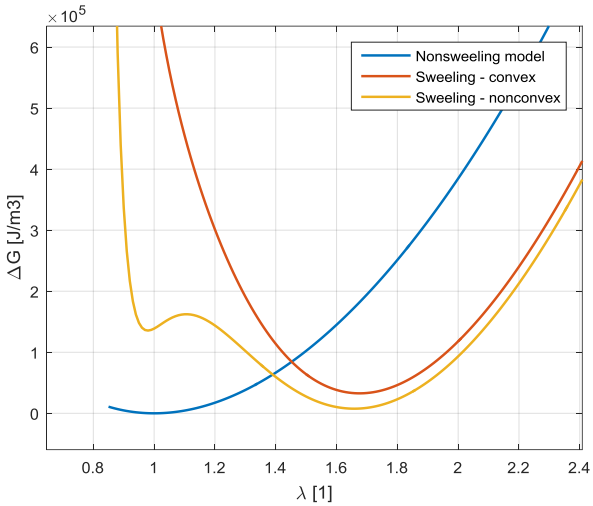

Fig. 2. Potential for models Δg_1 – blue, Δg_2 – red, Δg_3 – yellow

There are 3 possible solutions for the potential from the Fig. 1 is the local maximum potential. This is an unstable solution. The remaining 2 are stable. The choice of solution for use is arbitrary. One way is based on Maxwell's construction. It is based on the calculation of the chemical potential. The sign of the chemical potential integral between stable roots determines a more stable root [1]. A simpler procedure is to select the root with the lowest potential. This is the most stable solution.

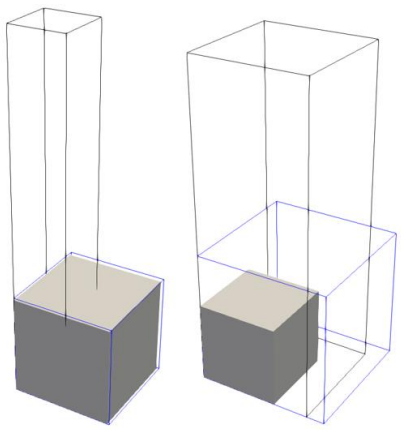

Fig. 3. Initial, swollen and tense shape of cubes

The calculation can be demonstrated on a cube model. If we select the constants in the potential, we get the waveform according to Fig. 1. There are 2 sizes of swelling. For the first root, the material swells only a little less than the original. The second root swells to about 50% larger than the original. Fig. 2 shows the waveforms of potentials when we want to find a solution to the second root. The first step starts with the first potential (blue curve) and gradually switches to the second potential (red curve). In the second step, the second potential is switched to the third potential in the same way. The figure shows that the first potential does not swell and the second potential has a minimum equal to the second minimum of the final potential. If we were to find a solution at the first root, we would use a different second potential. Let the cube swell to both roots and then try to stretch it in one direction (1D stress).

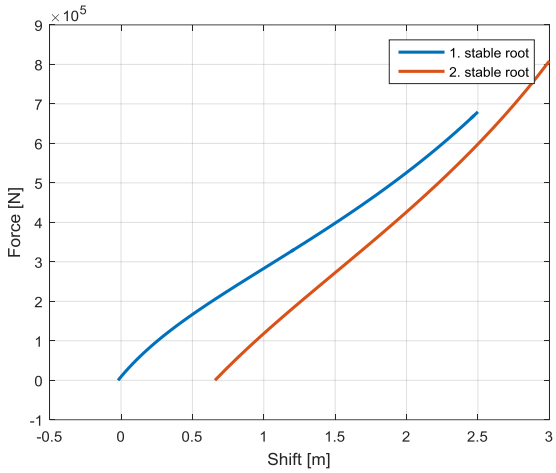

Fig. 4. Tensile curves

3. Conclusion

The test procedure showed how to find multiple solutions for one task, if exists. An example is a cube model with a material description allowing 2 swellings. One will swell to a smaller volume and the other to a larger volume. In both cases the geometry, boundary conditions and material description were identical. The results are shown in Fig. 3. The original configuration, the swollen state, and the stretched state are shown. Fig. 4 shows the tensile curves. In case of greater swelling, the model is stiffer. Without the described procedure, the calculation would probably not find any solution. If he converged, then he would find only one and the other could not be found.

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References

[1] Barra, A., Moro, A., Exact solution of the van der Waals model in the critical region, Annals of Physics 359 (2015) 290-299.
[2] Dušek, K., Dušková-Smrčková, M., Šomvársky, J., Effect of constraints on swelling of polymer networks, Macromol, Symposia 358 (2015) 120-127.
[3] Gent, A.N., A new constitutive relation for rubber, Rubber Chemistry and Technology 69 (1) (1996) 59-61.
[4] Nakamoto, C., Kitada, T., Kato, E., Pressure dependence on the Flory-Huggins interaction parameter of poly (N-isopropylacrylamide) gels, Polymer Gels and Networks 4 (1) (1996) 17-31.