

Summary Report of
Modelling The Formation of Blisters

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Background

The aim of this project was to review the existing literature that couples chemistry and mechanics with the long-term aim of developing a model for the formation of Uranium Hydride blisters. After an initial exploration of the literature and summary of the appropriate theory we chose to concentrate on model problems in two papers:

- *Interaction between diffusion and chemical stress* by Yang (2005).
- *A continuum mechanical model for the description of solvent induced swelling in polymeric glasses* by Wilmers & Bargmann (2015).

Although the latter paper is somewhat removed from the application, it represents a modern “state of the art” approach to chemical transport, reaction and mechanics. Our aim was to investigate whether the theoretical framework would be of any use. Note that explicit reaction terms are not included in either model, but could be incorporated without too much difficulty.

The fundamental physics can be divided into three parts:

1. Transport (and Reaction) of Chemical(s).
2. Transport and Conversion of Heat.
3. Deformation of the material.

These processes are coupled in the following ways:

Mechanics → Chemistry/Energy:

1. Deformation changes the domain in which the chemicals and heat are being transported.
2. Deformation will affect the internal energy and hence chemical potential and diffusion coefficients.
3. Mechanical work can induce heat.

Chemistry/Energy → Mechanics:

1. Chemical concentrations can lead to local expansion that induces stresses.
2. Thermal expansion can also induce stresses

Standard thermodynamics should also be taken into account.

Typically, the timescales of each process are well separated so that dynamic deformation effects (oscillations) occur fastest, on the order of seconds or faster. The transport of heat is the next fastest and finally the transport and reaction of chemicals is typically the slowest process. The large separation of timescales leads to sets of equations that are “stiff”. Stiff systems are not well-suited to explicit timestepping schemes because the timestep is limited that of the fastest process. This lead to the numerical problems that Gareth encountered at the end of the project. Designing an implicit timestepping scheme would obviate these problems and is therefore, in our opinion, absolutely essential in the study of these systems.

Using the separation of timescales: Yang’s approach

In Yang’s paper (section 3), solving a system with disparate timescales is avoided by assuming that the two fastest timescales are instantaneous. In other words, by assuming isothermal conditions and that the solid is always in mechanical equilibrium. Following the classic work of Prussin (1961), Yang couples the chemistry to mechanics by using an analogue to classic linear thermoelasticity:

$$\text{strain} = \frac{\text{stress}}{\text{Young's Modulus}} + \textit{Isotropic chemical expansion},$$

where the chemical expansion term is linearly proportional to the concentration. In alternative theories, more complex non-local behaviour has been introduced to account for the fact that the stresses are transmitted at faster speeds than the chemical. In other words the solid “feels” the approach of the chemical via the stress. Yang solves the mechanical equilibrium equation analytically and substitutes the resulting stress into the remaining chemical transport equation which is then solved numerically. Yang uses a chemical potential that is linearly proportional to the hydrostatic stress so that gradients in stress also drive chemical transport.

Thus, a generalised version of Yang’s approach can be summarised as follows:

1. Assume system in thermal and mechanical equilibrium.
2. Assume constitutive relationships. In the most general case, this will be a relationship for the internal energy.
3. Solve equilibrium equations directly to find deformation as a function of chemical concentration.
4. Substitute results from 3 into the chemical transport equation and solve.

Note that the numerical solution of the equilibrium equations may be incorporated into an implicit timestepping solution.

Gareth was able to implement an explicit timestepping scheme in MATLAB that could reproduce all results in Yang’s paper. The initial hope was that this scheme could be extended to the more general set of equations considered in the next section, but although this is possible (with some difficulty), it is not the best way to solve that set of equations.

Brute force numerics: Wilmers & Bargmann

Wilmers & Bargmann do not make explicit use of the separation of timescales, but instead use an implicit timestepping technique combined with a finite element method to solve their complex system of equation. This is a difficult paper to follow and has its fair share of typographical errors and obfuscation. We feel that the essential results could be captured by a simple one-dimensional model, although time ran out before the model could be solved numerically.

Wilmers & Bargmann do not assume anything is in equilibrium and also assume a Case II diffusion scenario, which complicates the chemical transport equations, but these can be accommodated. The chemical diffusion coefficient depends on chemical concentration, temperature and deformation, but not explicitly on the stress.

Unlike Yang, Wilmers & Bargmann do not assume that the concentration affects the thermomechanical constitutive law directly. Instead they modify the model’s Lamé parameters depending on the value of the concentration relative to a critical value. The separation of timescales is such that the system is in approximate mechanical equilibrium and the observed expansion is actually driven entirely by the thermal expansion. The effective constitutive law is of the form

$$\text{strain} = F(\text{stress}) + \text{Isotropic thermal expansion (coefficients depend on chemical)}.$$

Effectively, the difference between chemical to mechanical coupling is that Yang uses an expansion term of the form αC , whereas Wilmers & Bargmann use $\alpha H(C)$, where α is a constant, C is the chemical concentration and, $H(C)$ is the Heaviside (step) function. Necessarily, the latter form leads to more abrupt changes in behaviour.

Examining their results it appears that the separation of timescales approach used by Yang could have been used even with this more complex system of equations. The system is driven entirely by the chemical transport equation coupled to the abrupt transition in mechanical parameters.

Conclusions & Future Work

We believe that both papers studied in detail in this project can be incorporated into the same general one-dimensional framework and that the most efficient solution method is to assume thermal and mechanical equilibrium and then solve the single (or multiple) chemical transport equation.

The key areas for future work are:

- Development and implementation of the general one-dimensional model framework using implicit timestepping methods. [Estimated Timescale: 1-2 months full time]
- A more comprehensive investigation of constitutive models and their influence on generic transport properties. [Estimated Timescale: rather open-ended depending on how many different classes can be identified, probably 3-6 months full time]
- Inclusion of reaction terms in the equations. [Estimated Timescale: again somewhat open-ended, but if restricted to specific reactions then approximately 3 months full time]