Modeling of the Behavior of a UF_6 Container in a Fire—DIBONA Code

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INTRODUCTION

Uranium hexafluoride (UF6), the raw material from which the fuel for nuclear power stations is obtained, is stored in the solid state in industrial containers called 48Y. The International Atomic Energy Agency (IAEA) envisages a revision of current regulation and suggests that a container withstands a specific fire test (engulfing fuel fire of 800°C for half an hour, for a steel emissivity of 0.8 and flame emissivity of 0.9). To study the safety of the containers under these conditions, a numerical model was elaborated by the French Atomic Energy Commission (CEA). A 2-D model using the finite element computation code ANSYS was therefore developed. It takes into account thermal and mechanical phenomena as well as mass transfers. Recently some tests have been performed on a Tenerife container (Saroul et al. 1995), and our model has to be validated with this results. As the analysis and the interpretation of physical phenomena have been carried out (Pinton et al. 1995) therefore this document will only show how these phenomena have been introduced in the numerical model. This paper is chiefly concerned with the study of thermohydraulic behavior inside the container, with no reference to the mechanical aspects of the problem. The extrapolation to a IAEA fire test on 48Y will be a future step. Note that this model is the pursuit of the work begun by DURET B. (1992). So, only the new developments will be more detailed.

DEFINITION OF THE PROBLEM

Tenerife Project

Over the past 30 years a number of experiments have been performed, the best instrumented and the most interesting being that of Suzuki et al. (1988). However, they do not in any way reflect real conditions. Therefore, many uncertainties remain about the thermohydraulic behavior of the UF6 under a realistic fire. Also, the numerical model can only be partially validated with these results. Consequently, an experimental project called Tenerife (Casselman et al.1922) was defined and conducted in the scope of a joint research programme between France and Japan, managed by IPSN. The fire is simulated by an Inconel electric furnace. The Tenerife container is identical to a 48Y container except for its length that is reduced by one-third to limit the quantity of UF6 and the overall dimensions of the furnace.

Different Rupture Modes and Description of the UF6

The UF6 is the only material inside the container. It is a colorless solid at ambient temperature that sublimates without melting, as shown on the phase diagram Figure 1, that also shows the following:
- that for a temperature lower than 64°C it can only have a gas or solid phase
- that the UF6 melts at a constant temperature of 64 °C
- that for a pressure above that of the triple point the three phases coexist

- that the vapor pressure rises steeply with the temperature of the liquid to reach the critical point at a value of 46 bars for a temperature of 230°C. Boiling phenomena may also be involved that would lead to a rapid increase in pressure up to the rupture of the metal casing.

The melting of UF6 entails a significant decrease in density (Figure 2), so that the liquid level will increase progressively over time until it occupies all the inner volume. There is then a risk that the container will tear open under the force of hydraulic pressure.

The model will serve to determine whether rupture takes place, and, if so, the type of rupture mode that occurs first.

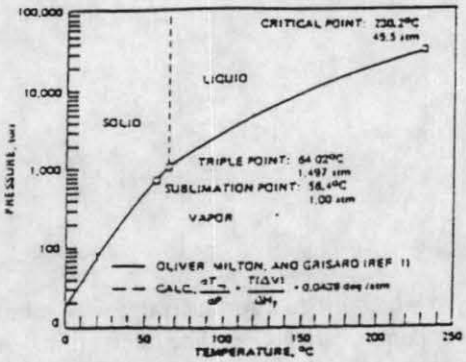

Figure 1. UF_6 phase diagram

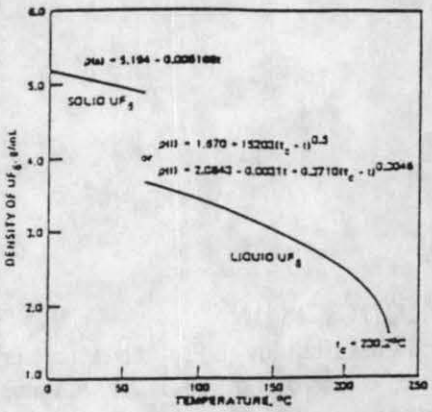

Figure 2. Density of UF_6

Physical Properties of UF6

We referred to the compilation of Anderson J.C. et al. (1994), that assembles and criticizes practically all the literature published on the properties of UF6.

INITIAL STATE AND COOLING OF THE UF6 AFTER FILLING

The initial structure of the solid UF6 within the container was estimated by DURET B. et al. (1992) by analysing the cooling process of the UF6 after filling. For the time being, the modeling of the UF6 in contact with the UF6 gas is approximated by a horizontal surface. The top crust is assumed to be circular in shape and to have a uniform thickness. The initial height of UF6 is determined by the model depending on the thickness of the crust (Figure 3). The steel and the UF6 are modeled by 2-D quadrilateral thermal elements with 4 nodes.

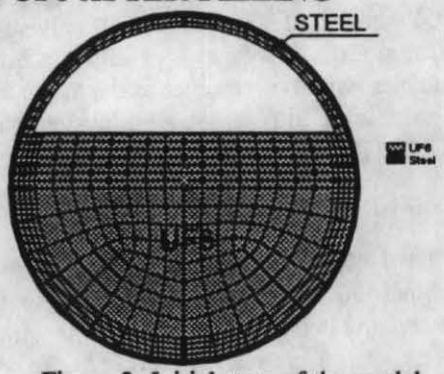

Figure 3. Initial state of the model.

PHYSICAL PHENOMENA INTRODUCED IN THE MODEL

A series of multiphase and transient phenomena takes place within the container. These phenomena are closely dependent on the existence of the top crust, so the analysis will be presented in two parts: the first describes the phenomena when the crust is present, and the second, the phenomena without the crust. Before developing internal heat transfers, external transfers during heating and cooling will be presented.

Heating

This is a purely radiant fire since the furnace and the container are enclosed in a vacuum.

The thermal loading of the container is assimilated to a radiant flux through a transparent medium, emitted by an Inconel casing surrounding the container and heated to a temperature for a specified duration. The temperature profile of the fire during the heating phase is fixed by the user. The

radiant transfers are modeled using the relations for an enclosure made up of N-radiating surfaces (in this case N elements) and assuming these surfaces to be gray and diffuse in emission.

Cooling

In the case of a Tenerife test, cooling will be solely through radiation between the furnace and the outer surface of the metal casing. The furnace is thermally insulated so that heat exchange with the ambient atmosphere will be negligible. Therefore zero exchange is applied to the outer Inconel surface, and the imposed temperatures on Inconel are suppressed. Its temperature falls steadily to equilibrium.

Internal Heat Transfers With the top crust

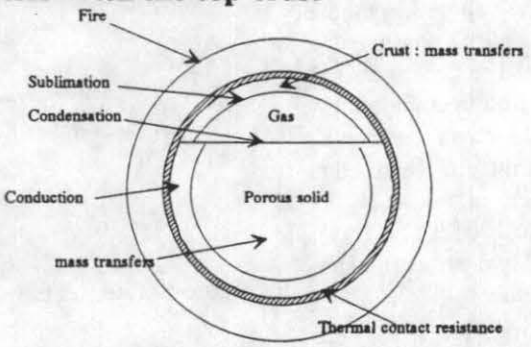

Figure 4. Internal transfers with an existing crust.

Contact Between Steel and Solid UF6

Heat transfers through an interface composed of two bodies in contact generally entail a temperature difference at the interface due to a contact thermal resistance in this zone.There are 3 types of exchange modeled at the interface: conduction when the UF6 adheres to the steel, convection and radiation when the UF6 is separated from the steel by a gas layer, convection alone when the UF6 becomes liquid. A parameter defines the percentage of the surface of UF6 that is assumed to adhere perfectly to the steel, thereby defining the surfaces allocated to each type of exchange.

Contact Between Steel and Liquid UF6

The Tenerife tests (Saroul and al. 1995) have shown that the liquid appears before the crust collapses. These tests indicate that the presence of the liquid has little effect on exchanges at the metal surface. Consequently, exchanges governed by contact resistance are retained in the model.

Transfers in Solid UF6

The solid UF6 is a porous material where inner transfers are generally due to mass transfers through sublimation and condensation of the gas phase which entails an increased transfers in the solid. Therefore an equivalent thermal conductivity much higher than the theoretical value is adopted and applied to the top crust and within the solid.

Mass Transfers at Solid UF6/Gas Blanket Interface

When the temperature of the crust rises, the solid UF6 at the interface sublimates. On the other hand, at the cooler horizontal interface, the UF6 gas condenses. The mass flow and the thermal flux associated with mass transfers through sublimation and condensation are determined and applied as a boundary condition at the interface so that thermodynamic equilibrium is preserved; in other words, so that the steel/gas interface temperature is equal to the internal pressure saturation temperature.

Transfers in the Steel

Heat transfers from the fire to the UF6 are modeled by conduction through the steel.

Internal Heat Transfers without the Top Crust

Break-up of the Top Crust

The physical phenomena responsible for the collapse of the crust have not yet been identified. The model sets the collapse of the crust at the time when the pressure reaches the triple point vapor pressure, i.e., 1.5 bar, as found during the Tenerife tests. Radiant exchanges through the gas pocket between the steel and the horizontal free surface, as well as convective exchanges between the steel and the gas are then activated. All transfers associated with contact resistance are eliminated.

The mass of the crust is transferred onto the horizontal surface of the UF6 and activates the corresponding expansion layers With the collapse of the crust, the liquid trapped between the solid UF6 and the steel is exposed to the gas blanket (pressure 1.5 bar). Boiling phase transfers can now develop and replace the single phase transfers. Transfers through conduction and radiation governed by the contact resistance are cancelled. Convection transfer is retained to simulate boiling phase exchanges, and the exchange surface develops to 100%. The modeling of these particular two-phase transfers is described in the next chapter.

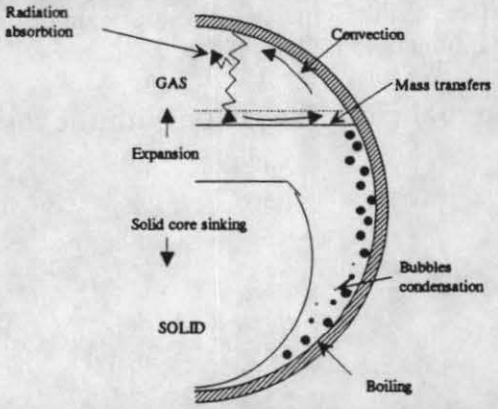

Figure 5. Internal transfers without the crust.

Contact Between Steel and Liquid UF6

Heat transfer during boiling is characterised by the phase change of the fluid from liquid to vapor. The energy exchanged from the wall to the UF6 (q_{Boil}) in a boiling regimen concerns three types of transfer (Figure 6):

- a latent heat flux associated with the Liquid/Gas phase change, $q_{EvapLat}$
- a gas sensible heat flux against the superheated wall, $q_{EvapSens}$
- and a convection flux with the liquid, q_{liq} giving:

$$q_{Boil} = q_{liq} + q_{EvapLat} + q_{EvapSens}$$

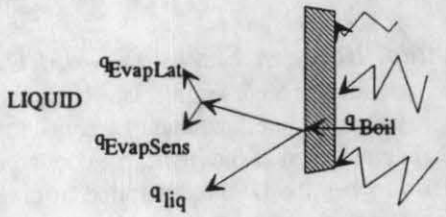

Figure 6. Transfers at the surface during boiling.

A study of the literature on boiling regimen heat transfers was carried out. This enabled us to find and build up relations in order to determine, as a function of the pressure (physical properties), the wall superheat, the surface slope, and subcooling:

- the flux exchanged at the boiling surface (that is imposed as a boundary condition on the inner surface of the steel in contact with the liquid)
- the flux associated with the latent heat of vaporization during boiling (and therefore the quantity of vapor generated)
- the flux of sensible heat of the vapor in contact with the hot wall
- the convection flux with the liquid (applied as a boundary condition on the outer surface of the UF6 in contact with the steel).

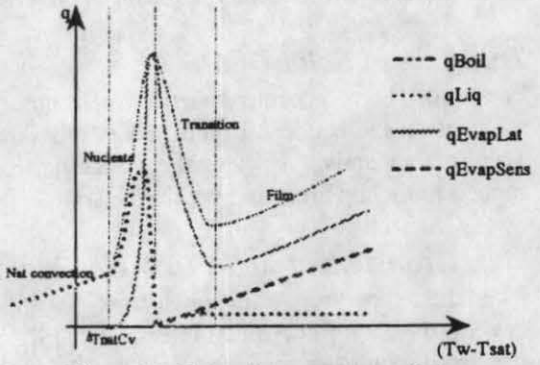

Figure 7. General appearance of the characteristic curves for different transfers boiling regimen.

The general appearance of these four fluxes in relation to the superheat for a set pressure is indicated on Figure 7. It shows that for a same flux at the metal surface the heat transfers may be very different depending on whether the regimen is nucleate boiling or film boiling. In the first case (and for a heat flux equivalent to that of a fire), convective exchanges with the liquid predominate, whereas in the second case, latent heat exchanges predominate.

Modeling of Natural Convection

After the stop of the heating, boiling will be replaced by natural convection. The correlation introduced in the model to simulate natural convection exchange is that of an annular cylindrical space. For a given pressure, i.e., for a known saturation pressure, the superheat in natural convection ΔT_{NatCv}, that characterises the transition natural convection/nucleate boiling is defined by $q_{NatCv} = q_{Nucl}$.

Transfers in liquid UF6

Modeling of Transfers in the Liquid

The computation code used was chosen because it can deal correctly with mechanical phenomena, radiation, and conduction. However, it does not take into account fluid mechanics. Transfers in the liquid are therefore treated by assimilating them to conduction. Finally, to simulate the increasing exchanges due to the turbulence from the bubbles, an equivalent liquid conductivity is adopted. As the bubble generation is different according to the boiling type, the conductivity used depends on the turbulence in the liquid and therefore depends on the type of exchange at the interface.

Modeling Condensation of the Vapor Bubbles

As mentioned in the literature, a two-phase boundary layer begins to develop for a surface with a slope of more than 70° (Figure 8) and that is superheated in the middle. This means that any vapor generated on a surface with an angle of slope of between 0 and 70° will condense. On the contrary, within the bubble boundary layer ($\theta > 70°$) all the vapor generated will not change phase but will reach the gas blanket. So for $\theta < 70°$, all the energy exchanged on the boiling surface will be absorbed by the liquid. So a boundary condition qliq=qboil is imposed at the Steel/Liquid interface. On the contrary, for $\theta > 70°$ the same boundary conditions are kept at the interface. The quantity of vapor from the boiling surface that reaches the gas pocket is that generated on the surface where $\theta > 70$.

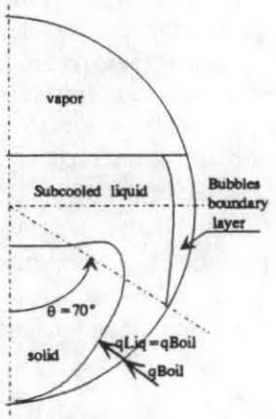

Figure 8.

Melting

For the numerical simulation of melting, the latent heat of melting is incorporated in the enthalpy equation. Melting of a pure material takes place at a constant temperature (64°C) which entails numerical difficulties. So a melting temperature band must be defined: for instance 64 +/- 1° for a temperature band of 2°C.

Expansion

In the initial state the whole of the UF6 is represented in the model. As the temperature rises, the UF6 will expand. Since the UF6 is modeled by a fixed volume *V*, part of this expanded UF6 will no longer be represented by the elements of *V*. When the unrepresented volume (that is determined by the model) exceeds the volume of a predefined expansion layer, this layer is activated as shown on Figure 9.

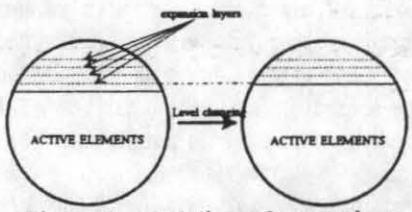

Figure 9. Modeling of expansion

Sinking of the Solid

Since the solid is denser than the liquid, it must find a support at the bottom of the cylinder. The solid begins to sink as soon as the depth of liquid between the bottom of the container and the solid UF6 reaches a value fixed by the user. Assuming this value to be 1 cm; elements are chosen that have at least one solid node ($<64°C$); then the temperature of the UF6 located at 1 cm above the nodes is attributed to the nodes.

Transfers With the Gas Blanket

Convective Exchanges

To model the convective exchange between the metal surface and the gas blanket we use a relation which is a function of the surface slope and defined for a flat plate that is at a higher temperature than the fluid and slopes downwards. The authors claim that this relation is applicable for both laminar and turbulent natural convection.

Exchanges between the gas and the free surface of the UF6 are modeled by the classical relation of for a horizontal surface that is colder than the fluid.

Radiant Exchanges

The gaseous UF6 totally absorbs radiation at certain wave lengths. The ANSYS computation code does not take into consideration the absorbing effect of the gas. To model radiation, the ANSYS method is used with the addition of the absorbing effect of the gas as boundary condition for the surfaces under consideration. To do this, we use the enclosure relations applied through a semi-transparent isothermal gas. It will be noted that the absorbing effect is not in fact very significant. The net flux does not vary by more than 10% compared with the net flux in a transparent gas. However, the energy absorbed by the gas, even if low compared to the surfaces (15%), is high enough to influence gas temperature evolution.

Mass Transfers at the Liquid/Gas Interface

As for the solid/gas interface, the thermal flux and the mass flow associated with mass transfers at the liquid/gas interface are determined and applied as a boundary condition so that thermodynamic equilibrium is preserved.

Pressurization

The pressure obeys the equation of state in which it depends on the volume V, the mean temperature T, the mass of gaseous UF6 m, and the compressibility factor in the gas pocket C. R and M are the ideal gas constant and the molar mass of UF6, respectively:

$$P = C\frac{R}{M}\frac{m}{V}T$$

Volume

The volume of the gas is calculated by taking into account:

- the variation in the volume of UF6 associated with expansion and mass transfers.
- the expansion of the steel and the deformation of the steel induced by internal pressure. These two parameters are determined by ANSYS from the mechanical computation.

Mass of Gaseous UF6

The model determines the quantity of UF6 vapor generated at the boiling surface that arrives in the gas blanket, the quantity of UF6 evaporated or condensed at the horizontal interface, and, therefore, the mass of UF6 in the gas pocket.

Mean Temperature of the Gas

The model calculates the convective exchanges with the steel and the liquid UF6, the gas absorption of infrared radiation, the energy contributed by the vapor produced through boiling at the wall, and the energy associated with mass transfers at the interface. By determining the energy balance for the gas, and referring this to the mass of gas, the mass enthalpy of the gas is deduced. Since the evolution of enthalpy with temperature for a given pressure is known, the temperature of the gas can be found.

Compressibility Factor

The relation used in the model is that defined by Malyshev V.V. (ANDERSON et al. 1994). It depends on the density and the temperature of the gas.

NUMERICAL SUPPORT

Data Processing Features

The present model is in the form of a command file of about 240000 bytes that is run with ANSYS 5.0A.The computations described in the following paragraphs (heating for 18 minutes and cooling for 19 minutes) were performed on a workstation HP 720. Running time required was about 90 hours.

Resolution

Resolution is an iterative process (because of the non linearities) that uses the Newton-Raphson algorithms. For a transient analysis the evolution in time is managed by the Cranck-Nicholson algorithm.

Outputs

During the computation the program generates an archival storage of the model and temperature maps as well as monitoring of the interface elements. In addition there is a specific post-processing that uses a series of menus to treat interactively the file result.

VALIDATION OF THE MODEL WITH TENERIFE RESULTS

This model was validated by the experimental test performed on July 7, 1995 on the TEN2 container enclosed in a furnace at 800°C for 18 minutes (Saroul **et al. 1995).**

Steel Temperatures(Figures 10, 11)

The steel temperatures for the lower part of the container are in quite good agreement with experimental findings. This means that the transfers at the Steel/Liquid UF6 interface and Steel/Solid UF6 interface (contact resistance, boiling, natural convection) as well as the transitions from one or other of the boiling regimen, are correctly modeled.
However, during the cooling phase, the steel temperature in the model for the upper part of the container seems to decrease too quickly.

UF6 Temperatures (Figures 12, 13)

The general trend of the temperatures in the model is similar to that found experimentally:
- all the UF6 reaches the melting temperature at about 560 seconds;
- the time the liquid appears, starting from the top to the bottom, is reasonably respected;
- the stratification of the liquid is present in the upper part;
- the UF6 located at the level of the thermocouples in the lower part remains solid;

Gas Temperatures

The calculated temperature evolution follows that of the model (Figure 14). Note that T2 measures a temperature in a specific point whereas the calculated temperature is a mean.

Pressure

The pressure computation is in quite good agreement with the experimental values (Figure 16, 17). Remark that the pressure determined by the equation of state is equal to the saturation pressure of the mean temperature of the interface. Consequently, thermodynamic equilibrium is conserved throughout the computation. It is found on Figure 15 that the interface temperature is practically uniform over its whole surface. It also shows the stratification of the liquid and that the solid keeps to the bottom of the container.

CONCLUSION

A 2-D, finite element model was developed to simulate the behavior of an UF6 container exposed to a fire. It takes into account practically all the phenomena encountered in this type of situation and can be considered a comprehensive model. This model was validated by the experimental tests performed on July 6, 1995 (Saroul et al.1995; Pinton et al.1995) on the TEN2 container enclosed in a furnace at 800°C for 18 minutes. Overall, the numerical results are quite close to experimental results. Before the model can be completely validated, it will be necessary to check the reproducibility of the phenomena in the next container test. It will then be possible to extrapolate for a 48Y industrial container and the IAEA fire conditions.

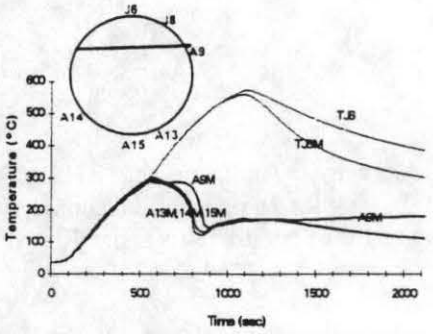

Figure 10. Steel temperature/Modeling.

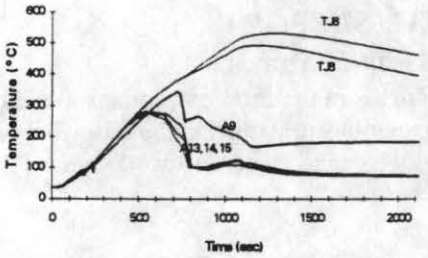

Figure 11. Steel temperature/Experiment.

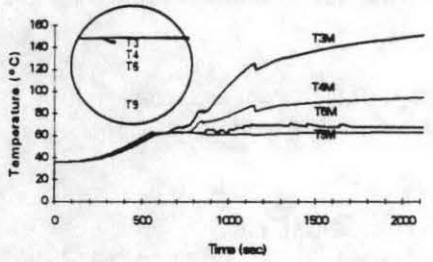

Figure 12. UF6 temperature/Modeling.

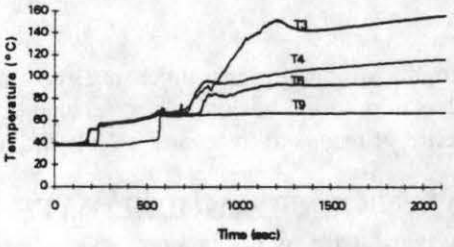

Figure 13. UF6 temperature/Experiment.

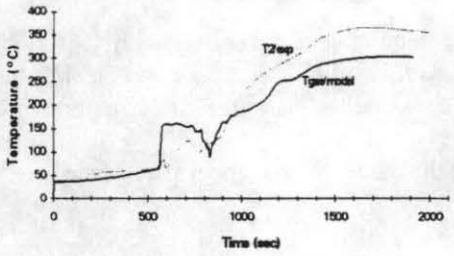

Figure 14. Gas temperature.

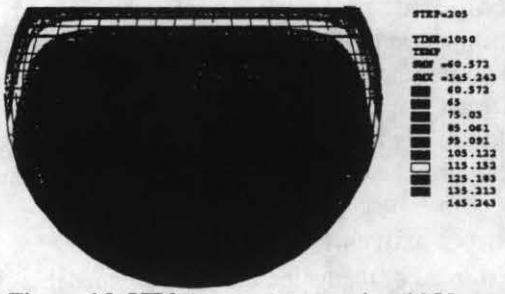

Figure 15. UF6 temperature card at 1050 sec.

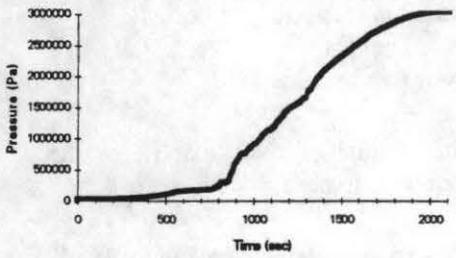

Figure 16. Pressure/Modeling.

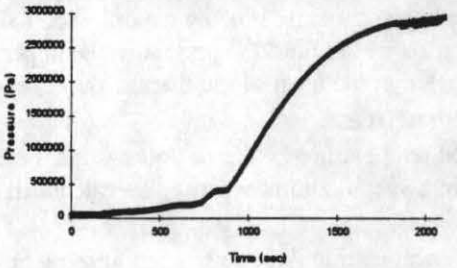

Figure 17. Pressure/Experiment.

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