2D Modelling of a UF6 Container in a Fire

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INTRODUCTION

In France, large quantities of uranium hexafluoride are used in the various uranium enrichment and reprocessing plants. Natural or depleted UF6 is stored and transported in containers called 48Y. The risk entailed is essentially chemical not radiological so the containers used are classified as "industrial-purpose" and do not need to undergo any accident test. No fire test, in particular, is required. However, IAEA recommendations with a view to future regulations specifically mention a fire test on the UF6 containers, so this has become a priority research topic.

The only real fire tests date back to 1966 (e.g., Malett 1966) and were limited to about only a hundred kilograms of UF6. Interpretations of results (e.g., Duret and Bonnard 1983, Williams 1988) do not lead to any definite conclusions on the fire resistance of the 48Y container, but point out the difficulty in achieving a fine model of the processes involved during a fire. Other experimental works at lower temperature and on small quantities of UF6 (e.g., Suzucki et al. 1988, Shin Park 1983) were referred to. These tests were better instrumented and served to refine the theoretical models (e.g., Yamakawa et al 1988, Clayton et al. 1991, Duret 1988, Williams 1991). After consideration of the uncertainties subsisting, the model developed in our laboratory has a twofold objective :

- interpretation of the TENERIFE tests programmed for 1993-94 in which a container having the same diameter as the 48Y container will be heated in a furnace. A description is given in a paper (e.g., Casselman et al. 1992) presented at this conference.
- extrapolation to define the safety margin up to actual bursting of the container. This will permit optimisation of fire-fighting action and inform on the state of the UF6 if and when bursting occurs in order to assess the quantity of UF6 that is likely to be released.

DEFINITION OF THE PROBLEM

Apart from the actual thermohydraulic modelling of UF6, several "outside" causes contribute to the difficulty in understanding the behaviour of a UF6 container exposed to fire.

Properties of UF6

Uranium hexafluoride is a crystal solid at ambient temperature that sublimates readily. At 20°C its vapour pressure is 0.1 bar, which corresponds to the initial internal pressure of the container. This vapour pressure increases with rising temperature : 1 bar at 56°C, 4.1 bar at 100°C, 12 bar at 150°C, and reaches the critical point of 45.5 bar at 230°C.

Solid UF6 tends to crumble and is riddled with cracks and voids due to the solid-liquid phase change. The voids and the high vapour pressure can induce considerable mass transfers and significantly increase internal

heat transfers in UF6.
This explains the variability of thermal conductivity noted by different authors : $\lambda = 0.16$ W/m°C (e.g., DeWitt 1960), $\lambda s = 0.05$W/m°C (e.g., Barber 1988), $\lambda s = 0.4$W/m°C (e.g., Duret 1983).These differences may be due to mass transfer that has a significant effect on the measurement, e.g., $\lambda s = 3$W/m°C at 55°C (e.g., Duret 1983, Williams 1988, Yamakawa 1988). They have important consequences with respect to the quantity of unmelted UF6 during a fire and the temperature increase of the steel casing. The sensitivity of these parameters is studied later. Other properties that are important for heat transfer are still unknown or not well understood :

- * total hemispheric or spectral emissivity of UF6sol and UF6liq
- * radiation absorption of UF6 gas
- * UF6 equation of state, especially gas compressibility factor
- * properties of supercritical UF6

Initial filling and cooling of the container

The container is generally filled with liquid UF6 at about 100°C in accordance with international recommendations (e.g., ORO 1991). Initially the steel is at ambient temperature and the UF6 solidifies at 64°C. If the quantity of UF6 is such that at 120°C 95% of the volume is occupied, it can be calculated that at the onset of solidification this ratio will have become 85% and the liquid UF6 free surface level is then about 35cm above the horizontal axis of the 48Y vessel.

Given that the density ρ in relation to temperature is the following : ρ= 5090 kg/m3 at 20 °C, ρs= 4920 kg/m3 ρl= 3674 kg/m^3 at 64 °C , then ρ= 4130 - 7.13t kg/m^3 with t in °C ,we can reach the evolution of the filling rate versus temperature (Fig. 1).At ambient temperature the surface area filling ratio is 63% , and if the temperature is uniform the hydraulic rupture will occurs above 144°C depending on the cylinder dilatation.

Assuming that a free horizontal level is maintained, the value of Rap (height of the free level above the axis, divided by the radius) can be calculated as a function of temperature or of relative density, both assumed to be uniform for example at 20°C Rap=0.17, that is to say 10cm above the horizontal axis. In reality, firstly the porosity in the UF6 solid is not equal to zero, secondly a layer of UF6 is deposited at the top, due to mass transfer during cooling, so the ultimate distribution in the container, at ambient temperature, is governed by internal temperatures and the external cooling flow.

Depending on conditions, the filling pattern of the container will reflect one of the cases illustrated in figure 2.

Uncertainties concerning external flow

IAEA regulations for type B transport containers suggest taking a heat flow corresponding to a radiating environment at 800 °C with a factor of 0.9 on the basis of a container emissivity of 0.8. For a shape factor of 1, the maximal value is 54 kW/m^2, dropping to 40 kW/m^2, for instance, when the wall temperature is 500°C. In reality, heat flows can be much higher : recently (e.g., Gregory et al 1989), 3 tests lasting 30 min were carried out with a fire area of 18.3m by 9m engulfing a steel container diameter 1.4m and length 6.4m. Whereas the flame temperatures vary between 540 and 1050°C depending on the reference height, maximal heat flow is about 120 kW/m^2 and average heat flow over the 30 min at least 62 kW/m^2.Another unknown of a real fire is the spatial inhomogeneous repartition of this flow around the container.

MODELLING THE PHYSICAL PHENOMENA

In addition to the preceding aspects, modelling of other aspects related to the behaviour of UF6 inside the container is also not easy. Physical problems and the related modelling will be detailed to some extent below.

Heat conduction in steel shell

Assuming that the input heat flow is known, transient heat conditions in an isotropic medium can be expressed by the classical heat conduction equation.

$$\rho\, Cp\, \delta T/\delta t = \mathrm{div}\,(\lambda\ \mathrm{grad}\ T)$$

The steel shell is made with a commonly used industrial carbon steel. The actual variations on the steel composition do not permit setting any precise value for its thermal conductivity. In addition, the properties of such qualities of steel above 400°C are not well known.

We will take for our calculations: $\rho(kg/m^3)= 7800$ $Cp(J/kg°C)= 460$ $\lambda(W/m°C)= 60$

Heat transfer at the steel-UF6 interface

a) For solid UF6

The small layer of UF6 at the top (see initial geometry figure 2) is assumed to adhere perfectly to the steel. On the input of energy corresponding to its melting, the geometry switches to that of the horizontal interface.

In the upper part, only the radiation at the top is taken into account, taking as parameters the emissivity of steel and that of UF6. In this case gaseous UF6 is assumed to be transparent.

For the lower part, the model assumes the existence of a thermal contact resistance between solid UF6 and the steel shell. Heat exchange can be divided into three parts :(Fig 3 a)

1) a conductive type of heat exchange through the UF6 (assumed to adhere perfectly to the steel) over a surface representing Pc percent of the maximum surface area S of the steel facing the UF6. A conductivity of 0.4 W/m°C is taken but this conductivity will be considered as a parameter of the model. This layer has the same thickness as the thickness of the gas gap described in the following section,

2) a conductive exchange through a gas gap of thickness Eg, assumed to be uniformly distributed over a surface area representing (100 - Pc) percent of S.

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Eg (the thickness of the gas "gap") normally depends on the filling conditions for the UF6 but also on the transient expansion of the steel shell. In our model, this gap is considered to be constant and is supposed to be filled with UF6 gas at the temperature of the steel.Of course, Pc holds as another parameter of the model.

b) For liquid UF6

When the melting temperature of UF6 is reached, the interface between solid UF6 and the steel will fill up and heat exchange from the steel will be increased. The contact temperature between liquid UF6 and steel may be very high and many physical phenomena may appear. For instance, this contact temperature may be well above the critical temperature of UF6. In this case, it is assumed that film boiling will take place. The heat transfer due to film boiling is described with a suitable correlation.

When the steel temperature decreases, transition boiling and, later on, nucleate boiling may occur. Also correlations are used for the evaluation of the limits of the different types of boiling regimes and the description of heat transfer for these regimes.

For example the level of the critical heat flow is evaluated to be about $3\ 10^5$ W/m² for (Tsteel-TUF6)equal to 62°C. A single phase situation is also considered.

Fig. 3 b sketches the model.

At each instant and at the point involved, the vapour pressure Psat corresponding to the internal temperature of the steel shell is calculated. If Psat is below the pressure in the gas blanket then there is local boiling.

The heat input from the steel serves to vaporize the liquid UF6, to the increase of the temperature of the liquefied UF6 and finally to heat up and melt the solid UF6.

As a first approximation, the liquid is considered to stay at melting temperature, so the heat flow from the steel will split into heat for vaporisation and heat for UF6 heat up and melting. The ratio α of the heat serving for vaporisation to the total local heat input is a parameter of the model.

c) For gaseous UF6

Treated as an ideal gas, gaseous UF6 remains transparent to the radiation from the steel shell.

The net evaporation flow rate is used to calculate the pressure in the free volume at each instant. Free volume and transient changes in free volume (increase due to steel expansion, reduction due to melting of UF6) are taken into account.

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Internal heat transfer in UF6 subdivides into two components :

- pure conduction in the solid taking $\lambda = 0.05$ W/m°C
- mass transfer from the melting interface until internal cracks are filled

The later effect is taken into account by means of an global equivalent conductivity in the mass of UF6 that is another parameter of the model.

Internal pressure rise and failure

Two failure modes can be imagined :

* Boiling of UF6 generates a vapour flow resulting in an increase of the internal pressure. The resulting stresses in combination with a deterioration in the mechanical strength of the steel can lead to bursting.

* At any time the internal free volume reduces in relation with the decrease of the density due to melting. If the UF6 is completely liquid, the hydraulic failure occurs at about 148°C (figure 1) or 171°C (e.g., Williams 1988) allowing for expansion of the steel. There are many other pictures which may lead to hydraulic failure :

10% of UF6 not melted and mean temperature of liquid UF6 163°C

30% of UF6 not melted and mean temperature of liquid UF6 208°C

The pressure in the ullage space is not the vapour pressure corresponding to the bulk or maximum liquid temperature, but is calculated from the free volume and the gaseous evaporation flow coming from the previous boiling transfer at the steel-UF6liq interface.

Mechanical behaviour of the steel shell:

It is planned in the future to couple a mechanical model which will describe the effects of the internal pressure and the mechanical behaviour of the steel.

CURRENT DEVELOPMENT

Model

Our numerical development is centred on a general analytical program, based on the finite element method, called ANSYS developed by Swanson Analysis Inc. (USA). Its capabilities with respect to heat transfer analyses (transient conditions, non-linear analysis, radiation in cavities, phase change and convection conditions as a function of temperature) are adequate for our model and leave the possibility open for subsequently coupling mechanical analysis with heat exchange analysis. Parameters can be set for the geometrical input data. For instance, the vertical half section of a container can be obtained after automatic meshing (figure 4).

Sensitivity of the parameters in the initial phase

The actual effort of validation of the model is based on a Japanese experimental work by PNC (e.g., Suzucki 1988). Heating tests are carried out on about a hundred kilograms of UF6 occupying 95% of the inner volume of the container after filling at 120 °C. The container is a cylinder, inner diameter 0.21m and inner length 1m, with a steel shell thickness of 0.03m. Fire is simulated by an electric furnace radiating at 400°C for 10 minutes.

From the experimental results, we used the internal pressure results, the heating element temperature curves, the inner and outer wall temperature curves, and the UF6 temperatures measured at 3mm, 13mm, 23mm and 43mm from the steel shell surface.

When heating begins, the contact resistance between the steel and solid UF6 and the internal thermal conductivity of UF6 directly control the transmission of heat and the moment when UF6 becomes liquid at the interface with an appreciable increase in heat flow at the interface (Fig 5).With reference to the Japanese tests and to the modelling described earlier, we investigate which sets of parameters physically plausible lead to obtaining the main experimental results.We present in figure 6 the result obtained with λs= 45 W/m°C, λi=0.4 W/m°C, Pc= 10%, Eg =1mm and ε_{UF6}=0.9. If λs= 45 W/m°C we obtain also a good agreement for the following other parameters:

λi (W/m°C)	Pc (%)	Eg (mm)	ε_{UF6}
4	1	1	0.9
0.4	10	1	0.4
0.4	50	5	0.9

It is obvious that only one experiment could not lead to an single set of parameters.

It also appears that the radiative heat transfer in the gas gap is negligible for the considered test temperatures but that it would not be in the case of a real fire.

Melting and boiling phases

Subsequent interpretation of the Japanese experimental work (e.g., Suzucki 1988) permits identification of a short phase of natural convection heat exchange with a value close to 6 kW/m^2.

After 35 minutes in the considered test, the pressure gauge records a violent increase in pressure : this is thought to correspond to a nucleate boiling phase. To recover correctly the measured pressure, coefficient α should be adjusted to the value of 0.02 up to 37 minutes then of 0.07 and finally of 0.15 at the end of the test.

GENERAL DISCUSSION

Validation at high temperature

An interpretation UF6 tests (e.g., Malett 1966) has been undertaken. These tests have been conducted up to the burst of the cylinders. The mass of UF6 is low and comparable to that of the Japanese tests; so the same values of the parameters are taken for equivalent internal conductivity and initial contact resistance. The wall thickness is lower , the external heat input in a real fire situation is greater so expansion of the shell should entail a rapid increase in contact resistance. Therefore the main parameters, in relationship to our model, seem to be Eg and Pc .

To obtain hydraulic failure within approximately 9 minutes (corresponding to the experimental result), the following set of parameters is used: λs= 45 W/m°C, λi=0.4 W/m°C, Pc= 10%, Eg =5mm and ε_{UF6} =0.4. An emissivity increase up to 0.9 results in a rupture time reduction of about 1 minute (11%).

48Y Container

It is not physical to use the set of parameters obtained from the interpretation of the Japanese tests (e.g., Suzucki 1988) and the American tests (e.g., Malett 1966) directly for the extrapolation to the 48Y situation, for the following reasons :

* the mass is more than 100 times greater

* cooling on filling takes much longer time with, obviously, consequences on the initial solid UF6 configuration and on the thermal resistance between solid UF6 and the steel shell.

* the equivalent thermal conductivity of the UF6 mass depends on the diameter, which is six times greater in the case of the 48Y. This governs the time to reach melting .

However some calculations were attempted.

Assuming that a radiative heating (800°C with an emissivity of 0.9 on the basis of a container emissivity of 0.8) is applied during 1500sec followed by adiabatic conditions, it appears than the wall temperature becomes so high (more than 500°C when liquid forms after about 10 minutes) that film boiling occurs ; finally the steel and UF6 are predicted to reach a homogeneous temperature equal to 90°C. The same calculation with a 2000sec fire predicts a hydraulic rupture 90 minutes after fire stop.

FUTURE DEVELOPMENT

Our model is still in the development stage. Very soon it will need to take into account :

- a description of the increase in the level of the UF6 and its consequences with the enlarged surface of heat exchange with the steel
- an energy equation for the liquid UF6

On the other hand, model validation by the experimental TENERIFE program (e.g., Casselman et al 1992) seems essential, in particular for unknowns such as :

- the value of equivalent thermal conductivity under conditions representative of a real size container and a fire engulfing the 48Y.
- the type of boiling at the onset of melting which is strongly related to the interface resistance between solid UF6 and steel
- the value of parameter describing the net vapour flow

Nor must we fail to realise that 3-D extrapolation to the case of a fire will always suffer from uncertainties.

The real heat flow of a fire is not known and it is far from uniform in space. Nor must we forget the container valve through which UF6 can escape from the container. On the same lines, information is needed on the possibilities of external cooling in order to prevent from container burst due to heat transfer from the still hot steel to the UF6 when the fire is stopped. Typically, if the average external heat flow is 100 kW/m^2, the energy input over 17 minutes is sufficient to entrain hydraulic failure of the 48Y cylinder. This failure may be delayed but it is unavoidable unless there is external cooling.

NOMENCLATURE

λ	thermal conductivity	(W/m.°K)	D	cylinder diameter	(m)
			P	pressure	(Pa)
ρ	density	(kg/m^3)	S	surface	(m^2)
Cp	thermal capacity	(J/kg°C)	t	time	(sec)
ε	emissivity		T	temperature	(°K or °C)
Φ	thermal flow	(W/m^2)	Δ	difference	
Pc	solid contact percentage		**Indices**		
Eg	gas gap thickness	(mm)	s	solid	
σ	constraint	(Pa or hbar)	l	liquid	
			f	fusion	
α	$\frac{\text{energy for fluid evaporation}}{\text{total energy input to UF6liq- steel interface}}$		i	interface	

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Initial filling and cooling of the container

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0.4	50	5	0.9

It is obvious that only one experiment could not lead to an single set of parameters.

It also appeared that the radiative heat transfer in the gas gap is negligible for the considered test temperatures but that it would not be in the case of a real fire.

Melting and boiling phases

Subsequent interpretation of the Japanese experimental work [4] permits to identify a short phase of natural convection heat exchange with a value close to $6kW/m^2$.

After 35 minutes in the considered test, the pressure gauge records a violent increase in pressure : this is thought to correspond to a nucleate boiling phase. To recover correctly the measured pressure, coefficient α should be adjusted to the value of 0.02 up to 37 minutes then of 0.07 and finally of 0.15 at the end of the test.

GENERAL DISCUSSION

Validation at high temperature

An interpretation of Malett's tests [1] has been undertaken. These tests have been conducted up to the burst of the cylinders. The mass of UF6 is low and comparable to that of the Japanese tests; so the same values of the parameters are taken for equivalent internal conductivity and initial contact resistance. The wall thickness is lower , the external heat input in a real fire situation is greater so expansion of the shell should entail a rapid increase in contact resistance. Therefore the main parameters, in relationship to our model, seem to be Eg and Pc .

To obtain hydraulic failure within approximately 9 minutes (corresponding to the experimental result), following set of parameters is used: λs= 45 W/m°C λi=0.4 W/m°C Pc= 10% Eg =5mm and ε_{UF6} =0.4. An emissivity increase up to 0.9 results in a rupture time reduction of about 1 minute (11%).

48Y Container

It is not physical to use the set of parameters obtained from the interpretation of the Japanese tests [4] and the American tests [1] directly for the extrapolation to the 48Y situation, for the following reasons :

* the mass is more than 100 times greater

* cooling on filling takes much longer time with, obviously, consequences on the initial solid UF6 configuration and as on the thermal resistance between solid UF6 and the steel shell.

* the equivalent thermal conductivity of the UF6 mass depends on the diameter, which is six times greater in the case of the 48Y. This governs the time to reach melting .

However some calculations were attempted.

Assuming that a radiative heating (800°C with an emissivity of 0.9 on the basis of a container emissivity of 0.8) is applied during 1500sec followed by adiabatic conditions, it appears than the wall temperature becomes so high (more than 500°C when liquid forms after about 10 minutes) that film boiling occurs ; finally the steel and UF6 are predicted to reach a homogeneous temperature equal to 90°C. The same calculation with a 2000sec fire predicts a hydraulic rupture 90 minutes after fire stop.

FUTURE DEVELOPMENT

Our model is still in the development stage. Very soon it will need to take into account :

- a description of the increase in the level of the UF6 and its consequences with the enlarged surface of heat exchange with the steel
- an energy equation for the liquid UF6

On the other hand, model validation by the experimental TENERIFE program [10] seems essential, in particular for unknowns such as :

- the value of equivalent thermal conductivity under conditions representative of a real size container and a fire engulfing the 48Y.
- the type of boiling at the onset of melting which is strongly related to the interface resistance between solid UF6 and steel
- the value of parameter describing the net vapour flow

Nor must we fail to realise that 3-D extrapolation to the case of a fire will always suffer from uncertainties. The real heat flow of a fire is not known and it is far from uniform in space. Nor must we forget the container valve through which UF6 can escape from the container. On the same lines, information is needed on the possibilities of external cooling in order to prevent from container burst due to heat transfer from the still hot steel to the UF6 when the fire is stopped. Typically, if the average external heat flow is 100 kW/m², the energy input over 17 minutes is sufficient to entrain hydraulic failure of the 48Y cylinder. This failure may be delayed but it is unavoidable unless there is external cooling.

NOMENCLATURE

λ	thermal conductivity	(W/m.°K)
ρ	density	(kg/m^3)
Cp	thermal capacity	(J/kg°C)
ε	émissivity	
Φ	thermal flow	(W/m^2)
Pc	solid contact percentage	
Eg	gas gap thickness	(mm)
σ	constraint	(Pa or hbar)
α	$\dfrac{\text{energy for fluid evaporation}}{\text{total energy input to UF6liq- steel interface}}$	

D	cylinder diameter	(m)
P	Pressure	(Pa)
S	surface	(m^2)
t	time	(sec)
T	temperature	(°K or °C)
Δ	difference	
Indices		
s	solid	
l	liquid	
f	fusion	
i	interface	

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FIGURES

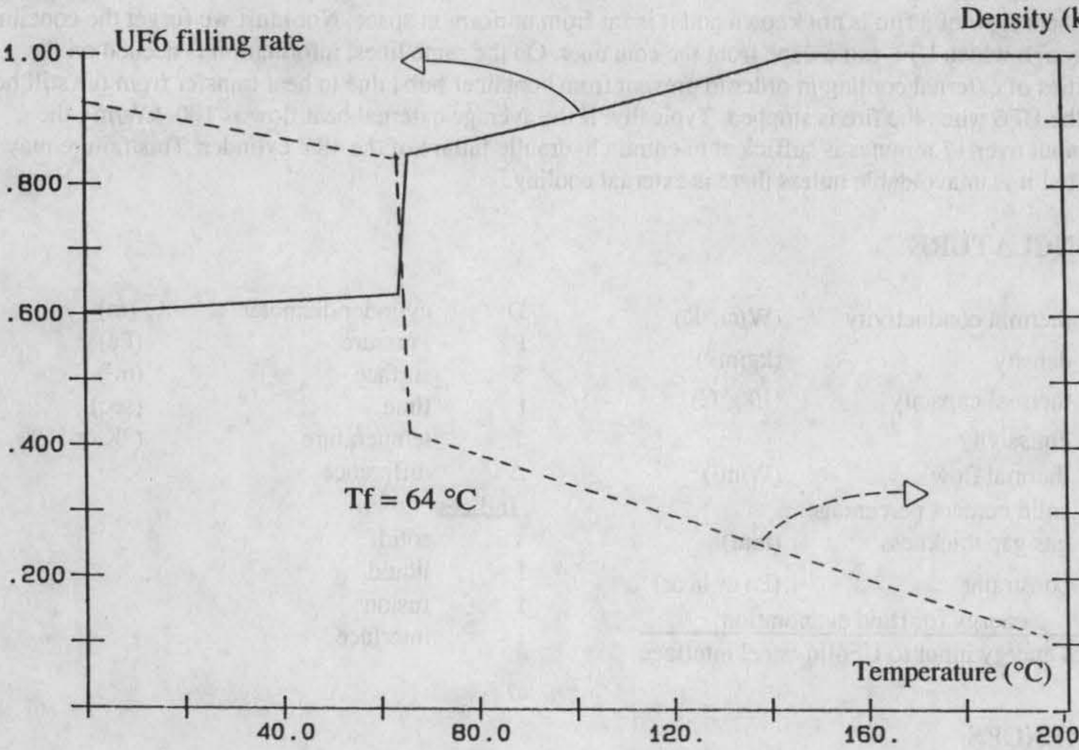

Fig.1 Filling rate and UF6 density versus temperature

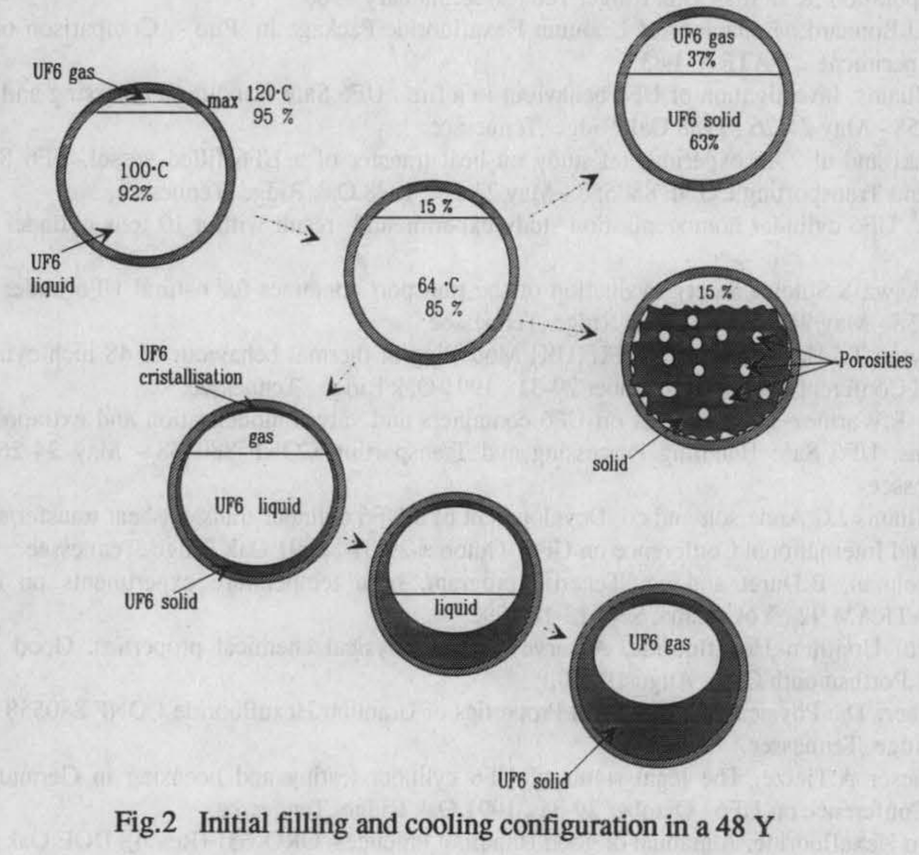

Fig.2 Initial filling and cooling configuration in a 48 Y

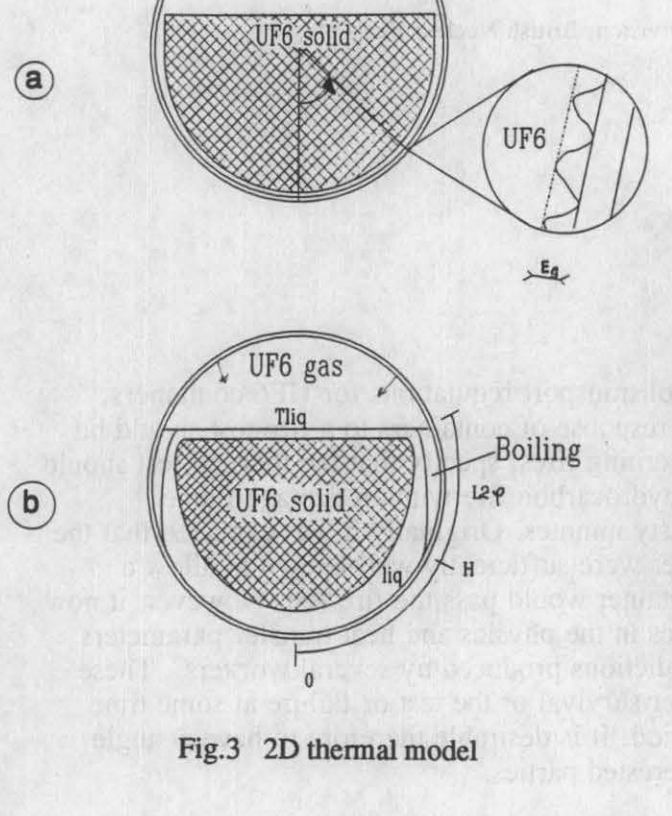

Fig.3 2D thermal model

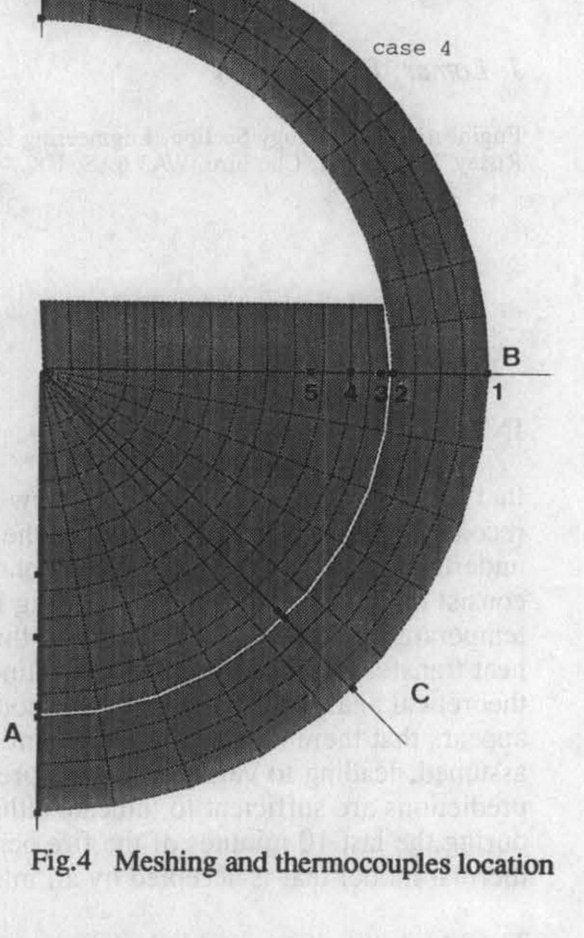

Fig.4 Meshing and thermocouples location

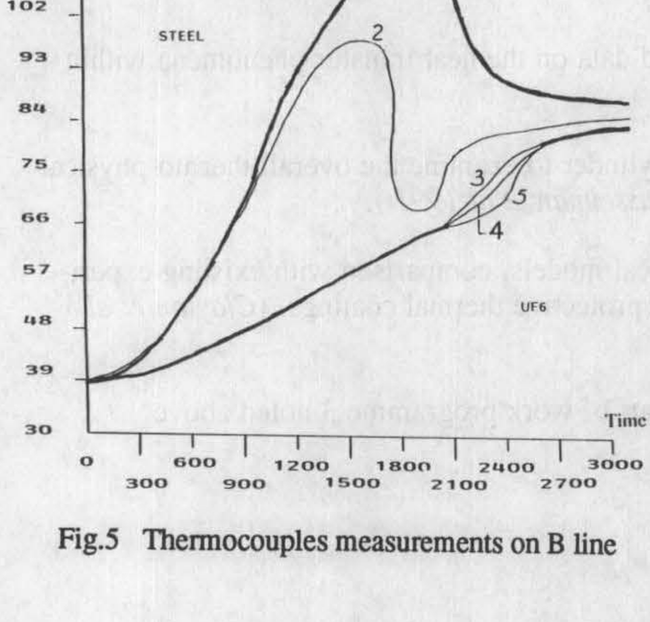

Fig.5 Thermocouples measurements on B line

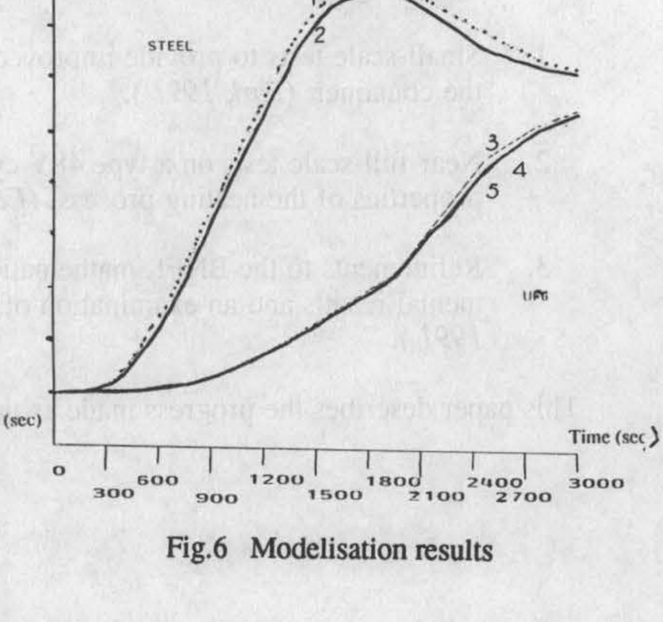

Fig.6 Modelisation results