BEHAVIOR OF A UF6 CONTAINER IN A FIRE : MODELLING AND VALIDATION WITH DIBONA CODE

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SUMMARY

The purpose of this study is to predict the thermohydraulic behaviour of a UF6 container in a fire. The coupling of the thermal model with the mechanical model must be considered to define if the resistance of a 48Y container can be guaranteed. To answer this question a 2D model using the finite elements code Ansys was developed. The scenario and the phenomenology have been introduced in the model from the analysis and the interpretation of the Tenerife tests.

This model is validated with 3 tests of the Tenerife program (TEN2, TEN4, TEN6). In the whole, the numerical results correlated successfully with the experiments.

The extrapolation to a IAEA fire test on 48Y container is presented at the end of this document.

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 Tenerife containers (Saroul et al. 1995, CRP meeting 1997), and our model has been 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. This model is the pursuit of the work begun by Duret B. (1992). So, only the new developments will be 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 may not in 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. 1992) 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 constraints of 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.

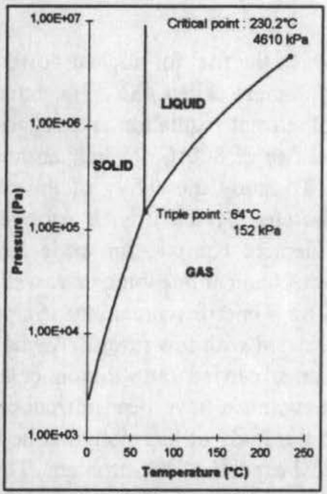


Figure 1 : UF6 phase diagram

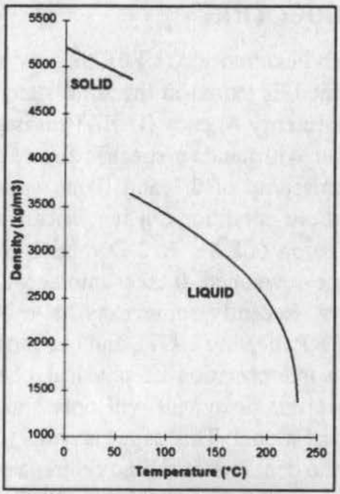


Figure 2 : Density of UF6

Physical Properties of UF6

We referred to the compilation of Anderson J.C. et al. (1994), that assembles and presents 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. (1995) by analysing the cooling process of the UF6 after filling.

For the time being, the modelling of the UF6 in contact with the UF6 gas is approximated by a horizontal surface (Figure 3). The top crust is assumed to have a uniform thickness. The initial height of UF6 is determined by the model depending on the thickness of the crust. The steel and the UF6 are modeled by 2-D quadrilateral thermal elements with 4 nodes.

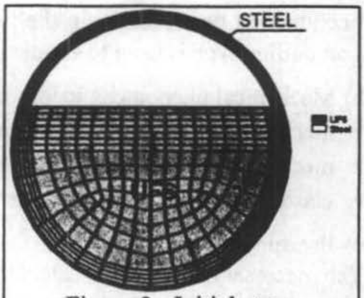
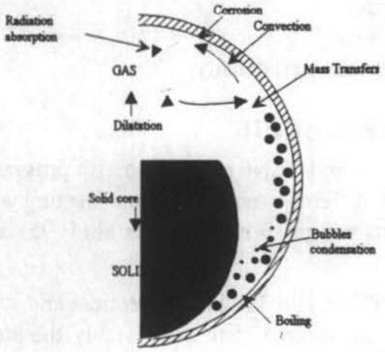


Figure 3 : Initial state.

PHYSICAL PHENOMENA INTRODUCED IN THE MODEL

A series of multiphase and transient phenomena takes place within the container. The two following figures summarize the main physical phenomena firstly with upper crust (figure 5) and secondly without (figure 4).



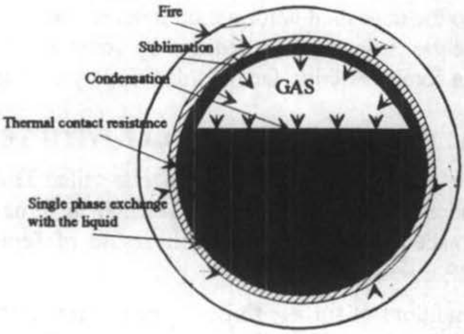


Figure 4 : Internal transfers without the crust Figure 5 : Internal transfers with a crust.

The detailed approach have been already presentated during PATRAM'95 (Pinton et al. 1995). Nevertheless we list below the main phenomena introduced in DIBONA code.

a) Thermal phenomena incorporated in the model are :

- radiation exchange through the vapour gap (which is an absorbing gas) between the inner top of the container and the free surface of UF6 liquid or solid,
- radiation exchange between the fire and the outer tank,
- thermal contact resistance between solid UF6 and steel,
- pool boiling heat transfer (q_{boil}) at the steel/UF6 interface by taking into account subcooled, surface state, orientation of the surface and pressure effects. Evaporation (q_{evap}) and convection (q_{liq}) heat flux linked with boiling are also included ($q_{boil} \approx q_{evap} + q_{liq}$),
- UF6 melting,
- mechanism of solid core sinking,
- volume expansion of UF6 (which is very important during the solid/liquid phase change),
- steel shell expansion,
- vapour condensation and sublimation on the solid or liquid interface in contact with the gaseous plenum,
- vapour bubble condensation in the subcooled liquid,
- conduction heat transfer in the solid UF6 and steel,

- conduction heat transfer in the liquid UF6 (an equivalent heat conductivity which depends on boiling type is used to simulate the heat exchange increase caused by boiling).

b) Mechanical phenomena incorporated in the model are :

- mechanical stress induced by steel expansion and by temperature gradient,
- mechanical stress induced by internal pressure,
- elastic and plastic deformations.

c) Pressurisation :

The necessary values to calculate the pressure are the mass, the volume, the average temperature in the vapour gap and the compressibility factor, these three parameters being linked to the pressure by an equation of state. The model determines :

- the vapour quantity created at the boiling surface which arrives in the gaseous plenum,
- the vapour quantity condensed or sublimated on the free surface of UF6.

By using those two parameters of gas can be deduced .

To calculate the gas volume, we take into account the UF6 and steel thermal expansion, but also the tank shell deformation induced by internal pressure.

The gas temperature is deduced by doing an energy balance.

The compressibility factor is deduced by the gas temperature and density.

VALIDATION OF THE MODEL WITH TENERIFE RESULTS

The model is validated by three tests called TEN2, TEN4 and TEN6 of the Tenerife program, where an UF6 container is placed in a furnace with different ranges of time heating and furnace temperatures. The main results of Tenerife tests are given by Saroul et al (1995) and CRP meeting (1997).

Conditions of surface finish of the external surface of TEN4 and TEN6 are identical and well known whereas this is not the case for TEN2 where the surface state and notably the steel emissivity is not well identified. Moreover container TEN4 and TEN6 have not undergone a first heating as TEN2. The internal configuration of the UF6 in TEN4 and TEN6 is such that it exists just after the filling. Unlike TEN2 and TEN6, the measure of the two pressure sensors of TEN4 is uniform and does not saturate (TEN6 : a defective sensor).

The test TEN4 is considered as that of reference for the adjustment of parameters and the validation of the model. Nevertheless, a calculation with conditions of the test TEN2 and TEN6 has been correlated, but will not be presented in this paper.

Validation with TEN4 test

The initial temperature is 17°C for a 19mn30s (1170s) heating at 800°C. The inconel and steel emissivities are 0.8 and 0.6 respectively. These values were defined from the experimental results of the calibration test (CRP meeting 1997) with an empty container. The meshing is identical to that previous paragraph.

The positions of the thermocouples mentioned in this paragraph are illustrated on figure 6.

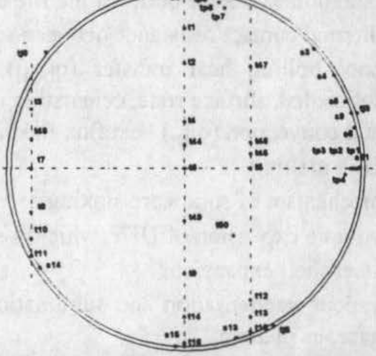


Figure 6 : Instrumentation of TEN4 container in the median plane

The following uncertainties have been taken into account :

- some parameters unknown experimentally for example the thickness of the top solid crust, the percentage of the contact surface at the steel/solid UF6 interface,
- the thermal conductivity of the liquid which depends on the liquid agitation (influenced by the type of boiling),
- adaptation of the parameters linked with relations supplied by the literature.

Steel temperatures

The steel temperatures for the lower part of the container are in quite good agreement with experimental findings (Figures 8-9). 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 modelled.

Consequently, the phenomenology presented in CRP meeting (1997) is correlated. After the collapse of the top crust (at 750 s, P=1.5 bar), the regime becomes film boiling where the wall temperature decreases slowly. Then near 1050 s, the transition phase begins where the flux increases as the superheat decreases. The temperature then drops rapidly until equilibrium is achieved at about 840s ($q_{Boil}=q_{Fire}$). The regimen is now nucleate boiling and the temperature evolves with $T_{sat}(P)$. When the heating is stopped (1170s), the energy supplied by the furnace is no longer sufficient to maintain nucleate boiling. Natural convection sets in and the temperature of the steel decreases.

UF6 temperatures

UF6 temperatures in the upper part

The general trend of the temperatures in the model is similar to that found experimentally (Figures 7-10) :

- the time the liquid appears, starting from the top to the bottom, is reasonably correlated,
- the stratification of the liquid is present, The modelling of transfers in the liquid by conduction is suitable.

UF6 temperatures in the lower part

In this case, the correlation of Tenerife measurements remain uncertain because of the perturbations induced by the tubes supporting the thermocouples as shown in Pinton et al 1995. These measurements only reveal the presence of solid UF6 at the level of the lower thermocouples. This presence is confirmed by the model (Figures 11-12 : $T_{UF6} \leq 64°C$) except for T8 and T10 thermocouples placed close to the wall. However, they remain in the vicinity of the solid/liquid frontier.

Inconel temperatures

The temperature profile evolution given by the model in the heating or cooling phase is correctly simulated as illustrated on figures 15 and 16.

Gas temperatures

The calculated temperature evolution follows that of the model (Figure 14). Note that the mean temperature is an arithmetic mean of the local gas temperatures T1, T2 and T_{sat}. This result indicates that the determination of the energy balance and of the gas temperature is correctly done.

Pressure

The pressure computation (figure 13) is in quite good agreement with the experimental value (figure 18). The pressure evolution, in the mdoel, was adjusted solely by acting on mass transfers at the liquid/gas interface. This simply entailed adapting the thermal conductivity of the liquid, on the assumption that the quantity of vapour arriving in the gas blanket is correctly modelled. During film boiling and transition boiling, the agitation of the liquid is high and the conductivity chosen is 1000 W/m°C. For nucleate boiling where the liquid is stratified (less agitation), this conductivity is reduced to 25 W/m°C. Once the heating is stopped, it diminishes proportionally with the quantity of vapour arriving in the gas blanket.

Others

Figure 17 shows that the solid keeps to the bottom of the container, that the thermal stratification in the liquid is well represented, and that the interface temperature is practically uniform over its whole surface. The volume of gas increases slightly at the beginning because of the sublimation (figure 20). Then after the rupture of the top crust boiling appears and leads to a strong melting of the solid and a rapid decrease of the gaseous volume. Then it decreases slowly during the cooling phase. Nevertheless, at 7000 sec the volume is increased by about 20% in comparison with the initial value. The mass of gas is similar to that of the pressure (figure 19). It means that the mass of gas is the main factor acting on the pressure. The figure 21 present UF6 and steel temperature cards until 3000 s with an increment of 500 s. The profile at 500 s shows the strong thermal contact resistance. In fact, the steel temperature is 200°C whereas UF6 is still solid (T_{UF6}<64°C). Between 500 and 1000 s, the top crust collapsed (750 s) and a expansion layer got activate (980 s). At 1000 s the thermal gradient in steel at the liquid/gas interface is considerable : the wall temperature in contact with the gas is about 450°C and that in contact with the liquid is 150°C. UF6 melts strongly during the heating phase (t<1170 s) and remains always at the bottom of the container. During the cooling phase, the solid shape does not evolve a lot and steel temperatures decrease progressively.

Validation with the other test TEN6 and TEN2

A calculation with initial and boundary conditions (different from each other) for the test TEN2 and TEN6 was undertaken, but they will not be presented in this paper. Nevertheless the numerical results are used in good agreement with the experimental TN22TN6 results. It shows that the adjustment of the parameters used with TEN4 test are correct.

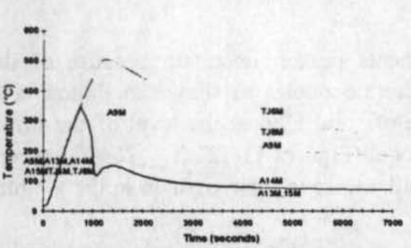


Figure 7 : Steel temperature/TEN4 modelling

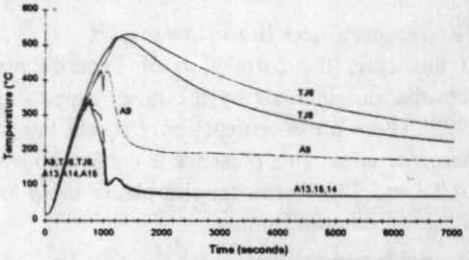


Figure 8 : Steel temperature / TEN4 test

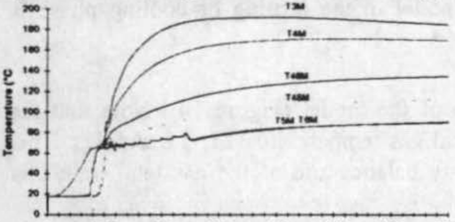


Figure 9 : Upper UF6 temperature/TEN4 modelling

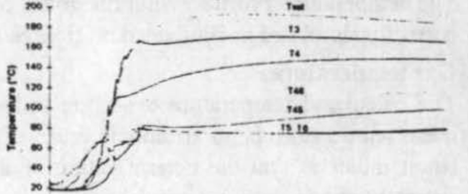


Figure 10 : Upper UF6 temperature/TEN4 test

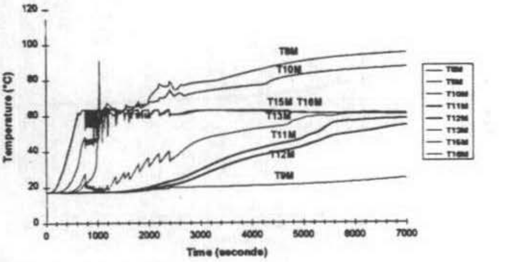


Figure 11 :Lower UF6 temperature/TEN4 modelling

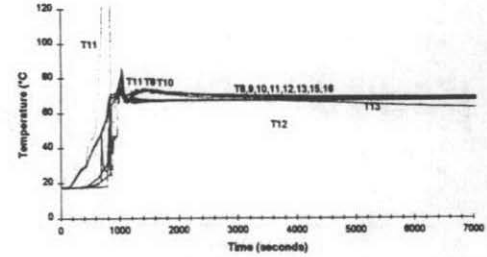


Figure 12 : Lower UF6 temperature /TEN4 test

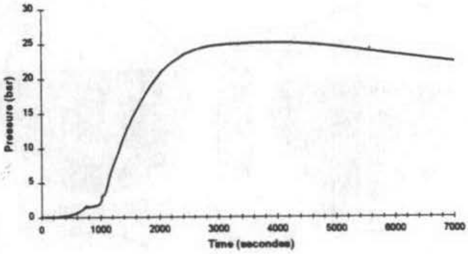


Figure 13 : Pressure evolution / TEN4 modelling

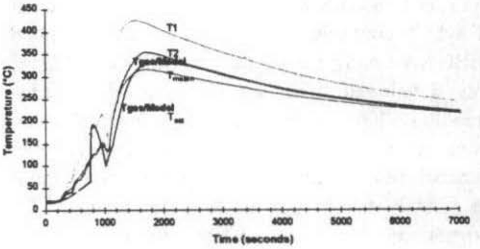


Figure 14 : Gas temperature of the model

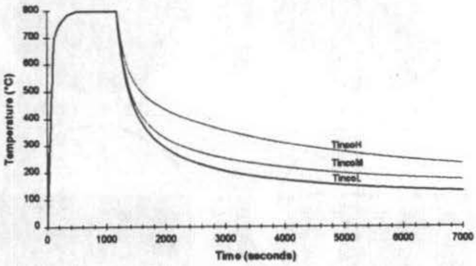


Figure15 : Inconel temperatures/ TEN4 modelling

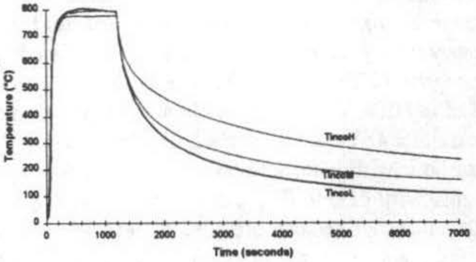


Figure 16 : Inconel temperatures / TEN4 test

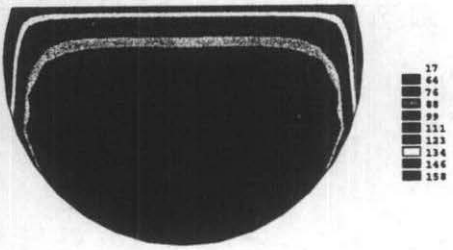


Figure 17 :UF6 (1500 sec)/ TEN4 modelling

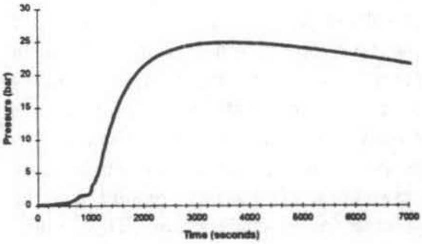


Figure 18 : Pressure evolution / TEN4 test

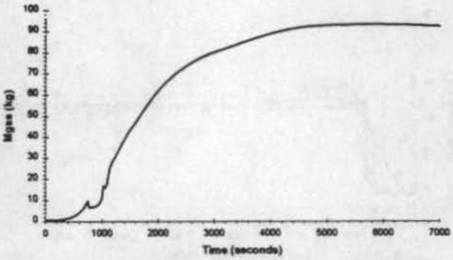


Figure 19 : Mass of gas/TEN4 modelling

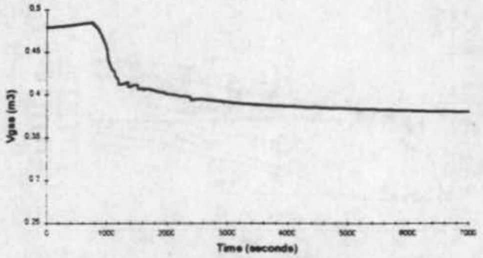


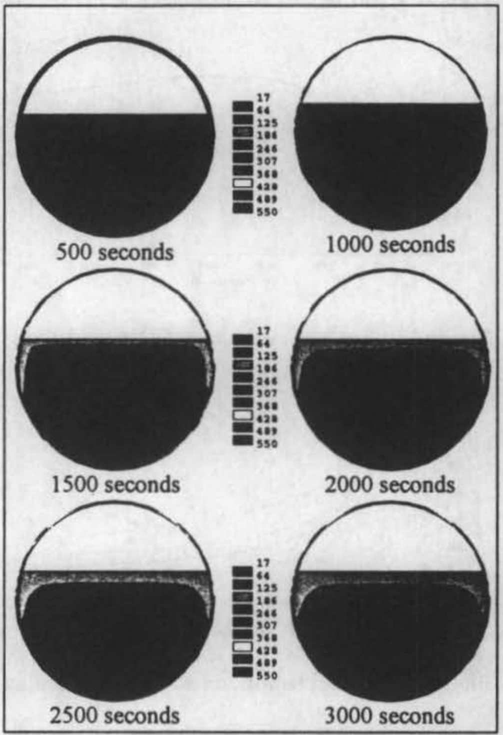
Figure 20 : Gas volume evolution/TEN4 modelling

CONCLUSION

In order to predict the thermohydraulic behaviour of a UF6 container in a fire, a 2D model called DIBONA (using the finite elements code Ansys) was developed. The scenario and the phenomenology have been introduced in the model from the analysis and the interpretation of the Tenerife tests. Many of these phenomena cannot be directly taken into account by Ansys (UF6 expansion, solid core sinking, thermal contact resistance, pressurisation, mass transfers, ...). Therefore, many modifications, under macro commands, have been brought to the code to take them in consideration. This final model has been successfully validated with 3 tests of the Tenerife program (TEN2, TEN4, TEN6).

Using DIBONA, it is possible to extrapolate in a case of a 48Y container in a IAEA fire test of half an hour with a steel emissivity of 0.8 and a fire emissivity of 0.9. The calculations show (figure 22) that the critical pressure is reached before 30mn and that a maximum steel temperature greater than 600°C exists. Above the critical conditions of the fluid (46.1 bar and 230.2°C) the model is unusable.

In fact, the distinction between the two phases is impossible (surface tension and latent heat of vaporisation are zero) and mass transfers do not exist. The computation method of the pressure is unstable and heat transfers are considerably modified. Nevertheless, the model can be used for the evaluation of internal pressure and the container steel roof temperatures. This model does not claim to be a perfect model of all the phenomena, but can take the credit for including them all and describing their general evolution correctly.



500 seconds

1000 seconds

1500 seconds

2000 seconds

2500 seconds

3000 seconds

Figure 21 : Temperature cards/TEN4 modelling

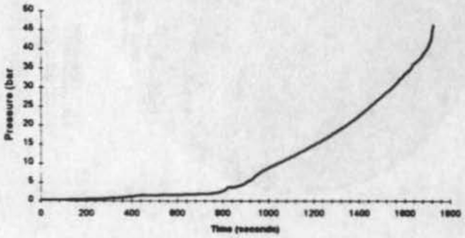


Figure 22 : Pressure / IAEA fire

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REFERENCES

Anderson JC, Kerr CP and Willams WR, *Correlation of the thermophysical properties of UF6 over a wide range of temperature and pressure*, Oak Ridge National Laboratory, Martin Marietta, ORNL/ENG/TM-51, Oak Ridge,TN, August 1994.

Casselman C et al., *High temperature experiments on a 4 tons* UF_6 *container Tenerife program*, PATRAM 92, Yokohama, Sept 13-18, 1992.

CRP IAEA meeting on UF6. French research program, *Behavior of a 48Y cylinder in fire Thermal results and Structural Resistance Evaluation.*, Vienna, Sept 1-5, 1997.

Duret B and Seiler JM, *2D modeling of a UF6 container in a fire*, PATRAM'92, Yokohama, Sept 13-18, 1992.

Duret B and Bonnard JC, *Interprétation des mesures faites au cours du remplissage en UF6*, note CEA STI/LASP/95.04, Grenoble, April 1995.

Pinton E et al., *Interpretation of TEN2 experiment*, Third International UF6 conference, Paducah, KY,Nov 28 - Dec 1, 1995.

Pinton E et al., *Modeling of the behavior of a UF6 container in a fire*, PATRAM'95, Las Vegas, Dec 3-8, 1995.

Saroul J et al., *Tenerife program UF6 transport container under fire conditions - experimental results*, Third International UF6 conference, Paducah, KY,Nov 28 - Dec 1, 1995.

Suzuki et al., *An experimental study on heat transfer of a* UF_6 *filled vessel*, UF6 Safe Handling Processing and Transporting CONF 880558, Oak Ridge,TN, May 24-26, 1988.