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RAPPORT DAS N° 555e

THERMAL TESTS ON UF6 CONTAINERS AND VALVES MODELISATION AND EXTRAPOLATION ON REAL FIRE SITUATIONS

B. DURET, P. WARNIEZ

Uranium hexafluoride - safe handling processing
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ABSTRACT

From realistic tests on containers or on valves, we propose a modelisation which we apply to 3 particular problems :

- Resistance of a 48 Y containers, during a fire situation.
- Influence of the presence of a valve.
- Evaluation of a leakage through a breach, mechanically created before a fire.

I. INTRODUCTION

The resistance evaluation to fire of the UF 6 container, type 30 B or 48 Y, without a protection shell, is the principal matter of our study. A preliminary work had been the purpose of a communication to the PATRAM 83 [1]. We are continuing with this work in the present note, by improving the anticipation of the behaviour of a package supposed to be impervious, but also, by supposing a leakage at the valve and in the case of a breach mechanically created before a fire.

II. INTACT CONTAINER AND TIGHT VALVE

Because the physical characteristics of the UF 6 cannot be simulated in a satisfying way, we have to base ourselves on tests done with UF 6.

In the case of a generalized fire applied to a container without a shell, we only have tests conducted in 1965 [2]. The experiment consisted in measuring the exposure time to a hydrocarbide fire, until rupture of the small containers (maximal mass 113 kg for UF 6 against 2.3 tons for 30 B and 12.6 tons for 48 Y).

For two reasons, the interpretation of these tests can by considered as partial :
- Unknowledgement of the type, of the thermal level and of spreading in space of the external heat flow.
- Lack of intrumentation in the container.

Other heating experiences exist, for example, a 10 tons instrumented container [3] or the experience conducted by the PRNFDC [4], but in both cases, the heating temperature, never exceeds, respectively 93°C and 400°C.

II.1. Modelisation of physical phenomena

a) External received heat flow in a container during a fire :
The AIEA reglementations, for type A packages (which does not at all correspond to the classification of a 48 Y or 30 B package) advises to suppose that the temperature is equal to 800°C and by choosing emissities of .9 for the flames and .8 for the envelope of the container. This means that the received heat flow depends on the steel temperature, therefore, on the internal thermal flow between the envelope and the UF 6.

Therefore, at a 20°C temperature, the heat flux will be 54 000 W/m^2 and, for example, if the steel temperature is 500°C the heat flux will be 40 000 W/m^2.
In addition to the radiative heat flow, must be added a part of natural convection (10 % of the radiant flow) and of forced convection heat transport.

A recente note by F. NITSHE [5] indicates that, during a hydrocarbide fire around some small containers, the heat flow value is of 25 $W/m^2{}^\circ C$.
The real heat flow, depends on the container's size compared with the size of the fire ; we can base on tests carried out in France, with a crude oil fire, covering a surface of 2000 m^2 [6] : it appeared, that the flame temperature decreases fast with altitude (which is maximum of 1 meter) and may reach 1250°C, an average radiance of the flames has been estimated at 30 000 W/m^2.
Depending on the case studied , (realistic calculation or pessimiste risk estimation) we can choose one of the previously described cases, as to introduce it as an incoming parameter for the calculations.

b) Internal transfers in the container
Principal physical phenomena which may have an influence :
- Conduction inside the steel lining
  - . well mastered (in spite of the lack of precise data on λ, ρ , Cp of the ordinary steel, as a function of the temperature).
- Thermal resistance between steel and UF 6
  - . depends on the type of filling (liquid ou gaseous)
- Thermal transfer internal to the UF 6
  - . Radiation : knowledge of an emissity of the UF6 in a solid state
  - . conduction : thermal problems with the circulation of UF 6 (solid, liquid, gaseous)
  - . Effect of a triple point at a temperature of 64°C
  - . Influence of UF 6 gas (at 54°C the vapour pressure above solid UF 6, equals 1 Bar) on the transfer of the mass inside the cracks (heat pipe effect).

. Unknown convection
. Unknown boiling
. Critical point at 230°C. State equation.

In addition to these phenomena, we must add some internal mass transfers, due to the important vapour pressures (70°C 1.8 Bar, 100°C 4.1 Bars and 120°C 6.5 Bars) ; such transfers may considerably increase the apparent thermal conductivity ; it may then, be multiplied by a factor 10, between ambiant temperatures and 65°C [1].
Our work is actually leading us to propose an internal heat transfer model), which has the following principal characteristics :
- Transient conduction inside the steel envelope
- Radiation heat transfer and conduction to the in-interface steel - Solid UF 6
- Apparent good conductibility in the UF 6, allowing for homogeneous temperature
- Increase of heat transfers while changing phases, from solid to liquid : transfer by boiling after phase changing at 64°C.

c) Rupture
Rupture happens if the internal pressure involves an over-strain of the rupture limit of the steel or if the liquid UF 6 fills all the available volume. This is a hydraulic rupture.
It appears that, in the case of the UF 6 thermal model that we have chosen, the hydraulic rupture appears before the mechanical rupture.

II.2. Application to real tests

The interpretation of the japanese experiments [4] on 110 Kg of UF 6 containers electrically heated permits to verify our model of heat transfer in the UF 6.
Main characteristics of a test at 400°C :
- Heating by radiation of electrical resistors
- Good knowledge of the STPT 38 steel temperatures
- Temperature measurements in the UF 6.

The application of our model has been accomplished by using a one dimensional transient approach.

The table 1 resumes the principal date used for the calculation.

Table 1

		SI Value
Geometry :	External diameter	.267
	Steel thickness	.0286
	External surface	1.286
	Internal volume	
Steel :	Thermal conductivity	55
	Density	7850
	Heating capacity	490
	Coefficient of thermal expansion	$1.2\ 10^{-5}$
	Density at 20°C	5090
	Density at 64°C (solid)	4920
UF 6	Density (liquid)	4130-7.13 t (t in °C)
	Heat capacity (solid)	487
	Heating capacity (liquid)	558
	Gas thermal conductivity	.007
	Fusion heat at 64°C	54480

The heat exchange between steel and UF 6 is controled through a gas film (the initial thickness is 0,9 mm); choosing an external emissivity of 0.6, which includes the radiation effect and a part of the convection linked to surrounding air, we then obtain, a steel temperature equal to 96°C after 20 minutes and 111°C at the 30th minute, the internal exchange by radiation, with an emissivity of 0.3 is equivalent to the exchange by conduction through the film ; about 700 W/m².
A calculation compared to a test is shown in fig.1. Steel temperature falls, when the UF 6 joins the melting point which is obtained at the 33 rd minute. then appears an homogeneisation of the temperature between steel and UF 6, the temperature is under estimated at 72°C, which means that the external heat flow, during the heating period, may be more important.
When is noticed, on the one hand, that the steel temperature does not exceed 120°C, and on the other hand, that the diameter of the container is only 1/6 th of a 48 Y. Then we will come to the conclusion that extrapolation of this model, on a big UF6 container during a fire at a temperature of 800°C or 1000°C, is hazardous.
Nevertheless, by to calculating the tests conducted by MALETT [2] (the steel has arrived at a temperature of 540°C during the test) we obtain a rupture after around 8 minutes if we suppose an external average emissivity of .7. This time is experimentally verified.

II.3. EXTRAPOLATION ON A FIRE AROUND A 48 Y

Supposing an internal heat transfer model, identical to the previous case, we study the time until rupture, with two assumptions :
- First case : maximum external flow with a temperature of 800°C and an emissivity Fire-Steel of .9 and .8, respectively adding a natural convection flow around the cylinder.
- Second case : more realistic external flow, with a value of 30 kW/m².

On the other hand we suppose that all the external surface receives the heat flow.
In fig. 2 we give the transient temperature histories between steel and UF 6, in both cas. By calculating the steel temperatures, which are important (above 500°C), we obtain internal radiative heat flows, which are 10 to 30 times above the heat flow due to conduction through the gas film.
In the first case, the hydraulic rupture (T of UF6 148°C) happens after 41 minutes and 57 minutes in the second case.

III. LOSS OF TIGHTNESS AT A VALVE

UF 6 industrial containers (30 B or 48 Y) are equipped with a valve and a plug. For the following reasons, we think that such accessories seem to be specialy sensible, in the case of a fire :
1) The tightness of the valves and of the plug in obtained by using a layer of alloy which is applied on their screwing points. Lead-tin alloy melts at 200°C.
2) The valves are made with materials having different thermal dilatation coefficients. For example the body is made of aluminium and the rod is made of monel (See fig. n° 3).

III.1. Experiment

The experimental system, sketched on fig. 4, allows a progressive or a sudden heating of a complete lot of valves.
The electrical oven, equipped with a steel tube used as a protection in case of a rupture of the valve, creates a hot surrounding (800°C).
In order to avoid an eventual oxidization, we have chosen nitrogen for the tests. A metallic container

fixed on the support, with an interposed gasket is used to receive the valve leakage flow. The gas is then conducted to nitrogen-water heat exchanges permitting to lower the nitrogen temperature to it's normal temperature before passing in a flow meter

a) Progressive heating :
The temperature measurements and the leakage measurements are conducted by imposing the oven temperatures by steps (100, 200, 300, 400°C) and then by varying the pressure (5, 10, 20 and 30 bars).
Five valves have been tested, (see recording in fig. 5). The melting of the alloy is evident, nevertheless, we have noticed the beginning of a leakage at 100-150°C, which seems to indicate that a leakage, can be initiated by a differential dilatation of valve elements, even more important leakages that the thermal gradient, internal to the valve, is imimportant.

b) Sudden heating :
All the devices are the same, but the valve and the plug are screwed on a sleeve which is welded same measurement thermocouples are in place.

The experiment consists to heat the oven, which is suspended above the valve, until obtention of a steady temperature (800°C) inside the steel tube and bring the oven over the valve, which is maintained at a pressure of 5 bars. The valve temperature increases, then we can note the time it takes and the temperature level of the surrounding space at the beginning of the leakage.
If the gasket, made of PTFE (see fig.3) does not exist, a leakage appears after 1 minute and 25 seconds, when the scriwing is at a temperature of 78°C.
For a completely equipped valve, the leakage starts when the temperature is of 200°C (melting temperature of the lead-tin alloy) after 4 minutes 50 seconds.

III.2. Extrapolation on gaseous UF 6

If the valve has lost it's tightness, the leakage depends, of the motor pressure, or during a fire, the leakage depends on the obtained temperature, which depends on internal thermal flow to the UF 6. Supposing that the UF 6 temperature is of 140°C, before a hydraulic rupture, the pressure would cause a leakage of about 2 $N.m^3/h$ of nitrogen in the experiment .
Supposing adiabatic behaviour of the UF 6, then :

$$Q_{UF6} = Q_{Nitrogen} \quad \frac{MN_2}{MUF6} \quad \frac{\dfrac{2\ \gamma\ UF6}{\gamma\ UF6 + 1}}{\dfrac{2\ \gamma\ N_2}{\gamma\ N_2 + 1}}$$

Based on the previous choices $Q_{UF6} = 0.55\ m^3/h$ which means that after hydrolysing we have an emanation of 0.5 g/s in HF.

IV. BREACH FOLLOWED BY A FIRE

The calculation supposes that the thermal flow incoming through the steel envelope allows to sublimate a certain quantity of UF 6 which will be then, evacuated out of the 48 Y container. We have chosen the model described in paragraph I, by adding the sublimation heat which is equal to $1.38\ 10^5$ J/Kg.

- First case : 800°C fire, of a duration of 30 minutes, with a fire emissivity of .9 and a container emissivity of .8 we considered also a contribution of natural convection.
- Second case : the heat flow is 30 000 W/m^2 and the fire lasts 15 minutes (see results in fig. 6 and 7) In any case, the steel envelope exits and has an influence by it's thermal inertia ; the heating continues up to the time at which, an external intervention permits a fast cooling of the steel.

In the following table we present the main results:

Table 2

	UF6 Density evacuated (Tons)	
	if intervention	without intervention
First case 800°C duration : 30 minutes	3.44	5.9
Second case 30 kW/m^2 duration : 15 minutes	1.1	2.7

If the breach is small enough, the pressure drop, and a shrinking related to a UF 6 deposition, can decrease considerably the leakage flow.

V. CONCLUSION

The uncertainty of our evaluations resides on two important points.

1. External heat exchange : it is variable because of the homogeneousity of the flames and the smoke influence, and the modification of the container's emissity. A modelisation is utopic because each fire is a different case, which depends on :
- the type of hydrocarbide
- the exact position of the container

We think that this exchange must be an incoming parameter of the calculation program, knowing that it can involve variations on the rupture time, for example around 50 %.

2. Steel-UF 6 heat transfers
Because we actually, don't have high temperature heating experiments with big diameter UF 6 containers, we cannot give a definition of the moments when ruptures will happen on industrial containers, as the estimations can vary between 15 minutes and 1 hour on different models, but joining the rupture time, in the MALLET tests [2].
Different assumptions are possible :
a) Considering the important UF 6 density, the heating can stay limited to the peripherical zones generating some growing pressures which are localised at the periphery.
b) A part of the UF 6 can be liquefied and would be accumulated at the bottom of the container, then two possibilities appears :

- The container is involved intirely by fire, including the bottom and involves an important spraying of UF 6, leading to a global heating with an excellent heat transfer and an early rupture of the envelope.
- The fire does not reach the liquid zone, and the existence of a bad coefficient of the heat transfer between the lining and the solid UF 6 delays the UF 6 heating and the rupture of the container.

The remaining problem is to determine the right manner, the conditions of phase changements, the mass transfers inside the container which would determine the conditions of the rupture time, as well as the quantities of UF 6 which are susceptible to be released if there is a leakage at the valve.
Actually a test is envisaged. It's principle is the *following* :

- Transient heating of a 1.3 m portion of a container 48 Y correctely instrumented, internaly and on the shell surface.
- Good knowledge of the external transient heat flow at a thermal fire level of 800 to 900°C.
- Stopping the test before rupture, but with an oven situated in a spacious surrounding, allowing a release of the UF 6.

The main parameters must be :

- The rate and the type of filling (gaseous or liquid),
- protection or not of the valve (leakage measurements),
- Oven temperature,
- Heating duration.

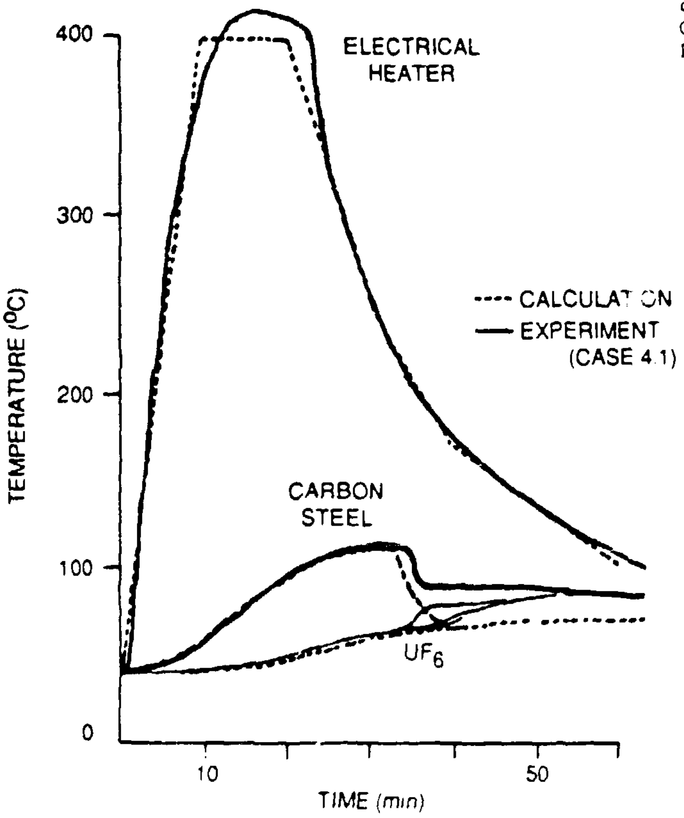

Fig. 1. Model comparison with Japanese experiment (4)

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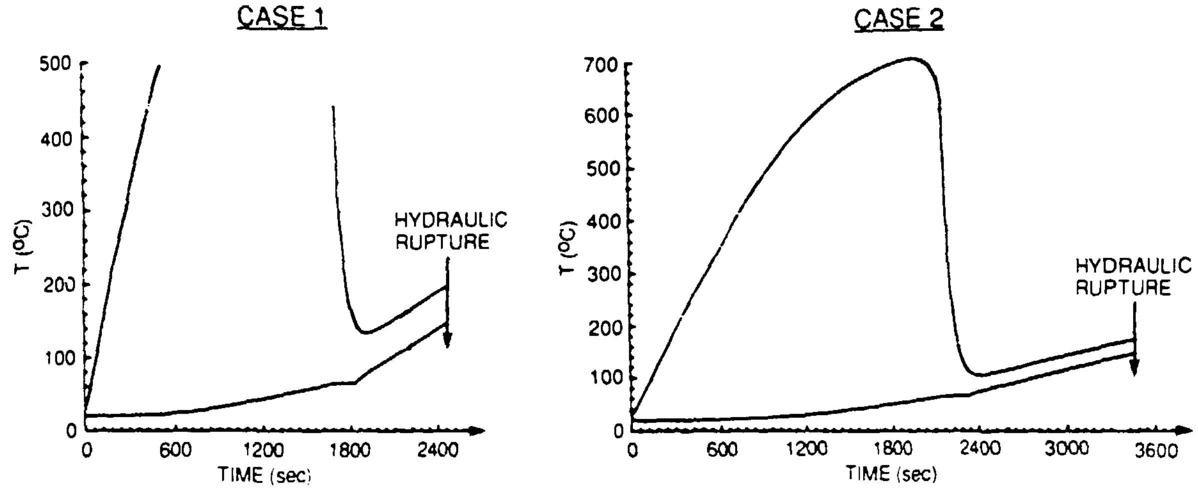

Fig. 2. Extrapolation to real fire on 48Y container

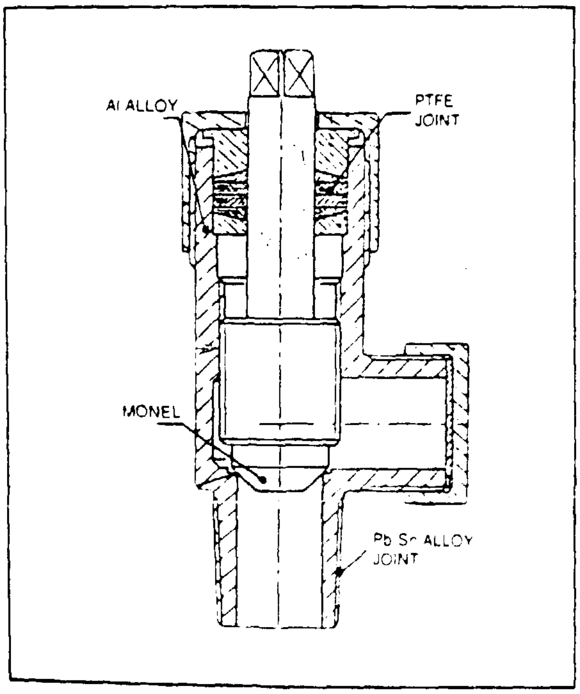

Fig. 3. 48Y valve schematic

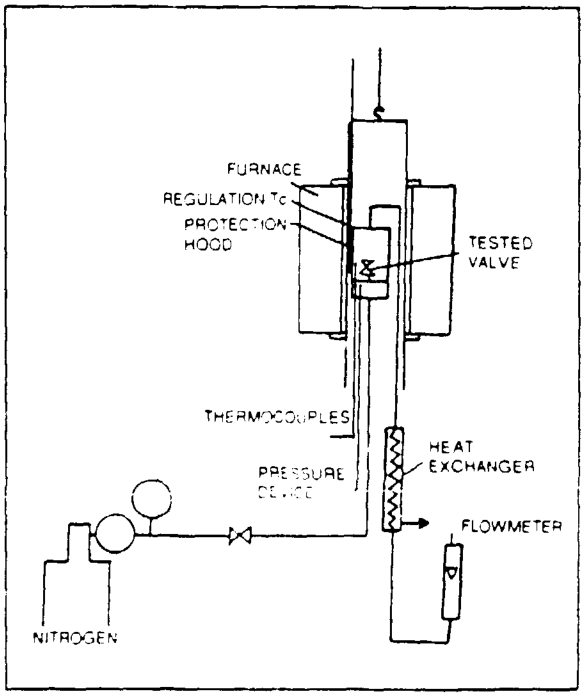

Fig. 4. Valve test loop

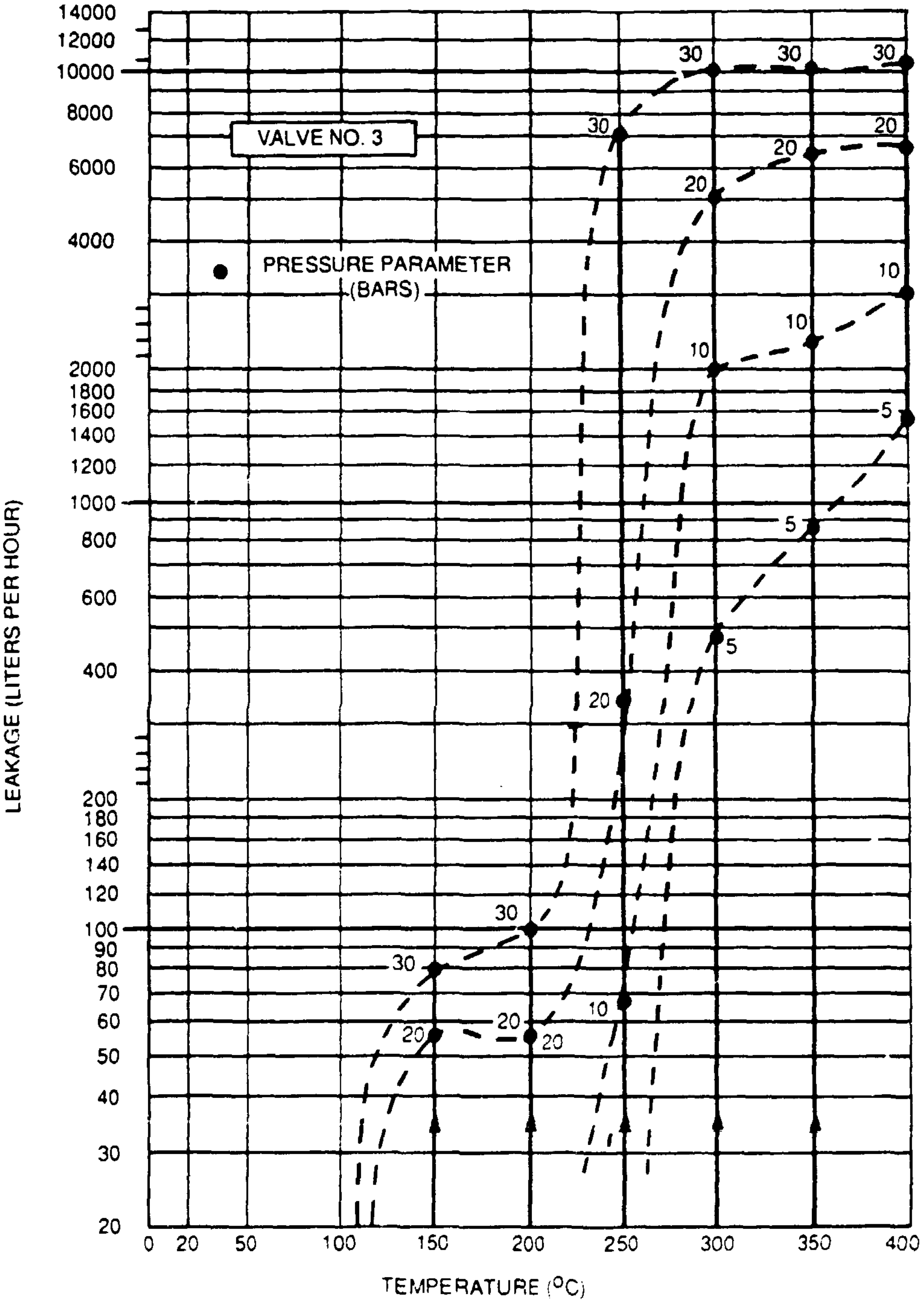

Fig. 5. Experiment measurements

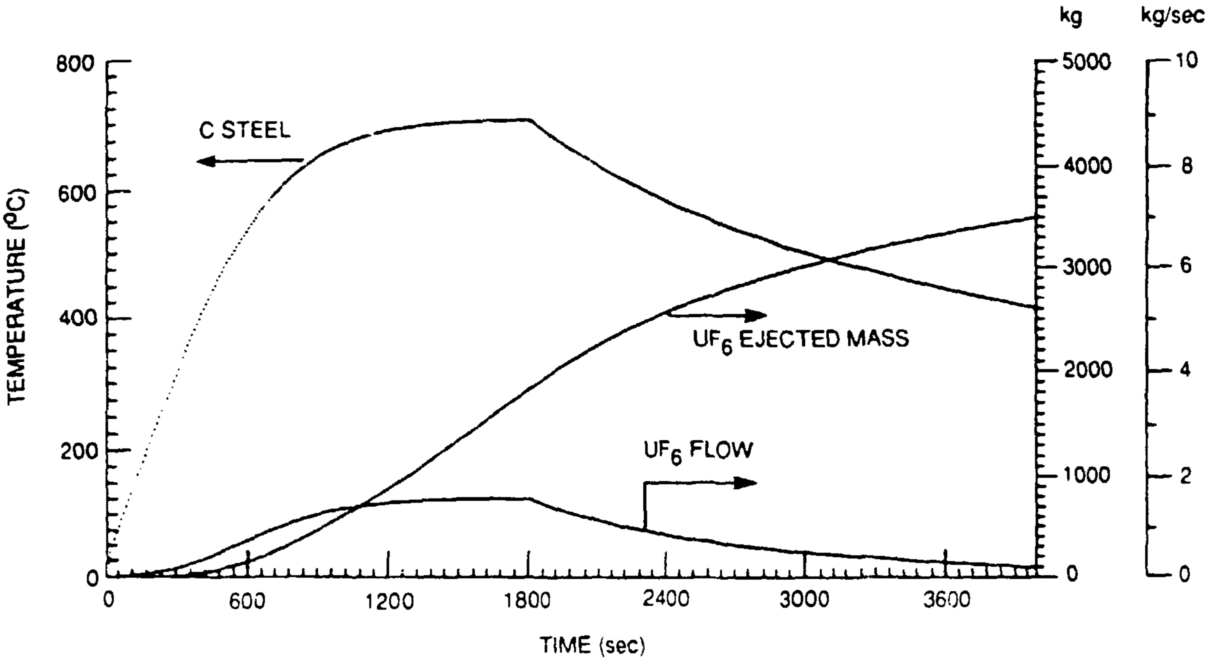

Fig. 6. Case 1 (fire duration = 30 min)

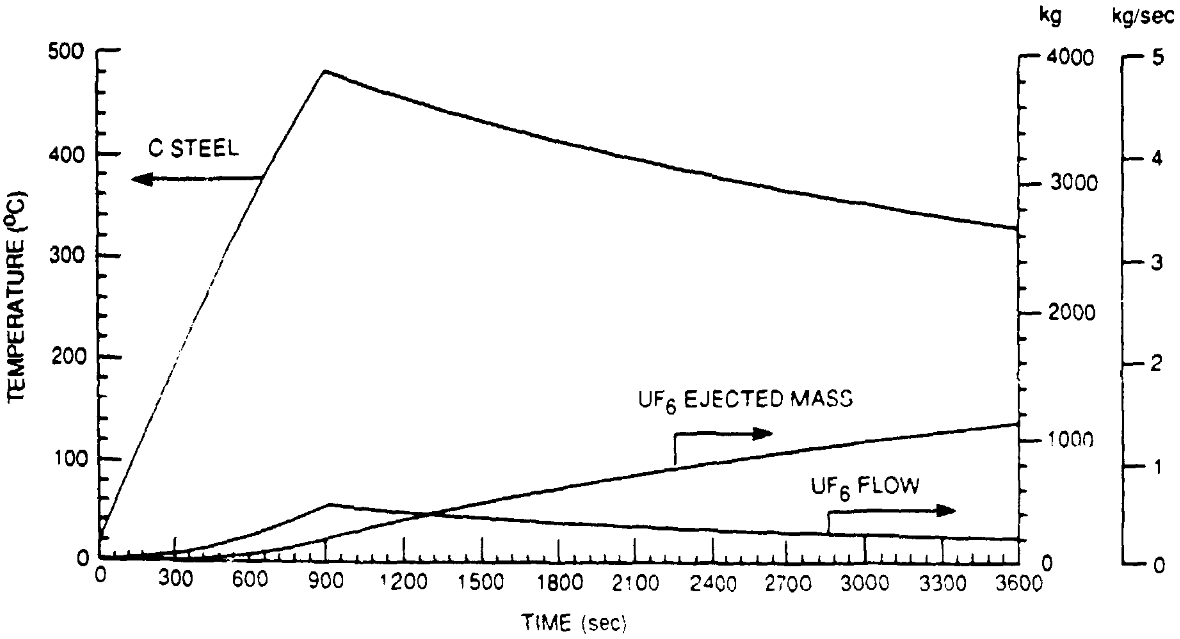

Fig. 7. Case 2 (fire duration = 15 min)

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