IMPROVEMENT OF THE IRIS PROCESS FOR INCINERATION OF VARIOUS RADIOACTIVE WASTE COMPOSITIONS

F. Lemort, J.P. Charvillat
Commissariat à l'Énergie Atomique (CEA), Rhône Valley Research Center
CEA/DEN/DIEC/SCDV, BP 171, 30207 Bagnols-sur-Cèze, France

ABSTRACT

Incineration represents a promising weight and volume reduction technique for alpha-contaminated organic waste. Following several years of laboratory research initiated in 1983 on a nonradioactive prototype unit at the CEA's Rhone Valley (Marcoule) Research Center, an innovative process, IRIS, has been developed to meet the need for processing nuclear glove box waste containing large amounts of chlorine. In March 1999, the first highly chlorinated alpha-contaminated waste was incinerated in the industrial facility based on the IRIS process at the CEA's Valduc Center. The nonradioactive prototype at Marcoule and the radioactive facility at Valduc demonstrated that the process is highly effective with a continuously fed rotating tubular kiln and with a very effective control of corrosion by pyrolytic decomposition of the waste initially at 550°C. The ash quality meets specification requirements (< 1% carbon, < 1% chlorine) and the volume and weight reduction factors are sufficient (around 30). The off-gas treatment system exhibits very high operating efficiency complying with gaseous emission standards.

On the basis of the initial radioactive operating experience, the IRIS process is being considered for use at the other operating sites. In addition to the future CEDRA incinerator at the CEA's Cadarache Center, two other facilities are planned: an incinerator on the Rokkacho Mura site in Japan for treatment of the waste arising in the nuclear facilities (RRP, J-MOX, d1) and another one for the ONDRAF in Belgium. An analysis of the requirements of these applications demonstrates that the process must be improved. The waste to be incinerated is very different from the waste materials in the Valduc facilities (high cellulose content in the Belgian waste, high polyethylene content in the Japanese waste). Experiments carried out in the nonradioactive prototype at Marcoule showed that it was very difficult to burn waste with a very wide composition range of because of clogging and overheating phenomena, especially in the pyrolysis kiln. For example, a large quantity of polyethylene causes waste to melt in the head-end area of the kiln, raising difficulties downstream. Research and development work was performed to improve the process and enhance its flexibility. The results clearly show that modifying the waste feed mechanism (the waste was continuously supplied to the process via a screw) and some parameter values (including the pyrolysis temperature in each area of the kiln) considerably extends the range of waste compositions that can be incinerated by this process. The left side of **Figure 1** shows the standard composition treated in the Valduc incinerator. Each summit of the octagon corresponds to a pure compound. PVCp is pink PVC containing a large amount of phosphorus. EVA is ethyl vinyl acetate which becomes liquid when heated. The improved process now enables incineration of all the compositions located within the dark region of the right octagon (each composition is bounded by a line for which the sum of the individual percentages for each compound is 100).

The studies carried out to date allow processing of the Belgian and the Japanese waste. The experiments in progress at Marcoule show that it could be possible to further extend the composition polygon without modifying the process technology. Solvents (including TBP or dodecane), polypropylene filters, or Plexiglas could also be incinerated in this way.

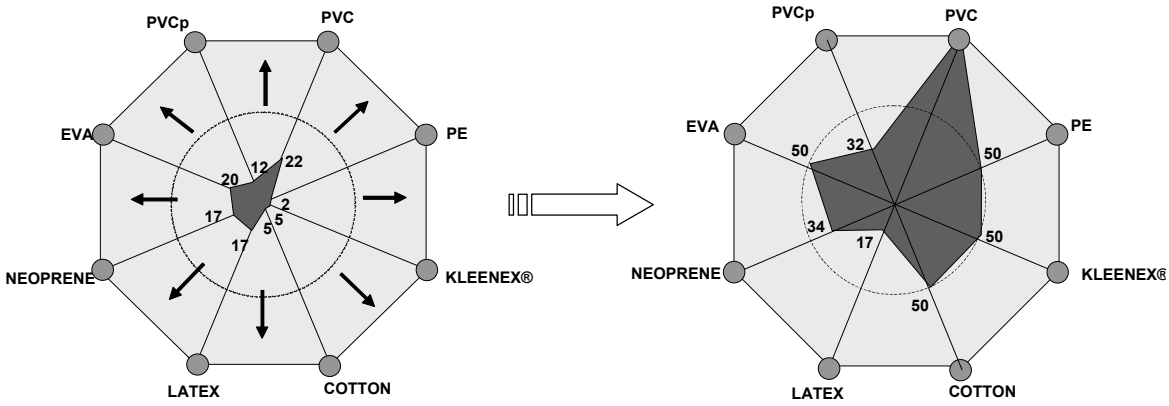

Figure 1. Evolution of the waste composition that can be incinerated by the IRIS process

INTRODUCTION

Incineration is an effective technique for reducing the weight and volume of radioactive organic waste produced during the operation of nuclear facilities. The IRIS process was developed for this purpose to process glove box waste from the CEA's Valduc Center and the MELOX (MOX fuel fabrication) plant. After several years of research and development at Marcoule the process was implemented in a unit commissioned in March 1999 at Valduc. Since then the process has produced very high quality ashes with a very satisfactory operating record. Although the waste contains about 50% chlorine, corrosion has been limited in all the process equipment.

Experience has shown that the IRIS process provides very good performance for incinerating waste of a well-defined composition. The reference waste composition at the Valduc site is indicated in **Table I**.

Table I. Average composition of "A0" waste incinerated at Valduc

Constituent	Content (wt%)
Cotton	5
Cleansing tissue	5
Neoprene	17
Latex	17
EVA	20
Pink PVC	12
White PVC	24

Routine operating experience at the site has shown that it is very difficult to guarantee a constant waste composition over time. For example, the quantity of neoprene in the waste may increase significantly following the replacement of a large number of glove boxes. This results in severe IRIS process control malfunctions; a new operating mode was therefore specified in terms of the upstream activity generating variable waste concentrations. This article describes the new process control procedures and the results obtained.

THE IRIS PROCESS

As noted above, the IRIS process was initially designed to process waste with the composition indicated in **Table I**. The relatively high inorganic content of the waste and the high chlorine concentration arising from a large PVC fraction led us to separate the process into two steps: an initial pyrolysis phase to prevent corrosion by eliminating the chlorine at moderate temperatures, followed by high-temperature calcining of the remaining carbonaceous residue. The two-step process also limits the gas flow and thus minimizes the entrainment of fly-ash. Numerous incineration tests with these wasteforms allowed us to define a multi-step process and specify the operating parameters. **Figure 2** shows the process sequence and indicates the corresponding temperatures.

The process and its main developments have already been detailed in several publications [1][2][3] together with a description of each step.

Operation experience with the process under inactive conditions at Marcoule or under active conditions at Valduc (during which over 4 metric tons of waste have been processed) demonstrates very satisfactory behavior for A0 waste treatment. The initial constituent elements are distributed among the fly-ash and bottom ash, comprising mainly (but not exclusively) Si, Ca, Al and Zn as oxides, chlorides or phosphates. The quantity and quality of the residues obtained vary according to the waste feed variations.

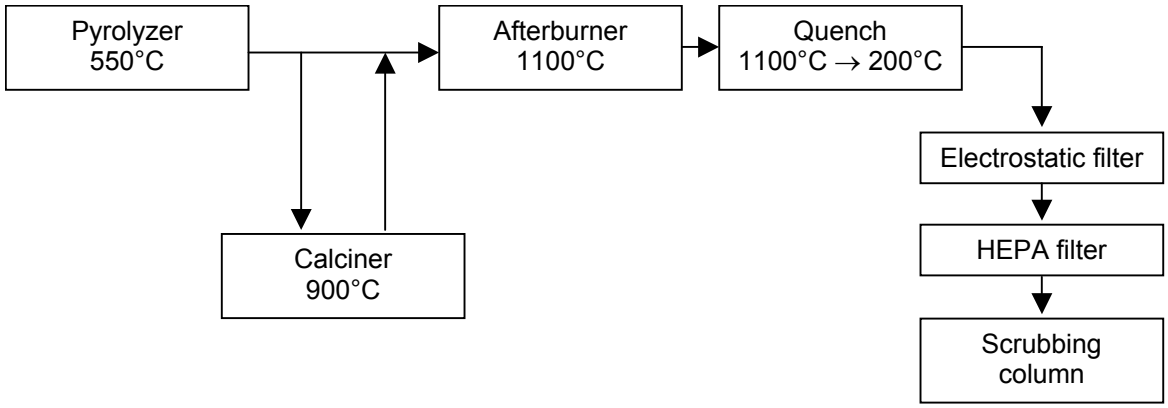

Figure 2. Schematic block diagram of the IRIS process

INTEGRATING A VARIABLE WASTE LOAD IN THE PROCESS

Observations

Experimental results obtained with incineration batches differing from the initial A0 composition show that the main problems arise during the first pyrolysis step: for example, major and very rapid temperature rises or serious plugging. **Figure 3** shows the result obtained after incineration of an extreme test batch containing 100% pink PVC (treated with a fire retardant, Reofos®).

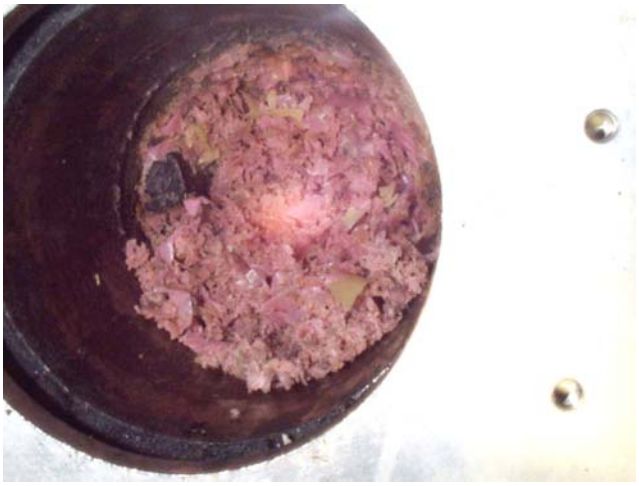

Figure 3. Inside view of pyrolyzer after incineration of a 100% pink PVC waste batch at 550°C

The figure shows the tube completely plugged after a few minutes by waste that was only slightly degraded. Similar results were obtained for pure polyethylene and EVA.

CEA tests as well as some published results [4] suggest that the waste materials exhibit independent behavior during pyrolysis. Extreme compositions containing very large quantities of PE, pink PVC or EVA thus raise processing problems.

The temperature rise observed in the pyrolyzer with mixtures highly enriched in a particular compound requires immediate action. **Figure 4** shows the temperature variation over time inside the pyrolyzer for a mixture containing 55% cellulose.

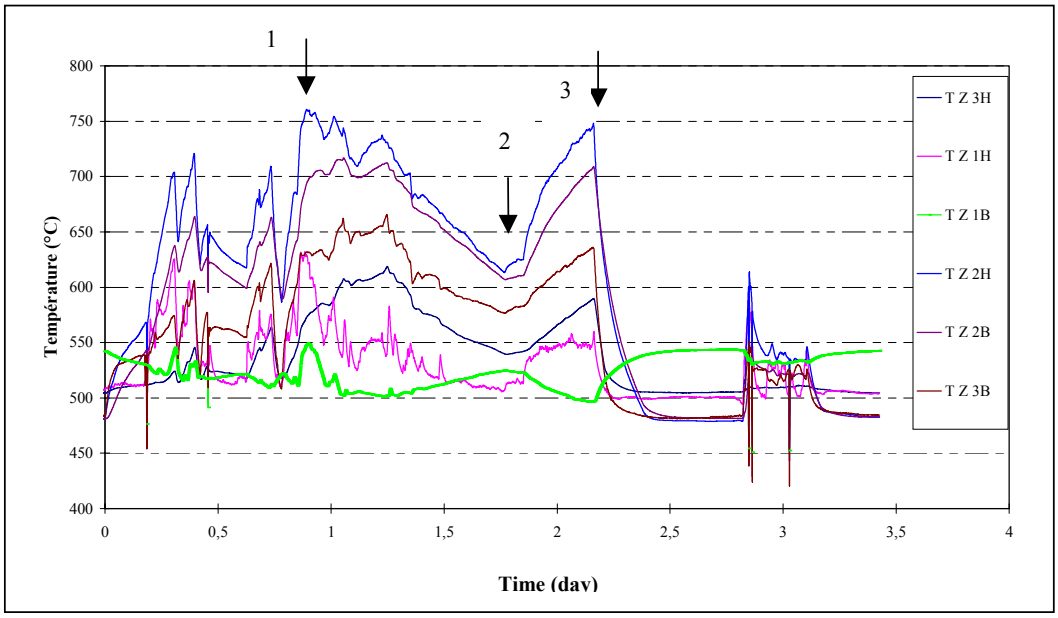

Figure 4. Pyrolyzer temperature versus time during incineration of waste highly enriched in cellulose (TZi corresponds to the temperature plot in zone I, subscripts H and B refer to the top and bottom of the zone respectively)

Figure 4 shows very strong temperature instability in the pyrolyzer (although the temperature remained relatively stable in the other portions of the facility). Very high values were reached in some cases.

Arrow 1 indicates the moment at which the air supply was reduced by half, arrow 2 shows when the normal air supply was restored, and arrow 3 corresponds to the interruption of the waste feed stream. It is important to note that the temperature rise can be very rapid and difficult to control. This may have serious consequences on corrosion; the process control must be very strict and cover interruptions of the feed stream, which is often at odds with industrial operating requirements.

Solutions considered

Examination of the pyrolysis furnace after processing showed that major deposits could form on the internal walls of the midsection. It is highly probable that these deposits are then consumed in the presence of air, and that the resulting heat release is responsible for the temperature rise observed in the second zone. Reducing the air flow and thus the oxygen flow rate diminishes the heat release, causing the temperature to drop.

- *Reducing the feed rate*

The desired result can be obtained by reducing the waste incineration feed rate. **Figure 5** shows temperature variations inside the pyrolysis furnace with a mixture containing 34% neoprene. Segment I on the graph corresponds to a throughput of 2 kg/h (i.e. half the IRIS design value); in segment II passage the feed rate was increased to 3 kg/h. The temperature in the first segment oscillated around 550°C, but quickly exceeded 650°C in the second segment. The pressure variation in the pyrolysis furnace is shown in **Figure 6**. The upper curve indicates the pressure drop and the lower curve indicates the suction pressure level at the pyrolyzer inlet (mm WG). Note the gradual rise in the inlet pressure together with the increasing pressure drop. In view of this trend and the major temperature rise phenomena, the feed rate was not increased to 4 kg/h. A subsequent test demonstrated that incinerating this type of waste at 4 kg/h caused major deposits to form in the tube, leading to plugging of the flow lines.

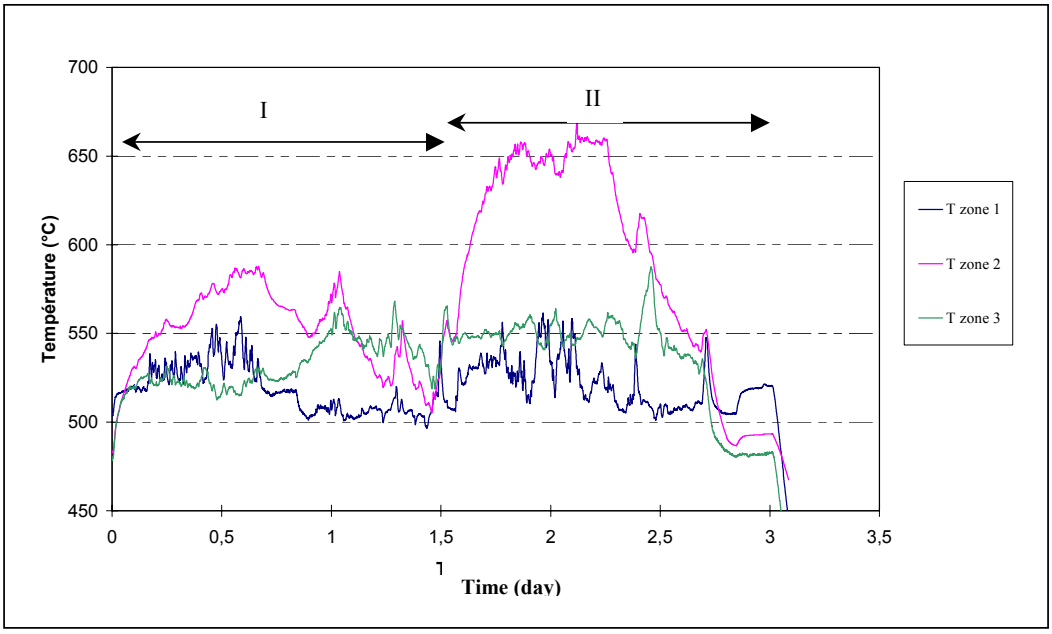

Figure 5. Temperature variations recorded in the three pyrolyzer zones during processing of waste containing 34% neoprene

A possible solution would be to diminish the feed rate for a given structure, or to increase the size of the structure for the same feed rate. The results obtained show that the temperature can be controlled in this case, but that the pyrolyzer pressure drop remains unstable.

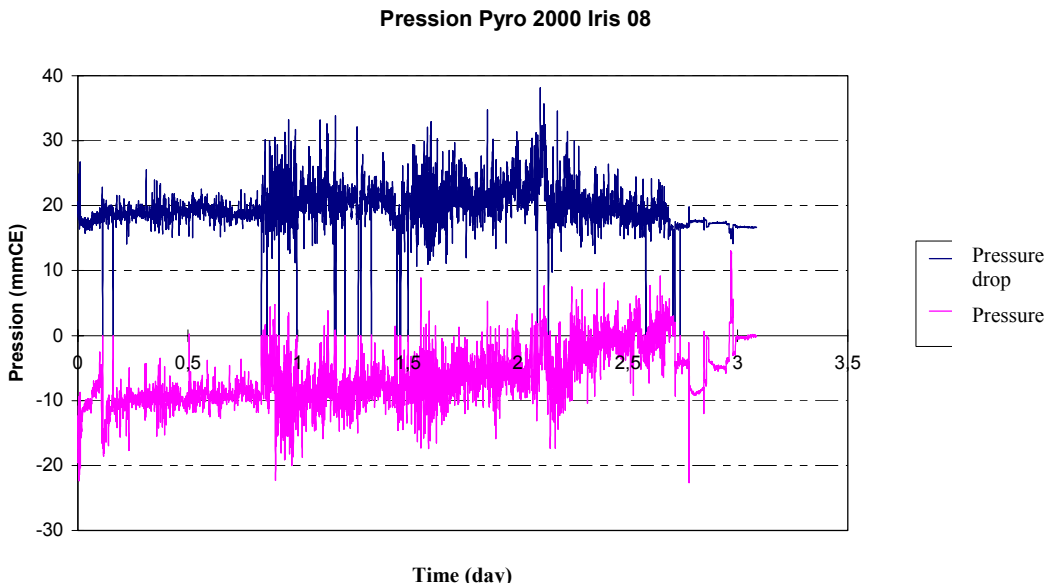

Figure 6. Pressure variations in the pyrolyzer during processing of waste containing 34% neoprene

Figure 4 shows that the temperature drops very rapidly when the waste feed ceases. It would therefore be advisable periodically to observe pyrolyzer relaxation periods to prevent overheating.

- *Feed sequencing*

In order to ensure the relaxation periods, it was suggested that the waste feed could be sequenced at a rate sufficient to ensure the nominal process throughput. The waste residence time in the pyrolysis furnace is 5 minutes; the relaxation period was therefore initially set at 6 minutes to purge the tube and eliminate any deposits using the furnace cleaning bar. The first tests of this type were carried out with waste highly enriched in cellulose; the results obtained at pyrolysis temperatures in continuous operation are shown in **Figure 4**. The waste feed cycle in this case was 20 minutes followed by a 6-minute interruption. In order to ensure a throughput of 4 kg/h, the instantaneous feed rate was increased to 5 kg/h with no technological modifications. The results obtained in this pseudo-continuous operating mode were significantly improved over those obtained in continuous operation. **Figure 7** shows the temperature variations recorded in the pyrolyzer during this test.

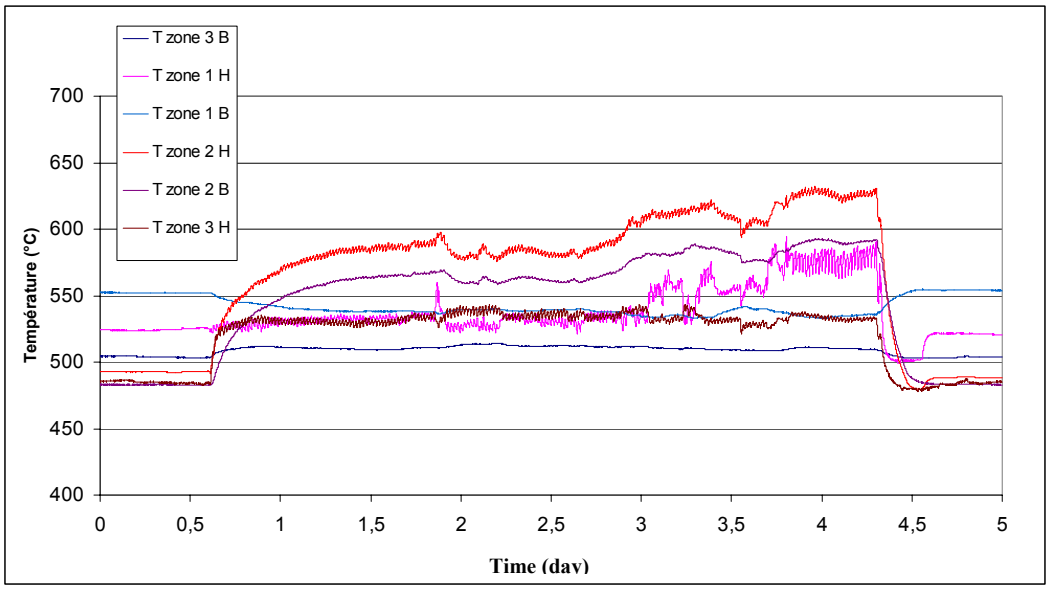

Figure 7. Temperature variations in the pyrolyzer
during sequenced incineration of waste highly enriched in cellulose

Comparing **Figure 4** and **Figure 7** shows that in the second test the temperatures were controlled, with no very rapid or uncontrollable temperature rise phenomena. In addition to this observation, the pressure drop across the pyrolyzer was very stable—this is an important safety-related consideration in the process. It may also be noted that the temperature in the first zone of the afterburner was subject to variations corresponding to the feed sequences. **Figure 8** shows the temperatures recorded in the ignition zone with a setpoint of 900°C. The temperature rise observed in this case was about thirty degrees, and had no effect on the operation of the unit.

A similar test was conducted with waste containing large amounts of neoprene (**Figure 5**). The results obtained in sequenced operation under the same conditions as above were also very conclusive. **Figure 9** shows the temperature variations within the pyrolyzer: the temperature was relatively well controlled, and stabilized at about 600°C. Compared with the results in **Figure 5**, the equilibrium temperature reached after 96 hours of incineration at a mean throughput of 4 kg/h was slightly higher than the corresponding temperature obtained in continuous operation at 2 kg/h with the same waste. The arrow in **Figure 9** indicates a shutdown due to external causes. Temperature oscillations were also observed in the afterburner. They were of higher amplitude in this case, reaching 100°C in the ignition chamber. Nevertheless, they had little effect on the refractory materials. These temperature rises could probably be limited by increasing the size of the afterburner.

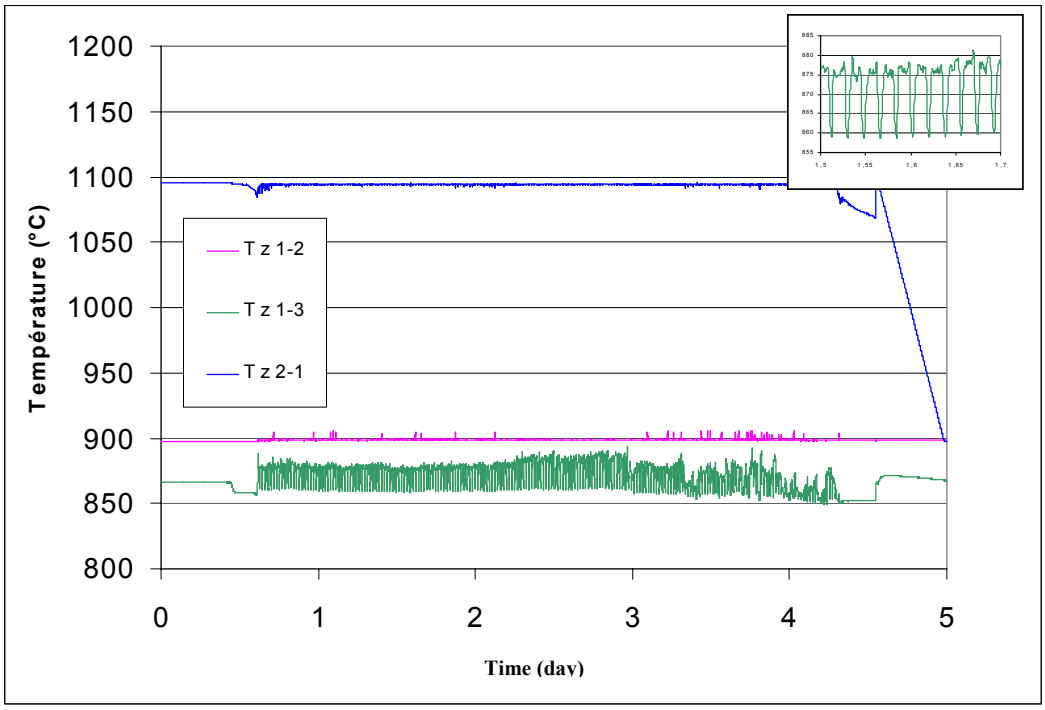

Figure 8. Temperature variations in the afterburner ignition zone during sequenced feeding of cellulose-rich waste

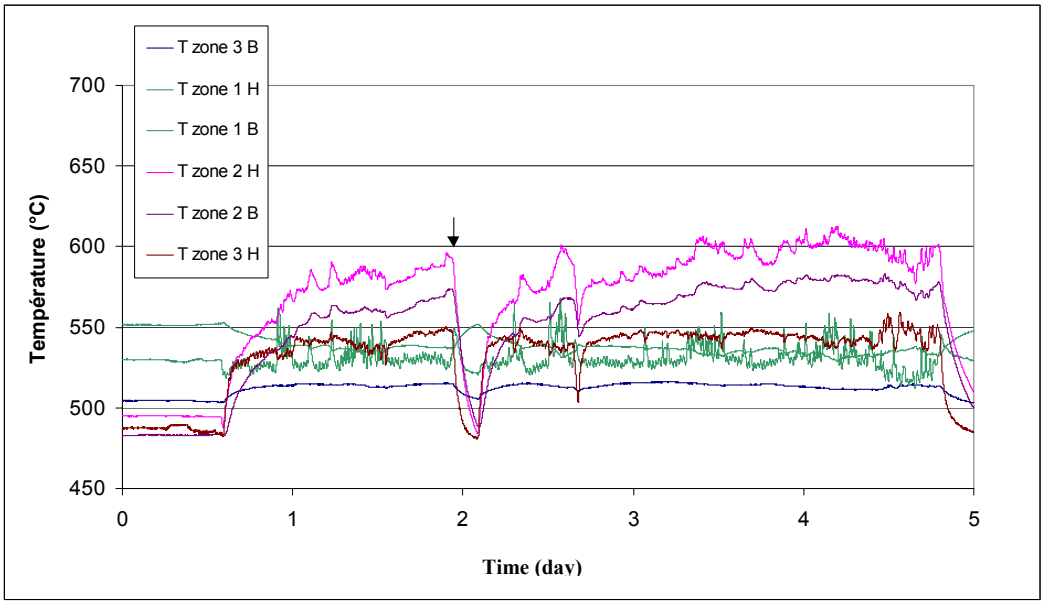

Figure 9. Temperature variations in the pyrolyzer during sequenced incineration of waste highly enriched in neoprene

CONCLUSION

Recent CEA studies were conducted to assess the possibility of extending the operating range of the IRIS process to accommodate waste with significant composition variations. This situation arises because the

waste-producing activities prior to incineration cannot guarantee the production of strictly identical waste, but also because of different operator requirements. For this purpose the behavior of each pure wasteform during pyrolysis must be known. The test results have shown that some of these materials, including polyethylene or flameproof polyvinyl chloride, cannot be processed in the IRIS facility as initially designed; most of the problems involved the pyrolysis step. The IRIS incineration tests with variable waste feed materials clearly demonstrated that some compositions are not incinerable (compositions enriched in EVA or neoprene for example) because they induce divergent phenomena detrimental to process control.

Sequencing of the waste feed stream was proposed to extend the scope of the application without modifying the technology (which is advantageous for the systems already installed). This mode allows for process relaxation and limits congestion phenomena. Very conclusive results have been obtained: regardless of the waste composition over the full test range, the process operating parameters were maintained within narrow limits. As a result, the incinerable waste composition range was significantly extended as indicated in **Figure 1**. It is important to note that the present composition range is not restrictive; further systematic studies should allow an extended composition range to be specified with additional refinement of the feed sequence.

REFERENCES

[1] A. JOUAN, J.P. MONCOUYOUX, R. BOEN, R. CARTIER, J.J. VINCENT AND T. LONGUET, "Incineration Processes for Radioactive Waste with High Alpha Contamination: New Developments", *IT3 Conference*, pp. 209-210, May 8-12, 1995, Bellevue, Washington, USA.

[2] F. LEMORT, J.P. CHARVILLAT, "Incineration of Chlorinated Organic Nuclear Waste: In situ Substitution of Phosphates for Chlorides", *ICEM 99 Conference*, September 26-30, 1999, Nagoya, Japan.

[3] F. LEMORT, T. LONGUET, J.P. CHARVILLAT, H. CHATEAUVIEUX, P. GUIBERTEAU, M. LORICH, "IRIS Alpha-Contaminated Waste Incineration Process: First Industrial Waste Processing Application at the CEA's Valduc Center", *IT3 Conference*, May 8-12, 2000, Portland, Oregon, USA.

[4] W. CHAO-HSIUNG, C. CHING-YUAN, H. JWO-LUEN, "On the thermal treatment of plastic mixtures of MSW: pyrolysis kinetics", *Waste Management*, Vol. 13, pp. 221-235, 1993.