Thermal Testing of Solid Neutron Shielding Materials*

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INTRODUCTION

General Atomics (GA) is currently developing two legal-weight truck casks for the Department of Energy (DOE), Office of Civilian Radioactive Waste Management (OCRWM). These casks, the GA-4 and GA-9, will carry four PWR and nine BWR spent fuel assemblies, respectively. Each cask has a solid neutron shielding material separating the steel body and the outer steel skin. In the thermal accident specified by NRC regulations in 10CFR Part 71 the cask is subjected to an 800°C environment for 30 minutes. The neutron shield need not perform any shielding function during or after the thermal accident, but its behavior must not compromise the ability of the cask to contain the radioactive contents.

In May-June 1989 the first series of full-scale thermal tests was performed on three shielding materials: Bisco Products NS-4-FR, and Reactor Experiments RX-201 and RX-207. The tests are described in *Thermal Testing of Solid Neutron Shielding Materials*, GA-A19897, R. H. Boonstra, General Atomics (1990), and demonstrated the acceptability of these materials in a thermal accident. Subsequent design changes to the cask rendered these materials unattractive in terms of weight or adequate service temperature margin. For the second test series a material specification was developed for a polypropylene based neutron shield with a softening point of at least 280°F. Table 1 lists the neutron shield materials tested. The Envirotech and Bisco materials are not polypropylene, but were tested as potential backup materials in the event that a satisfactory polypropylene could not be found.

TESTING SETUP AND PROCEDURE

Setup

Figure 1 shows a representative test article and thermocouple positions. Each test article consists of blocks of neutron shield contained in a box of dimensions 36 in. x 36 in. x 4.5 in. The box is 11-gage type 304 stainless steel with continuous welds along all seams and a 6-in. x 12-in. hole in the center of a 36-in. x 36-in. face. The hole simulates damage from the hypothetical drop and puncture events and extends 1.5 in. into the material. Six inches of mineral fiber insulation surround the box except for the face with the hole.

Six KNBS (Chromel-Alumel) 20-gage thermocouples (TCs) measure temperatures on the test article. The environment temperatures are determined using five TCs of the same type as on the test article, but shielded to prevent radiant heat loss to the test article surface. These TCs are positioned 6 in. away from the test article surface.

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As shown in Fig. 2, a vertical exposure furnace heats the uninsulated face of the test article. The back wall of the furnace contains a bank of seven natural gas burners. A single globe valve regulates the overall gas pressure to control the test exposure to the temperature range of 800° to 900°C.

TABLE 1
NEUTRON SHIELD MATERIALS IN THERMAL TESTS

Supplier	Material
Kobe Steel, Ltd.	PP-R01 polypropylene, 1% boron
Envirotech Molded Products, Inc.	High-density polyethylene (HDPE), 0.8% boron
Bisco Products, Inc.	Modified NS-4, 4.5% boron
Reactor Experiments, Inc.	High melt index polypropylene (HMPP), 1% boron

Procedure

The completed test article is conditioned at room temperature for at least 24 hr, then moved into position directly in front of the furnace. Recording of the TC data begins 5 minutes prior to ignition of the burners. After ignition, the average furnace temperature (i.e., average of the five furnace TCs) is maintained between 800° and 900°C for 30 minutes. The burners are then shut off and the test article is pulled away to cool in ambient air. When all temperatures have peaked, the test article is again conditioned at room temperature for at least 24 hr and then disassembled for inspection.

Material Acceptance Criterion

The neutron shield must not provide a source of thermal input to the cask sufficient to degrade containment integrity. More specifically, the material is acceptable if (1) temperatures on the back surface do not at any time exceed the maximum temperature of the thermal accident environment and (2) it shows no evidence of prolonged combustion (i.e., combustion lasting for a period of several hours) following the thermal accident.

OBSERVATIONS AND RESULTS

Two of the materials, the modified NS-4 and the HMPP, passed the test and are acceptable for use as the neutron shielding. The results for the PP-R01 and HDPE materials are inconclusive because the tests were stopped prematurely due to intense smoke combined with inadequate ventilation. Table 2 provides time-related observations for the four tests. Post-test inspection results and temperature plots are discussed below.

Kobe PP-R01

Disassembly of the test article after cooling indicated that despite the observed combustion a relatively small amount of material had been lost. The polypropylene was heavily charred and broken into loose tiles on the front surface. Some individual blocks had fused together but, contrary to expectations, there was little evidence of extensive melting. The total weight loss was 36.3 lb out of an initial 200.1 lb, or 18%.

Figure 3 gives the average environment temperature and the thermal response on the back surface of the test article, where the peak temperature is about 340°C. The responses exhibit abrupt increases within the first several minutes, a characteristic of all the tests. All temperatures on the back surface meet the acceptance criterion that they not exceed the accident environment temperature, but acceptability of the material cannot be established since the test was halted.

Envirotech High-Density Polyethylene (HDPE)

The test article was opened to reveal that most of the material remained in the box. Charring was concentrated mostly on material near the hole and in the lower region beneath the hole. Here molten material had collected and resolidified as indicated by a layer of dark material. In the upper portion, above the hole, charring was confined mainly to the surface and virgin, white material in the form of individual blocks was still identifiable. There was very little loose material. The total weight loss was 27.0 lb from an initial weight of 199.6 lb, or about 14%.

The average environment temperature and back surface TC responses are shown in Fig. 4. The responses display the sudden rises seen in the preceding test, although the peak response is less than 150°C. In this respect the HDPE performed better than the PP-R01 and its temperatures also meet the acceptance criterion. However results are indecisive due to termination of the test.

Bisco Modified NS-4

This material is similar to the NS-4-FR tested in 1989 but has a slightly lower hydrogen content and a lower weight. In this test the heating phase successfully proceeded the full 30 minutes, and the material was deemed acceptable.

A post-test inspection indicated immediately that the majority of the material had been retained. This was confirmed by a weight loss of only 6%, 14.5 lb out of 244.6 lb. There was a fairly uniform, black char layer about 1/8 to 3/16 in. thick on the front surface. With the exception of this char layer, which was fragile and separated easily from the remainder of the material, all blocks appeared nearly intact. An average of 2-1/16 in. of undamaged material remained in the front layer of blocks. Some charring was also observed along the sides of blocks that joined at the locations of TCs 1 and 2 and TCs 4 and 5. The back layer of blocks revealed localized discoloration near the top corners and a black, oily material that had condensed on the block surfaces. Apart from this, very little damage was noticed on these blocks.

Figure 5 gives the average environment and back surface temperatures The maximum back surface response is just less than 100°C. The initial peaking of TCs 4 and 6 before 50 minutes is followed by a gradual increase of these temperatures with subsequent maximums between 400 and 500 minutes. The later gradual increase is obviously due to conduction of heat through the material to the back surface; the initial peaks are therefore due to some other phenomenon.

Reactor Experiments High-Melt Index Polypropylene (HMPP)

This material also went through the full 30 minutes of heating and post-test cooldown. It was subsequently judged acceptable for use in the cask.

After complete cooling in ambient air, the total material weight loss was determined to be 109 lb from the initial 194 lb, some 56%. Disassembly of the test article confirmed a significant absence of material. No obvious char layer was noted. The space above the bottom edge of the hole was empty except for a column of partially melted blocks along the left side of the box. In the lower section, beneath the bottom edge of the hole, the material had softened and fused into a solid mass, although gaps between individual blocks could still be seen. The thickness of the fused material at the center of its top edge was approximately 5 in., indicating that the front steel had bowed outward 0.5 in. and allowed additional material to flow down from the top portion. Melted and resolidified material could be seen as distinct layers on the exterior surfaces of the fused, bottom portion.

Temperatures of the average environment and on the back side of the test article are shown in Fig. 6. The maximum back face temperature, 211°C, occurs in TC 6 at about 6.5 minutes. The sudden increase in back face temperatures at the beginning of the test is again noted, particularly in TCs 4 and 6, which were located in the bottom portion of the test article.

TABLE 2
THERMAL TEST OBSERVATIONS

PP-R01 polypropylene, 1% boron	High-density polyethylene (HDPE), 0.8% boron	Modified NS-4, 4.5% boron	High melt index Polypropylene (HMPP), 1% boron
3 min — Temp. = 800°C, slight smoking and flame from exhaust stack. **5.5 min** — Temp. > 900°C, smoke and flames fill furnace interior. Furnace burners turned down, then shut off. **6.5 min** — Temp. > 1000°C, extremely heavy smoke. **18.5 min** — Temp. = 1100°C, decision made to terminate test. **19 min** — Two failed attempts to extinguish flames with CO_2. Test article moved from furnace and flames extinguished with water.	**2 min** — Temp. = 800°C **2.5 min** — material ignited **4.5 min** — Temp. > 900°C, material flowing out of box front with vigorous flaming and smoking from exhaust port. Furnace burners turned down, then shut off. **5 min** — Temp. ≈ 1000°C, exhaust port damping reduced temp. to within test range. **17 min** — Test terminated due to copious smoke production, flames visible along bottom of test article. Test article moved from furnace. Flames observed inside furnace indicating material had been discharged into furnace. Flames extinguished with water.	Additional ventilating capability supplied. Environment achieved desired test range and maintained there for 30 min without difficulty. Some smoke was observed and some flames could be seen from the exhaust port, but intensity of combustion was less than in previous tests. **30 min** — Test article moved from furnace. Flames approximately 4 ft high issue from hole in test article. After about 5 min, the flames diminish and self extinguish. White smoke persists another 3.5 min.	**3 min** — smoke begins issuing from exhaust port. **5 - 6 min** — 3-ft column of flame appears, burners adjusted to control temp to specified range. **17 min** — Furnace burners shut off. Internal combustion maintains temp. in test range. Molten material observed flowing into water-filled catch pan. **30 min** — Test article moved from furnace. Flames issuing from hole and from material that had flowed out of the hole and become trapped in the insulation. Flames from hole gradually diminish and self extinguish after 12 min. Flames from material in insulation self extinguish after another 38 min.

CONCLUSIONS

The Bisco modified NS-4 and Reactor Experiments HMPP are both acceptable materials from a thermal accident standpoint for use in the shipping cask. Tests of the Kobe PP-R01 and Envirotech HDPE were stopped for safety reasons, due to inability to deal with the heavy smoke, before completion of the 30-minute heating phase. However these materials may prove satisfactory if they could undergo the complete heating.

Table 3 compares key results for all four materials. The Bisco modified NS-4 is best in terms of survivability and back side temperatures. Despite the more intense combustion of the HMPP during the 30-minute heating phase, the test article did not sustain this combustion when moved away from the furnace. Had the molten material not become entrapped on the test article insulation (which is not a part of the actual cask), the combustion would have expired much earlier. The back side maximum temperature of 211°C is well within the criterion of 800°C. This material therefore is also acceptable. It is preferable to the Bisco modified NS-4 since it is about 20% lighter and at the same time has a hydrogen density 20% higher.

The tests of the PP-R01 and HDPE materials were terminated for safety due to the extremely heavy smoke in the indoor test facility. The combustion of the materials without any external heat input led to the initial belief that they were unacceptable. However the HMPP test showed the same phenomenon, and combustion in the test article eventually ceased when it was moved from the furnace. This fact led to the conclusion that placing the test article against the well-insulated furnace formed an effective heat-retaining environment that kept the temperature high enough to support combustion. Neutron shield material became sufficient fuel in place of the gas burners. Moving the test article to cool in ambient air disrupted this process and the combustion ended. It is thus plausible that the PP-R01 and HDPE would behave like the HMPP and prove acceptable if these tests could proceed the full duration. Note that backside temperatures (see Figs. 3(b) and 4(b)) remained well below the acceptance criterion of 800°C.

TABLE 3
COMPARISON OF NEUTRON SHIELD THERMAL PERFORMANCE

Material	Time to End of Flames (min.)	Weight Loss (%)	Peak back side temps (°C)			
			TC 1	TC 3	TC 4	TC 6
PP-R01	Test terminated	18	341	169	328	336
HDPE	Test terminated	14	83	140	146	78
NS-4	5	6	96	41	47	42
HMPP	11.5 [(1)], 50 [(2)]	56	127	203	152	211

(1) Material inside test article
(2) Material outside test article entrapped on insulation

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REFERENCES

Boonstra, R.H., *Thermal Testing of Solid Neutron Shielding Materials*, GA-A19897, General Atomics (1990).

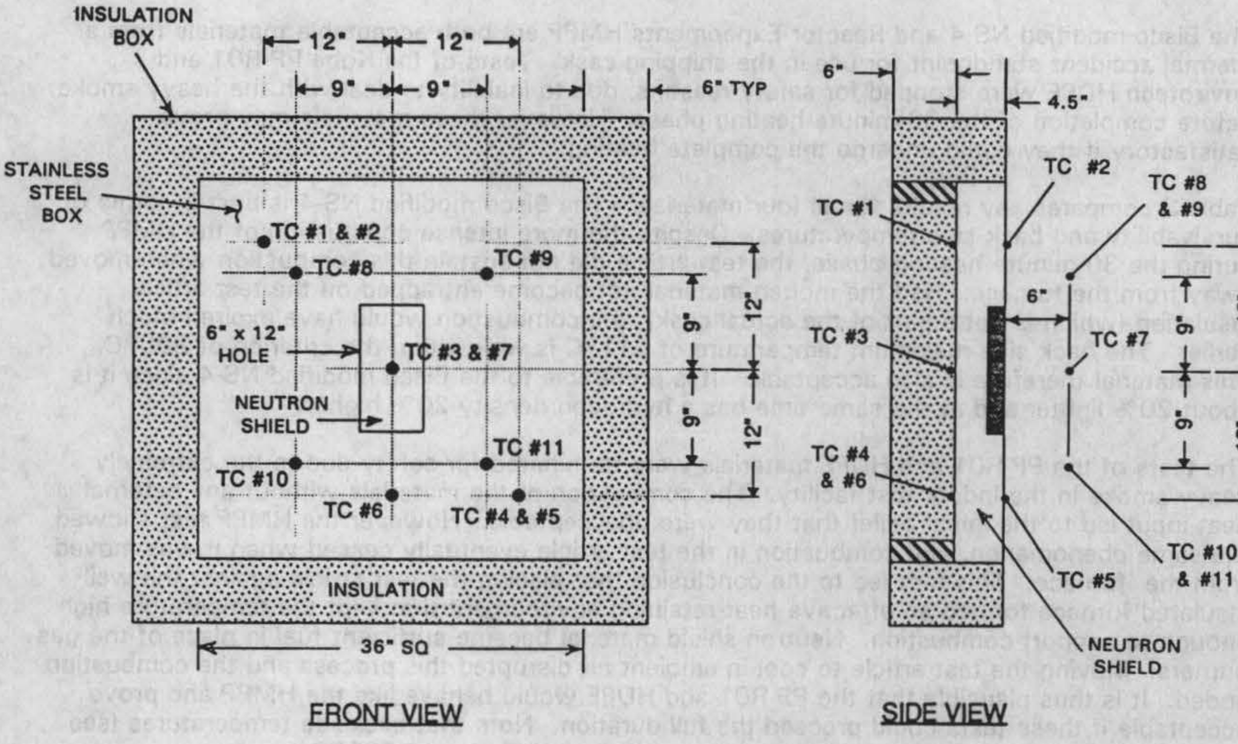

Fig. 1 Neutron shield test article with thermocouple locations

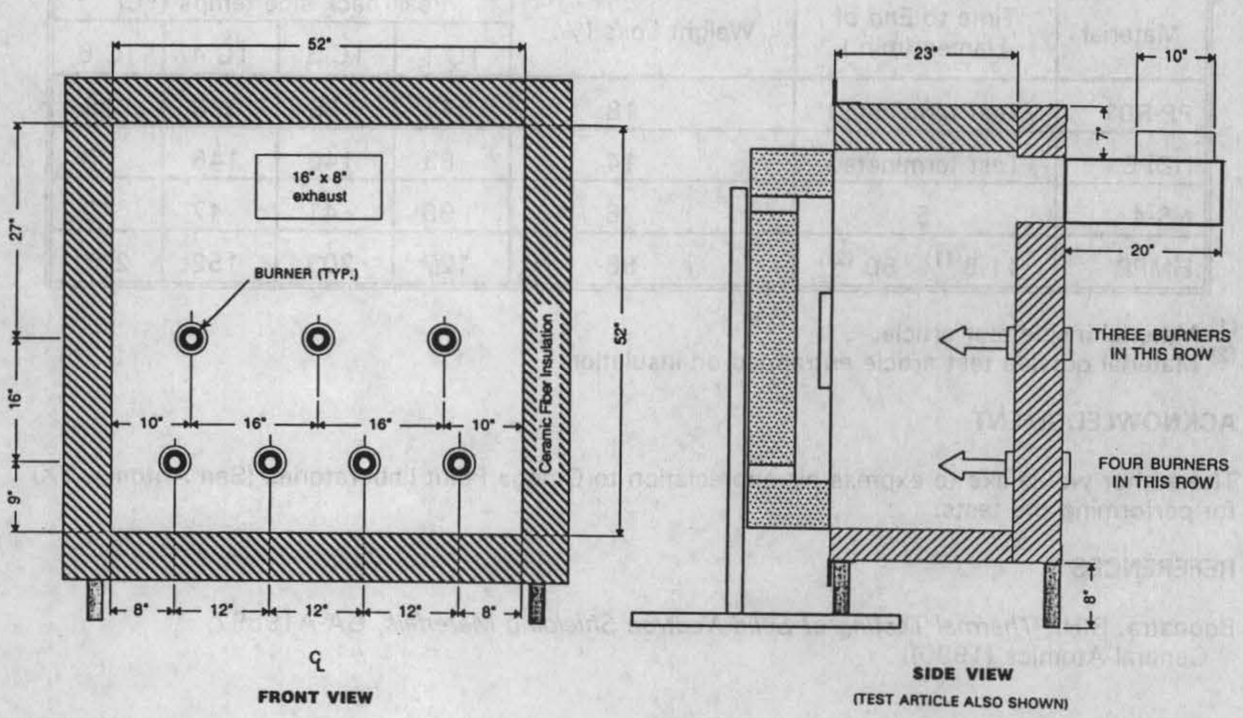

Fig. 2 Neutron shield test furnace

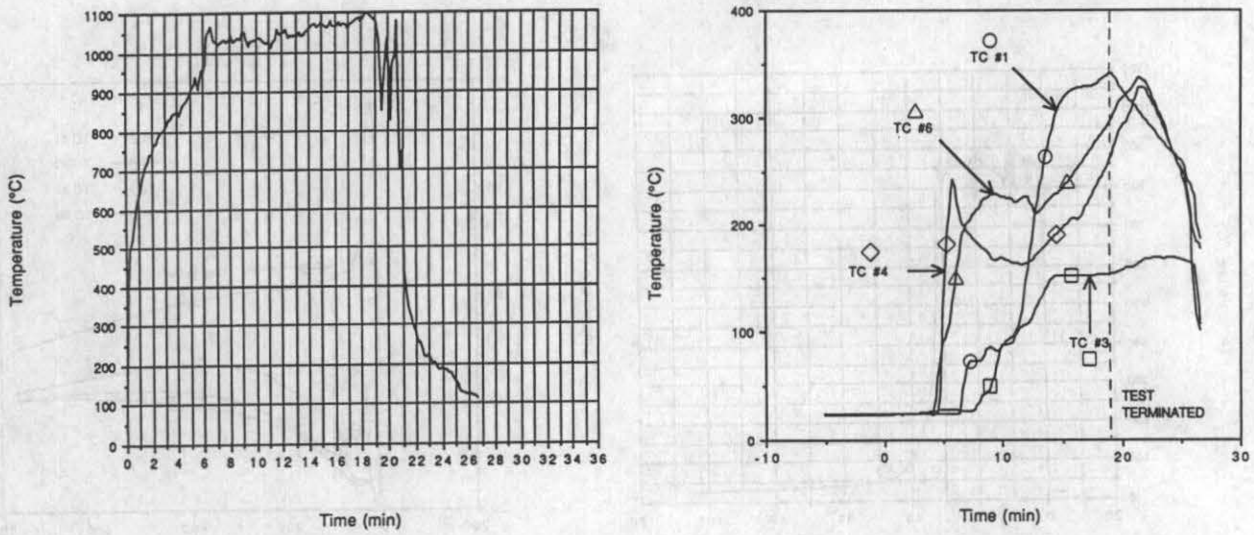

Fig. 3 Kobe PP-R01 test temperatures: (a) average environment, (b) back side response.

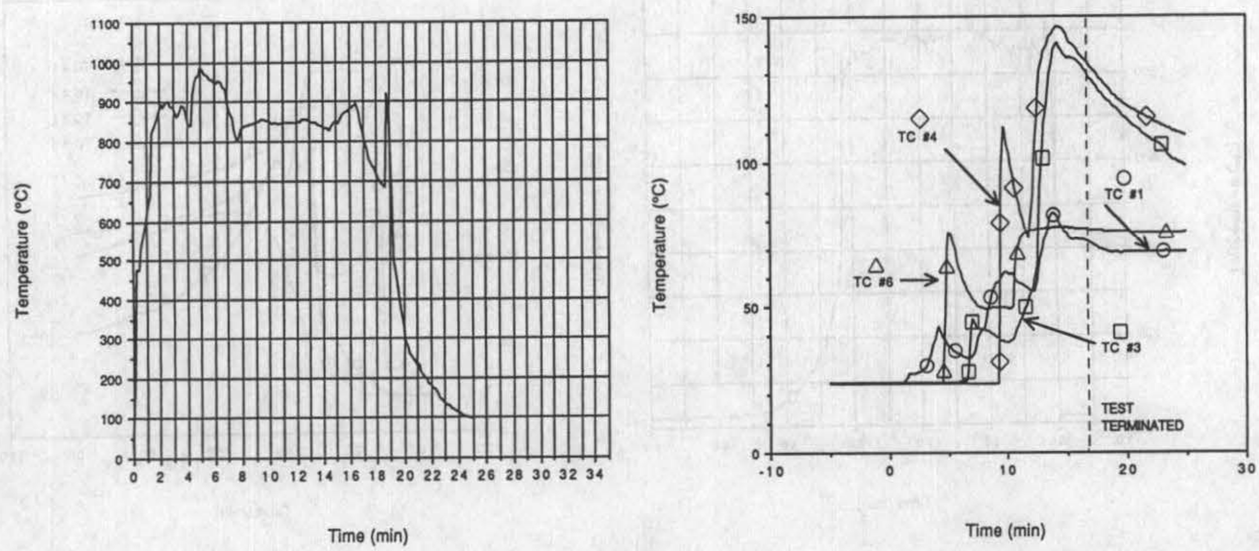

Fig. 4 Envirotech HDPE test temperatures: (a) average environment, (b) back side response.

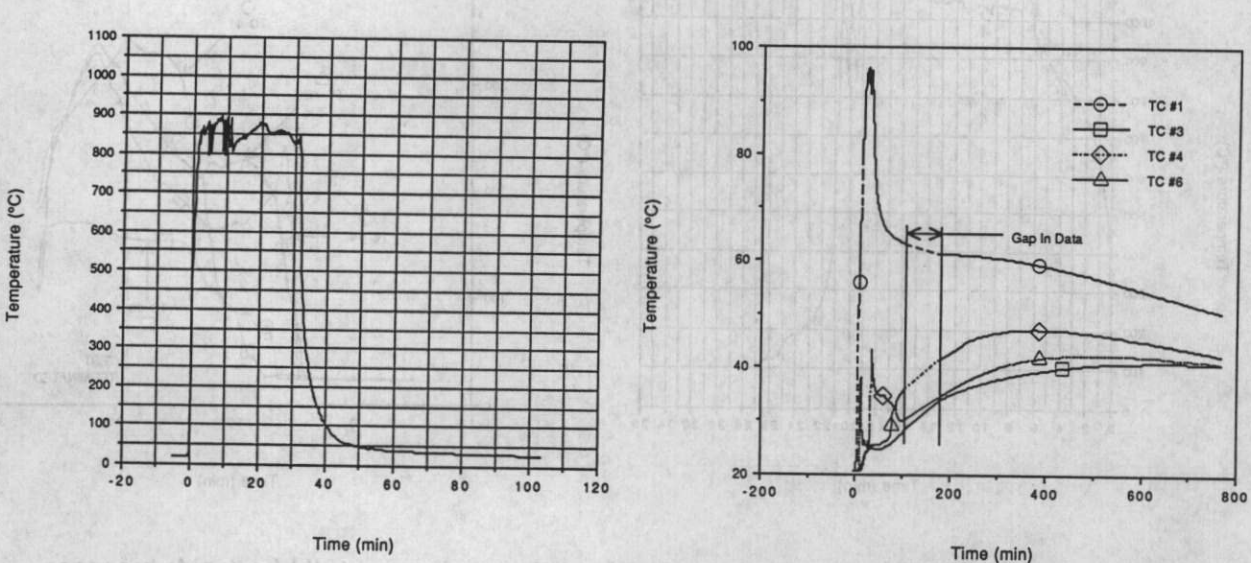

Fig. 5 Bisco Modified NS-4 test temperatures: (a) average environment, (b) back side response.

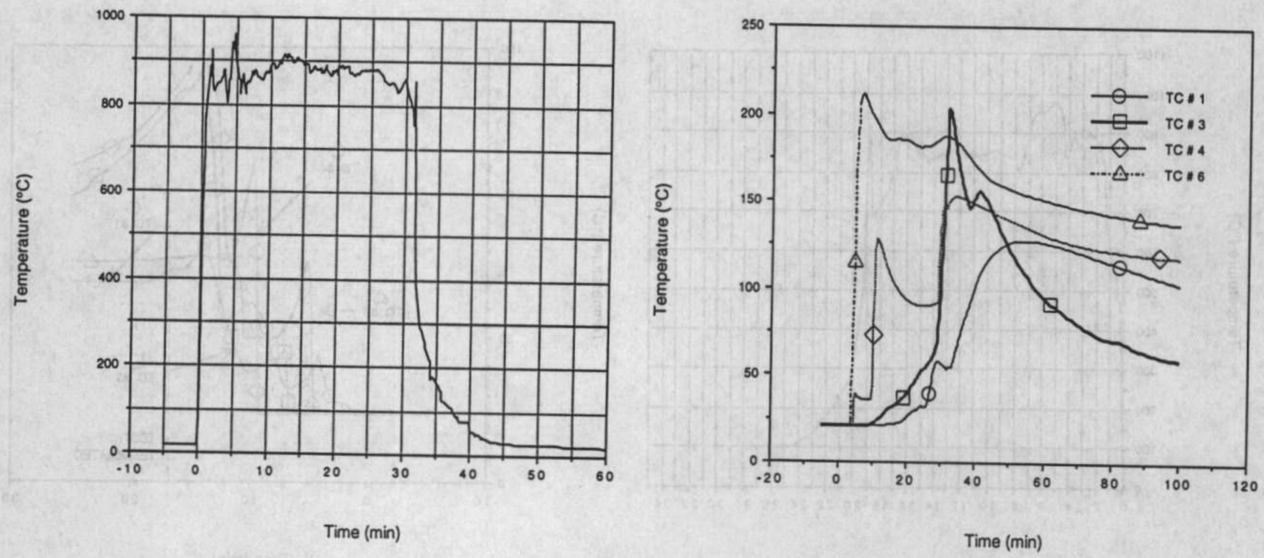

Fig. 6 Reactor Experiments HMPP test temperatures: (a) average environment, (b) back side response.