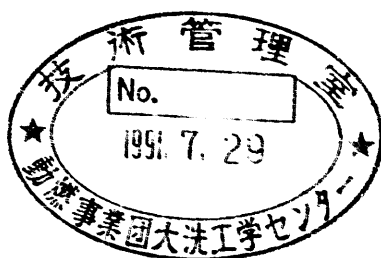


DEVELOPMENT OF CERAMIC LINER FOR FBR BUILDING



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ABSTRACT

To develop a ceramic liner, a selection test of materials, an improvement test of selected material, and a feasibility test of the liner have been conducted.

In the selection test, fifty commercially available high temperature cement and ceramics were subjected to thermal shock test (TST), sodium exposure test (SET), and sodium flame exposure test (SFET). From test results, alumina/silicon-carbide (Al_2O_3 -SiC) mixture base castable refractory was selected in consideration of material cost, and material availability for a simpler liner construction in the buildings.

The selected material was subjected to the improvement test. From the test, proper weight fractions of additives such as alumina cement and silica were determined. Drying conditions were also determined.

Finally, a sodium burning pan made of concrete whose inner surfaces were covered with the improved Al_2O_3 -SiC base castable refractory was fabricated and was used for a sodium burning test.

INTRODUCTION

In the FBR buildings, the concrete surfaces are covered with steel liners to contain a spilled sodium and to prevent sodium-concrete reaction following the postulated sodium spill accidents. Although the steel liners are highly reliable under the accident conditions, they need a complicated design and a long period for the construction. In a typical FBR plant, the liner construction becomes one of the critical paths for the plant construction. One of the possible way to eliminate the disadvantages associated with the liner construction is to adopt ceramic, instead of steel, for its material. In general, ceramic has smaller thermal expansion coefficient than steel and excellent high temperature resistance. Features of the ceramic liner, therefore, would be a simpler design that leads to a shorter construction period. Since no paper has been published so far in regard to the ceramic liner, the present authors have carried out its development.

SELECTION OF MATERIALS

Materials nominated for the selection tests were about 50 commercially available ceramics and high temperature cement. Their test pieces of 40mm in height, 30mm in width, and 20mm in depth were made and were used in the thermal shock test (TST), the sodium exposure test (SET), and the sodium flame exposure test (SFET).

Procedures of the tests are as follows. In TST, the test piece at room temperature was dropped into a sodium pool at 650°C to give a large thermal shock. After several minutes, the test piece was pulled out into air and was cleaned for examinations. In SET, the test piece was immersed into a sodium pool at 150°C. It was heated up to 650°C together with sodium and was kept at this temperature for 1 hour. Following to this, the test piece in the sodium was cooled down to 150°C during the next 3 hours. After extraction of the test piece into air, it was cleaned for examinations. In SFET, the test piece was exposed to a sodium flame for 1 hour. During the exposure, temperature of the flame exceeded 850°C. The temperature conditions for TST and SET described were based on an experience obtained from a series of sodium spill accident simulation tests in PNC⁽¹⁾, where the maximum spilled sodium temperature reached 650°C. The duration described in SET and SFET, however, are shorter than those postulated in the sodium spill accidents.

Results of the tests were as follows. The high temperature cement and some ceramics were destroyed shortly after their immersion into sodium or their exposure to the flame in SET and in SFET, respectively. Table I shows results for ceramics that survived and kept their original shape during the tests. It was found from TST that, in general, cracks were found in dense ceramics, while crack was hardly found in porous ceramics. The reason is probably attributed to the fact that dense ceramics have higher thermal conductivities than porous ones, and this leads to higher thermal stresses generated in TST as to generate cracks. In contrast, even if a very small initial crack is generated in porous ceramics, its propagation would be arrested by pores. Another findings from the tests are, as shown in the table, alumina (Al_2O_3) base material, magnesia (MgO) base one, silicon-carbide (SiC) base one, and silicon-nitride (Si_3N_4) base one are stable and were not corroded nor destroyed by a hot sodium and a sodium flame. Among these materials, silicon-nitride base was the best, although it is very expensive.

Essentials one should consider to select materials for the liner are material cost and their availability to a simpler liner construction in the plants. From the first view point, silicon-nitride (Si_3N_4) base material is the most expensive as explained, and magnesia (MgO) base one is more expensive than the others. Therefore, these two materials and materials that contain these two were eliminated from a list of materials for the liner. From the second view point, castable refractories are better than sintered ceramics, because any shape of the liner can be made easily. From these considerations and the results shown in Table I, SiC and Al_2O_3 remained. The results of the tests showed that $\text{Al}_2\text{O}_3/\text{SiC}$ base material had better properties, and therefore its castable refractory was selected finally.

IMPROVEMENT OF THE SELECTED MATERIAL

Physical and chemical properties of ceramic are generally very sensitive to its additives. Therefore, improvement of the selected material, i.e., $\text{Al}_2\text{O}_3/\text{SiC}$ base castable refractory, was conducted by changing its additives. Representatives of additives are alumina cement and silica that are essential to keep mechanical strength.

For alumina cement, its proper weight fraction was determined by conducting the similar tests to TST, SET, and SFET as presented. It was shown from results that the higher the weight fraction was, the higher the mechanical strength and the thermal shock resistance became, as expected.

In the similar way, proper weight fraction of silica was determined. As is well known, silica (SiO_2) reacts with sodium at higher temperature than about 530°C^(2,3). Once the reaction occurs in the porous refractory, the material will be destroyed. Figure 1 shows a relationship between penetration depth of sodium into the refractory and the weight fraction. It is seen that, with the fraction lower than a few percent, the sodium penetration into the refractory is very small or almost zero. Post-test destructive examination of the test piece

under optical microscope and X-ray diffraction analysis indicated that a thin silica and sodium reaction product layer was formed at a surface of the test piece. This layer became a barrier against further penetration of sodium into the porous refractory. With the higher fraction than a few percent, the penetration depth becomes larger. With extremely high weight fraction of silica, the test pieces were destroyed due to the reaction everywhere in the material. From these results, proper weight fraction of silica was determined to be a few to several percent. In this range, compressive strength of the refractory is more than 600 kg/cm^2 that is higher than that of structural concrete and a decrease in mechanical strength after SET and SFET becomes small.

Finally, drying conditions to remove residual water from the refractory were determined. According to an experience by the present authors, one problem caused by a residual water is a quick destruction of the test pieces when they were heated quickly by their quick immersion into a hot sodium. The destruction is caused by a quick expansion of residual water and a resultant quick build up of a steam pressure in the test piece. Another problem is a generation of hydrogen due to chemical reaction between sodium and released water when the test piece contacts with sodium. To overcome these problems, drying tests were conducted in a furnace, and a relationship between weight fraction of residual water and drying temperature was made clear. In parallel, measurements of hydrogen generation during the test piece contact with a hot sodium were conducted. Figure 2 shows results. It is seen that hydrogen generation, i.e., water release, proceeded in two steps. The first step is up to 110°C , and the second step is above 200°C . From this figure and results of the test in a furnace, the drying condition was determined to be at higher temperature than 350°C for several hours.

FEASIBILITY TEST OF CERAMIC LINER

A square box type burning pan made of concrete whose inner surface was lined with the improved refractory was fabricated. The pan is 140cm in width, 30cm in depth, and 30cm in thickness, while thickness of the liner was 75mm. In between the liner and the concrete, thermal insulator was installed. In the liner, a stainless steel mesh was installed for its reinforcement. Fabrication of the liner was conducted in three steps as follows. In the first step, a liquefied mixture of water, alumina, silicon-carbide, and additives was poured into a gap between the thermal insulator and a liner flame. In the second step, the flame was removed. In the last step, the liner was naturally and forced dried. These steps were completed in 6 days.

The pan made in this way was subjected to a sodium burning test in which a 30kg sodium was poured in the pan and allowed to burn in air atmosphere for 3.5 hours. Figure 3 shows an arrangement of test apparatuses and the pan for the test. As seen in the figure, the pan was installed in a cell so that a hydrogen concentration change in the cell can be measured. During the test, the maximum sodium pool temperature reached 625°C , and the upper part of the wall liners were exposed to a burning sodium flame. Figure 4 shows temperature histories at various locations in the pan. It is seen that the temperature at 10 mm below the concrete surface never exceeded several tens of degree-C indicating no liner failure. In addition, no hydrogen concentration build up was recorded.

After the test, sodium on the liner was removed, and the liner was subjected to the post-test examinations. Photograph 1 shows pictures of the pan before and after the test. An outer thick part is the concrete, and an inner comparatively thin part is the liner. A white part between the concrete and the liner is the thermal insulator. In the visual observation, a few hair cracks were found at a surface of the liner. But, no deeper sodium penetration into the crack than a few millimeters was observed in the destructive examination. This indicates that the wetting of sodium on the refractory is extremely poor. The maximum sodium penetration in an ordinal surface of the liner was also a few millimeters. Photograph 2 shows a fractured

cross section of the liner. A thin sodium penetration layer can be seen. The results presented are satisfactory. Therefore, one can conclude that the feasibility of a liner was demonstrated.

CONCLUSIONS

From more than fifty commercially available high temperature cement and ceramics, alumina/silicon-carbide mixture base castable refractory was selected as the material for the liner. The selected refractory needed improvement. Thus, its proper weight fractions of additives such as alumina cement and silica and proper drying conditions were determined. Thereafter, a concrete pan lined with the improved refractory was made and was subjected to a sodium combustion test for 3.5 hours. Post-test examination of the liner showed no failure of the liner. Sodium penetration depth in the liner was less than a few millimeters. The results showed feasibility of the ceramic liner.

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REFERENCES

1. Y.HIMENO, S.MIYAHARA, T.MORII, K.SASAKI, "Engineering Scale Test on Sodium Leak and Fire Accidents and Its Consequences in Auxiliary Building of Fast Breeder Reactors," *Int. Conf. Liquid Metal Eng. Technol.*, TS-15, #202, Avignon, France, Oct. 1988.
2. L.MUHLNSTEIN, et al., *Nucl. Safety*, Vol.125, No.2, 1984.
3. H.HIROI, Y.HIMENO, "Corrosion of Concrete by Sodium (in Japanese)," *Proc. 1990 Annual Mtg. Atomic Energy Society of Japan*, C18, p114, May, 1990.

Table I Results of Selection Test for Ceramics

No.	Material ⁽¹⁾	Main Composition (w/o)	Test Results ⁽²⁾			
			650°C TST	650°C SET	SFET	OAP
1 a	Dense Magnesia (s)	MgO : 99	○	◎	N.A	○
1 b	Porous Magnesia (s)	MgO : 95, SiO ₂ : 3	◎	△	×	×
4 a	Dense Alumina (s)	Al ₂ O ₃ : 98	×	destroyed	○	×
4 b	Porous Alumina (s)		◎	○	○	○
4 c	do. (c)		◎	◎	○	◎
8	Porous Alumina Titanate (s)	Al ₂ O ₃ : 54, TiO ₂ : 38	×	×	×	×
9 a	Dense Zirconia (s)	ZrO ₂ : 93, Y ₂ O ₃ : 5	◎	◎	△	○
9 b	Porous Zirconia (s)	ZrO ₂ : 88, Al ₂ O ₃ : 10	×	△	×	×
10a	Dense Silicon-Nitride (s)	Si ₃ N ₄ > 96	◎	◎	○	◎
10b	Porous Silicon-Nitride (s)	Si ₃ N ₄ : 98	◎	◎	◎	◎
20	Porous Alumina Silicon-Carbide (s)	Al ₂ O ₃ : 46, SiC : 47	◎	○	○	○

Note

(1) s : sintered
c : castable

(2) TST : Thermal Shock Test
SET : Sodium Exposure Test
SFET : Sodium Flame Exposure Test
OAP : Over All Performance
× : bad
△ : poor
○ : good
◎ : excellent

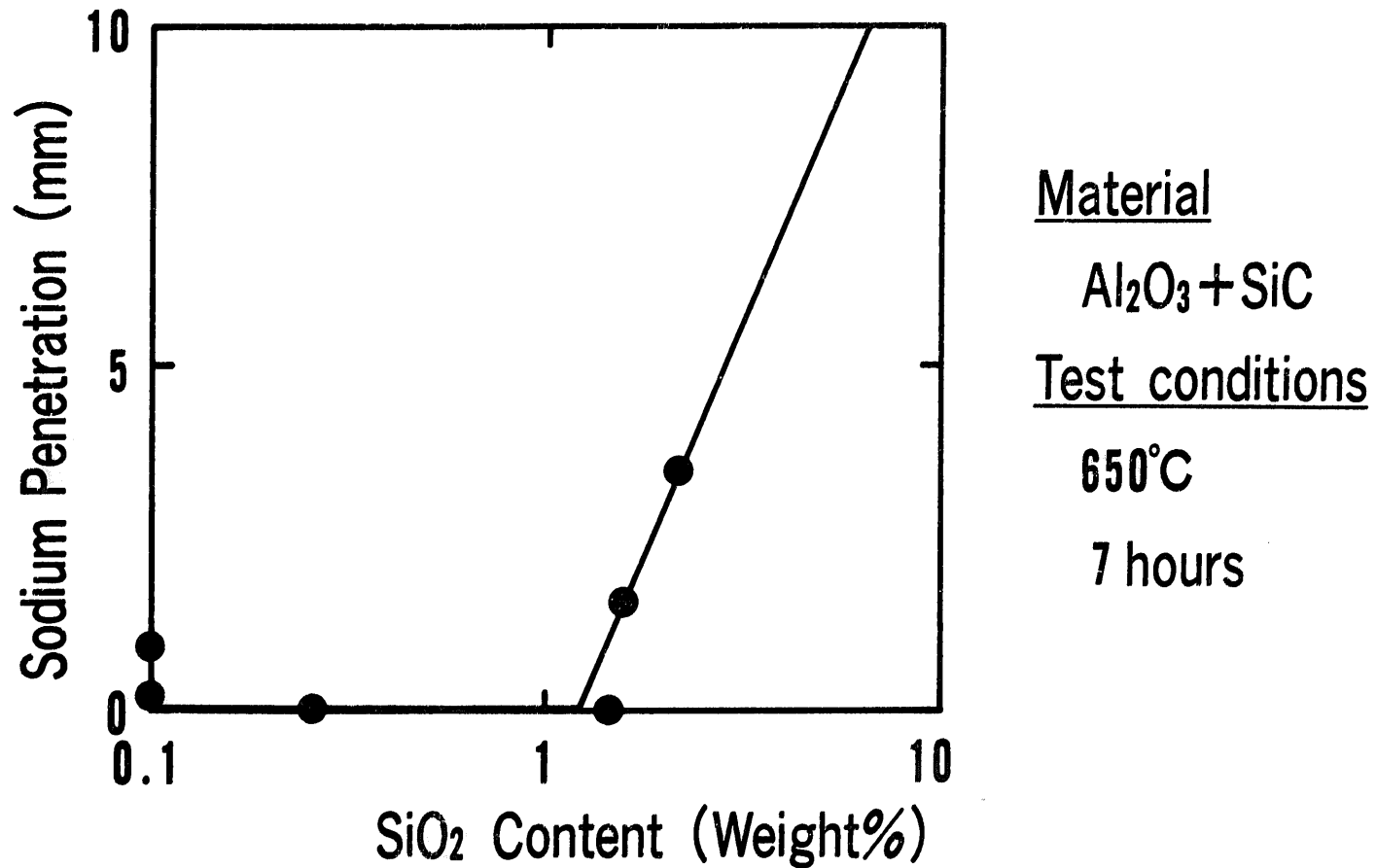


Fig. 1 Corrosion Resistance of Refractory
with Various Silica Contents

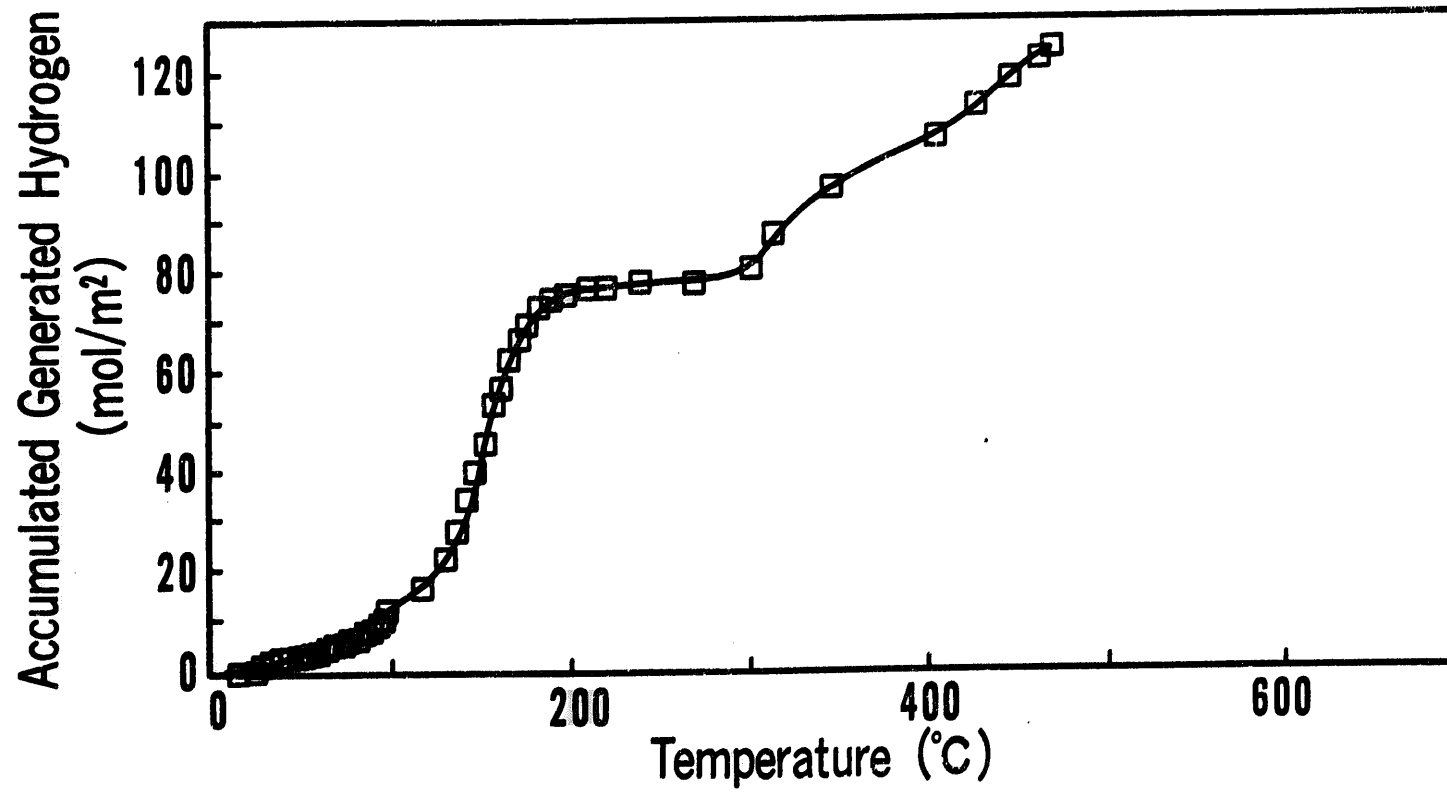


Fig. 2 Hydrogen Generation versus Temperature of Test piece

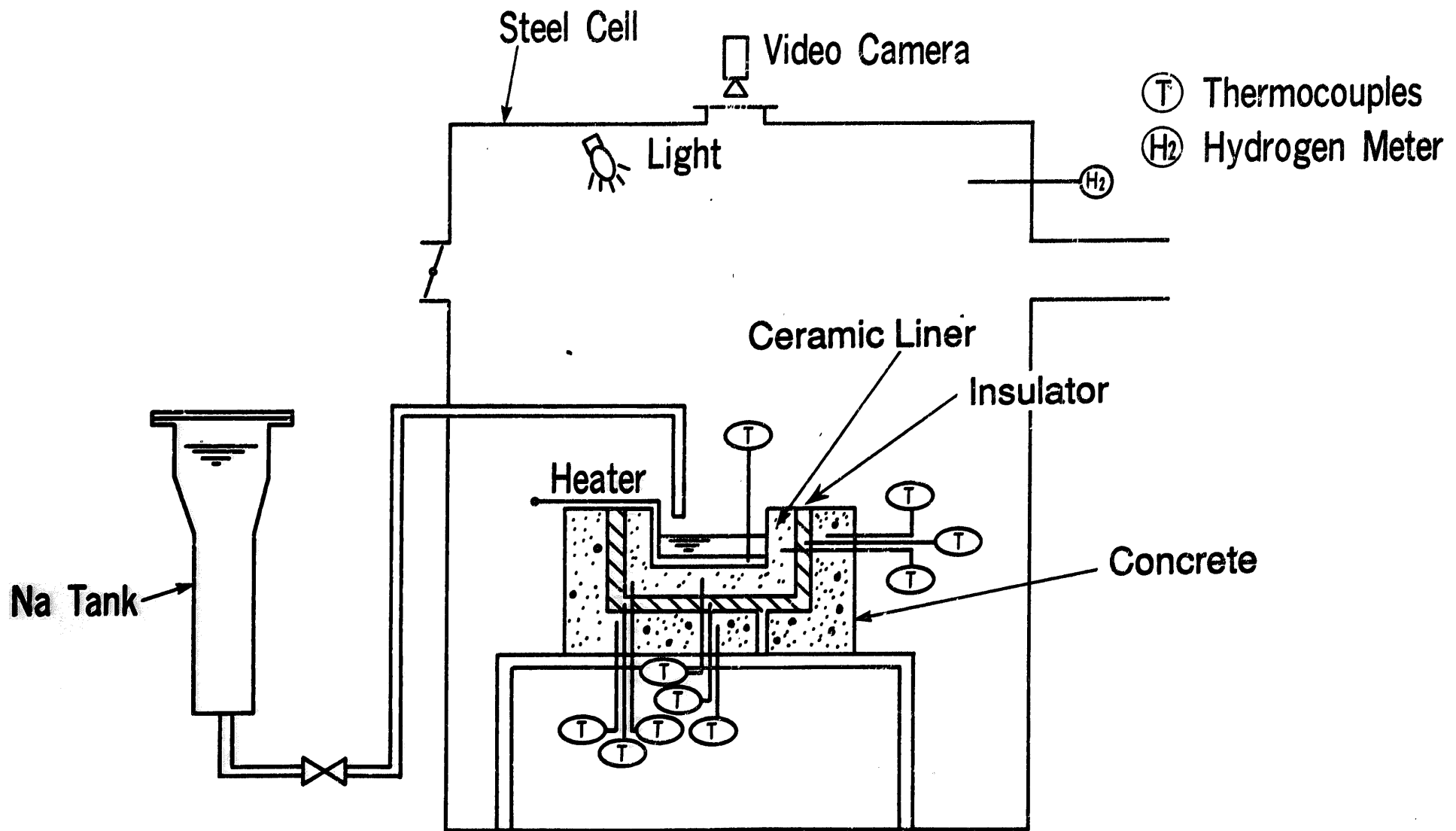


Fig. 3 Test Arrangement

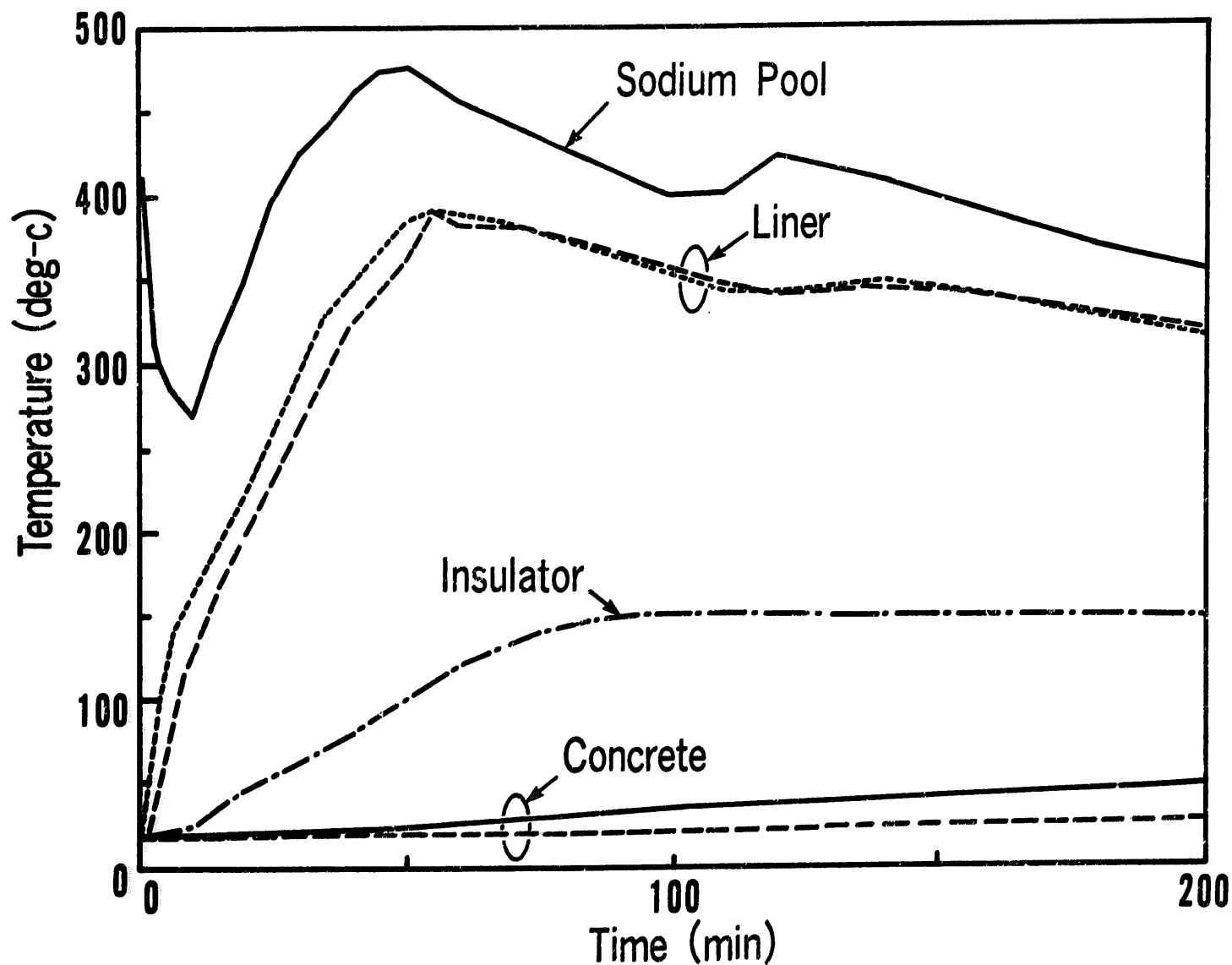
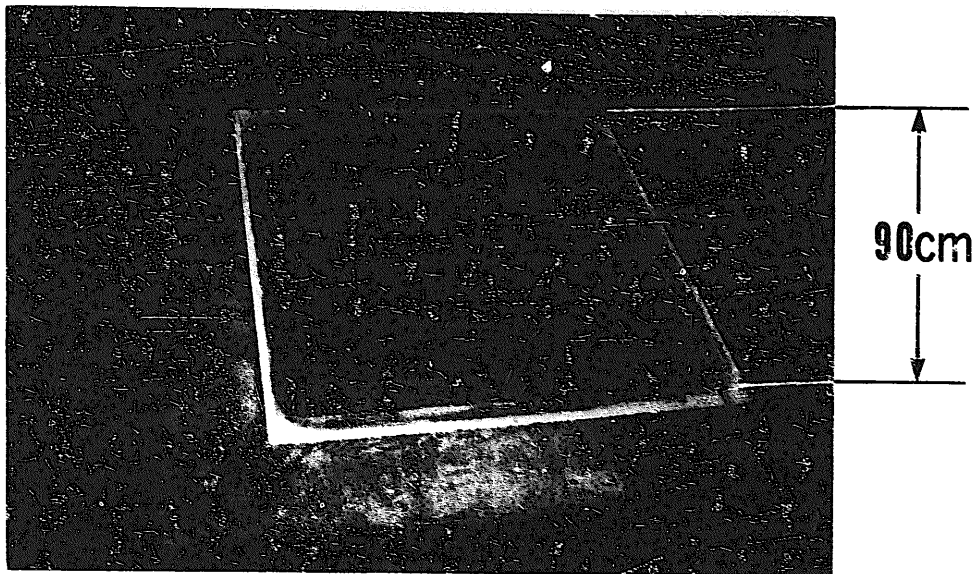
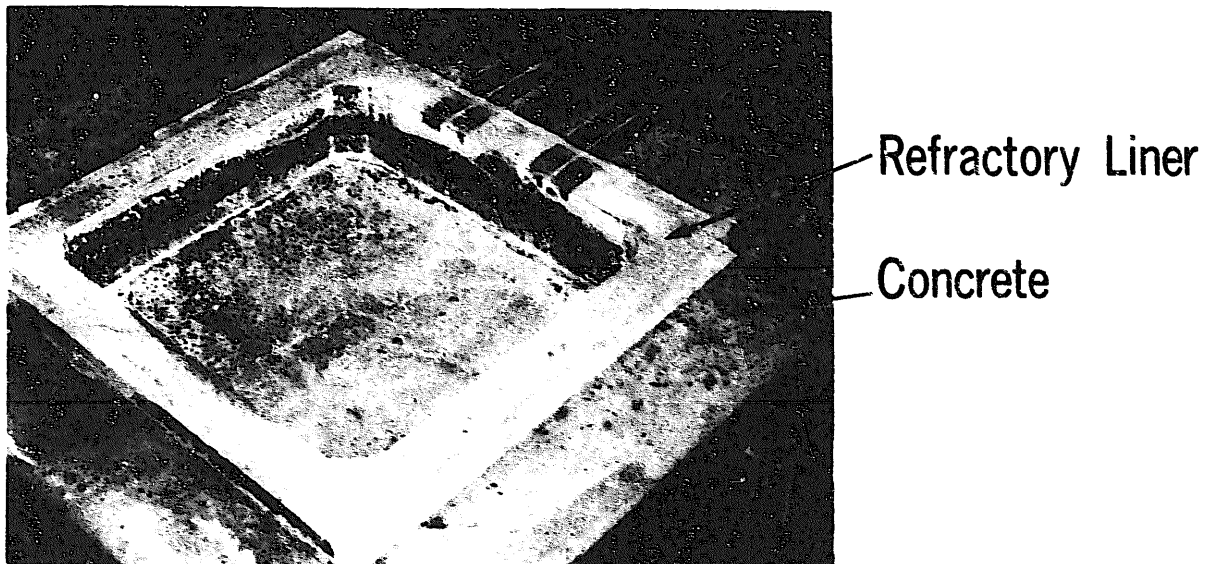


Fig. 4 Temperature Histories of Burning Pan with Liner



Pre-Test



Post-Test

Photo. 1 Burning Pan made of Concrete
Lined with Refractory for
Feasibility Test

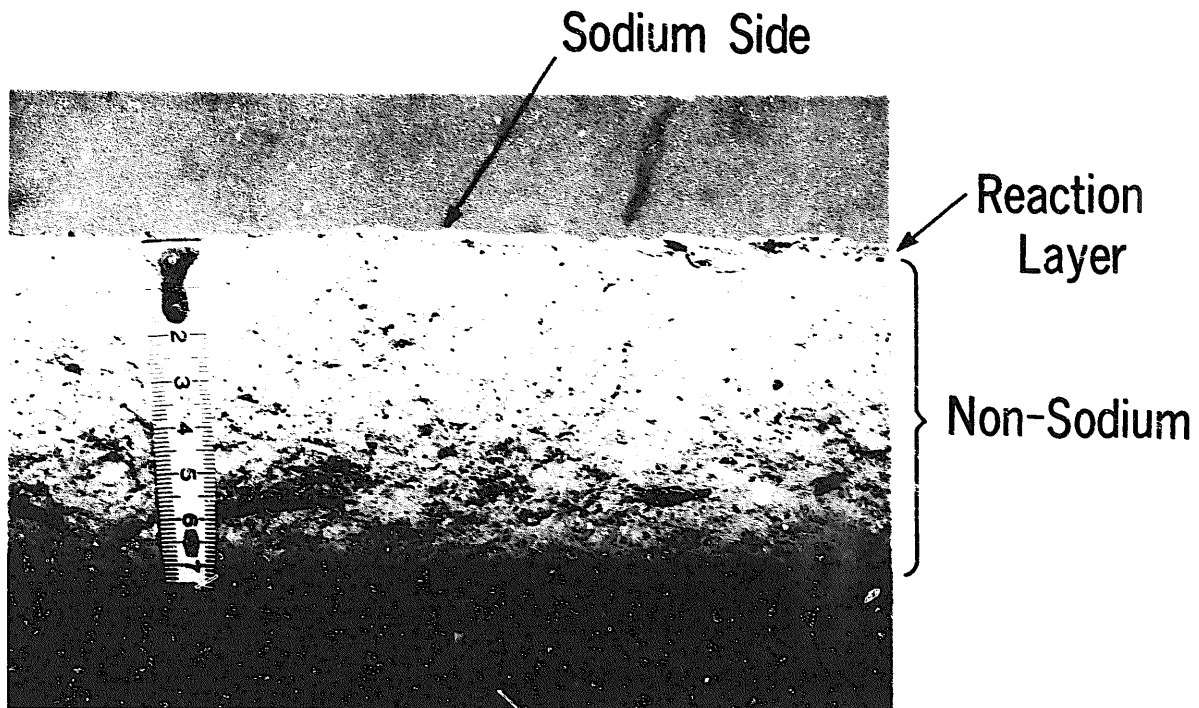


Photo. 2 Post-test Cross Section of Refractory Liner