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HIGH PRESSURE SOXHLET TYPE LEACHABILITY TESTING DEVICE AND LEACHING TEST OF SIMULATED HIGH-LEVEL WASTE GLASS AT HIGH TEMPERATURE

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A High Pressure Soxhlet Type Leachability Testing Device (HIPSOL) was developed to evaluate long-period stability of high-level waste (HLW) solids. For simulated HLW solids, temperature dependency of the leachability was investigated at higher temperatures from 100°C to 300°C at 80 atm. Leachabilities of cesium and sodium at 295°C were 20 and 7 times higher than at 100°C, respectively.

In the repository, the temperatures around solidified products may be hundred °C. It is essential to test them at such elevated temperatures. HIPSOL is also usable for accelerated test to evaluate long-period leaching behavior of HLW products.

Keywords; High-Level Waste, Long-Period Stability, Leachability, High Pressure, Cesium, Sodium, Solidified Waste, Soxhlet Type Device, Solidified Waste, High Temperature.

耐圧ソックスレー型浸出率測定装置および高レベル模擬廃液の高温浸出試験

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高レベル廃棄物固化体の長期安定性の評価および地層処分条件を考慮した浸出率測定を行なうため，新たに耐圧ソックスレー型浸出率測定装置の開発を行なった。

この装置を用いて，100℃から300℃までの間の模擬廃棄物ガラス固化体の浸出率温度依存性の検討を行なった。

295℃における浸出率は，100℃における値に比べ，セシウムでは約20倍，ナトリウムでは約7倍の増加が認められた。

処分地層内では，処分初期において，固化体近傍で，数100℃になることが予想されるので，高温における浸出率の評価は不可欠のものである。また，本装置は，長期間の浸出率の評価を行なうためにも，高温における加速浸出試験法として適用可能である。

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1. Introduction

The management of high level radioactive waste (HLW) from the reprocessing of spent fuel has been focussed into a series of confinement method by stable and less-dispersible conditions such as the vitrification with borosilicate glass and the isolation in a suitable geological formations after the interim storage [1].

The liachability of the radionuclides from the solidified waste products (glass, ceramics) is one of the most inportant parameters in the evaluation of the products in the repository because the leaching is the first step of the nuclide migration by ground water in the repository from which activity could be released to the environment over a long period of time.

A Soxhlet type leachability testing device [2] is one of the excellent apparatus for the leaching test of the radionuclides from the product because the sample is always leached by distilled water and does not come into contact with the balk leaching liquor containing the dissolved constituents. Therefore, the test is suitable for analysing the basic leaching mechanisms and it can be one of the standardized methods for the leachability test with different waste contents in the solid and for the leachability test for the geological disposal assessment. Almost all tests of this type were done below 100°C at 1 atmospheres.

In order to predict the long term leachability of the solidified products, it is desirable to accelerate the test at higher temperature more than 100°C. Moreover, to simulate the actual environmental conditions during the geological storage, the suitable leaching temperature more than 100°C might be essential [3-4].

In this report, a newly developed <u>Hi</u>gh <u>P</u>ressure <u>So</u>xhlet Type Leachability Testing Device named HIPSOL is described and the temperature dependency on the leachability has been investigated with the simulated HLW solid at higher temperature than 100°C upto 300°C at 80 atmospheric pressure.

2. Apparatus

In Fig. 1, the line drowing of HIPSOL is described. HIPSOL consists of a 3kW of Ni-Cr heater and a 2 ℓ of a pressure vessel

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2. Apparatus

In Fig. 1, the line drowing of HIPSOL is described. HIPSOL consists of a 3kW of Ni-Cr heater and a 2 ℓ of a pressure vessel made of SUS 316 for the test at various temperature up to 300°C at 80 atmosheric pressure. The vessel which is equipped with a reflux cooler, a sampling apparatus, a liquid supply and an evacuation line. The vessel also contains a 62 ml of leaching cell made of SUS 316 connected with an automatic syphon, from which signals are taken to be counted the number of the leachant exachange which is controlled at about 30 cycles per minutes manually. The testing sample in the form of a granular glass or a block can be placed in a 31 ml of 100-mesh wire made of SUS 316 cage in the leaching cell. The leachant with the leached waste is concentrated at the bottom of the vessel under the pressure, and the distilled water is introduced from the reflux condenser to the leaching cell and the leaching is carried out in the intermittent way by the automatic syphon.

3. Experimental

The component of the liquid simulated HLW [5] with 90.6 g/ℓ of solid based on metal oxides is shown in Table 1. The recipe of the borosilicate glass is shown in Table 2, in which natural zeolite is used as the source of SiO_2 and Al_2O_3. Zeolite was obtained from Hamado-kosan Ltd. Co. at Oshamanbe-Hokkaido, Japan and the component is shown in Table 3.

The simulated liquid waste was calcined at 650°C in the maximum temperature for 15 minutes as the residence time in the rotaly-kiln type calciner [6].

The vitrification was performed by induction heating of the calcine with the glass forming materials at 1200°C for 2 hrs in a metal melter made of KRIMAX® [7]. The molten glass was poured into a graphite mold and the glass was crashed to granules, which were screened between 35- and 60-mesh shieves.

Six grammes of the granular glass in a 100-mesh wire cage were placed in the leaching cell and 300 ml of distilled water was poured into the vessel. After the vessel was shut closely and evacuated, it was heated up to the setting temperature and was kept constant for 2 hrs. At the end of the leaching period, the vessel was cooled down to the ambient temperature.

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The volume of the recovered leachant was measured and the concentration of the cesium and sodium in the leachant was measured by an atomic absorption analysis method. The leaching rates of the cesium and sodium were calculated based on the recipe of the initial glass shown in Table 1 and 2 by using the concentration of each element and the volume of the leachant.

4. Results and Discussion

One of the operating conditions of HIPSOL is shown in Fig. 2. This figure shows the leaching temperature (A) was kept equal to the water temperature at the bottom of the vessel (B), and the interval of the leachant exchange (C) was also kept constant (ca. 31 cycles/hr) through out the test.

The leaching rates of the cesium and the sodium from the borosilicate glass obtaind at 100, 150, 200 and 295°C using HIPSOL are presented in Fig. 3. The leachability of the cesium was 0.2 and 4.0 w %/2hr at 100 and 295°C, respectively. On the other hand, the leachability of the sodium was 0.2 and 1.3 w %/2hr at 100 and 295°C, respectively. The temperature dependency on the cesium leachability was a little higher than that of the sodium. The reason of this difference is not clarified yet, however, it is possible that the diffusion controlled leaching rate of the cesium hydrated ion is higher than that of the sodium hydrated ion because the diameter of the sodium hydrated ion is bigger than that of the cesium hydrated ion [8-11].

It should be emphasized that the temperature dependency for leaching rate both of the cesium and the sodium was remarkable, therefore, the leaching test at such high temperature is suitable for predecting the long term leaching behabior during the storage or the isolation.

5. Conclusion

A newly developed High Pressure Soxhlet Type Leachability Testing Device (HIPSOL) for leachability testing at high temperature is described. The leachability of cesium and sodium depended strongly on the leaching temperatur (Fig. 3), therefore, it is essential to test at such elevated temperature as the condition at which the solidified HLW would be

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Reference

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Table 1 Recipe of simulated high-level wastes

Oxides	Oxide weight (g/l)	Weight-%
Na_2O	30.37	34.2
Rb_2O	0.62	0.7
Cs_2O	4.79	5.4
SrO	1.66	1.9
BaO	3.37	3.8
Y_2O_3	0.99	1.1
La_2O_3	2.61	2.9
Ce_2O_3	4.32	4.9
Nb_2O_3	12.11	13.6
ZrO_2	6.25	7.0
MoO_3	10.64	12.0
Fe_2O_3	6.06	6.8
CoO	0.45	0.5
NiO	2.17	2.4
TeO_2	1.21	1.4
Cr_2O_3	0.56	0.6

Total oxide weight; 88.18 g/l

Acid concentration; 2N HNO_3

Table 2 Recipe of boro-silicate glass

Component	Percent weight %
FP oxides	20.0
B_2O_3	12.0
Na_2O	16.0
CaO	5.0
Zeolite	47.0

Table 3 Composition of natural zeolite

Contituents	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	K_2O	Na_2O	MnO	P_2O_3	TiO_2
Weight-%	76.5	7.05	3.77	2.98	1.92	2.28	2.40	0.11	2.17	0.12

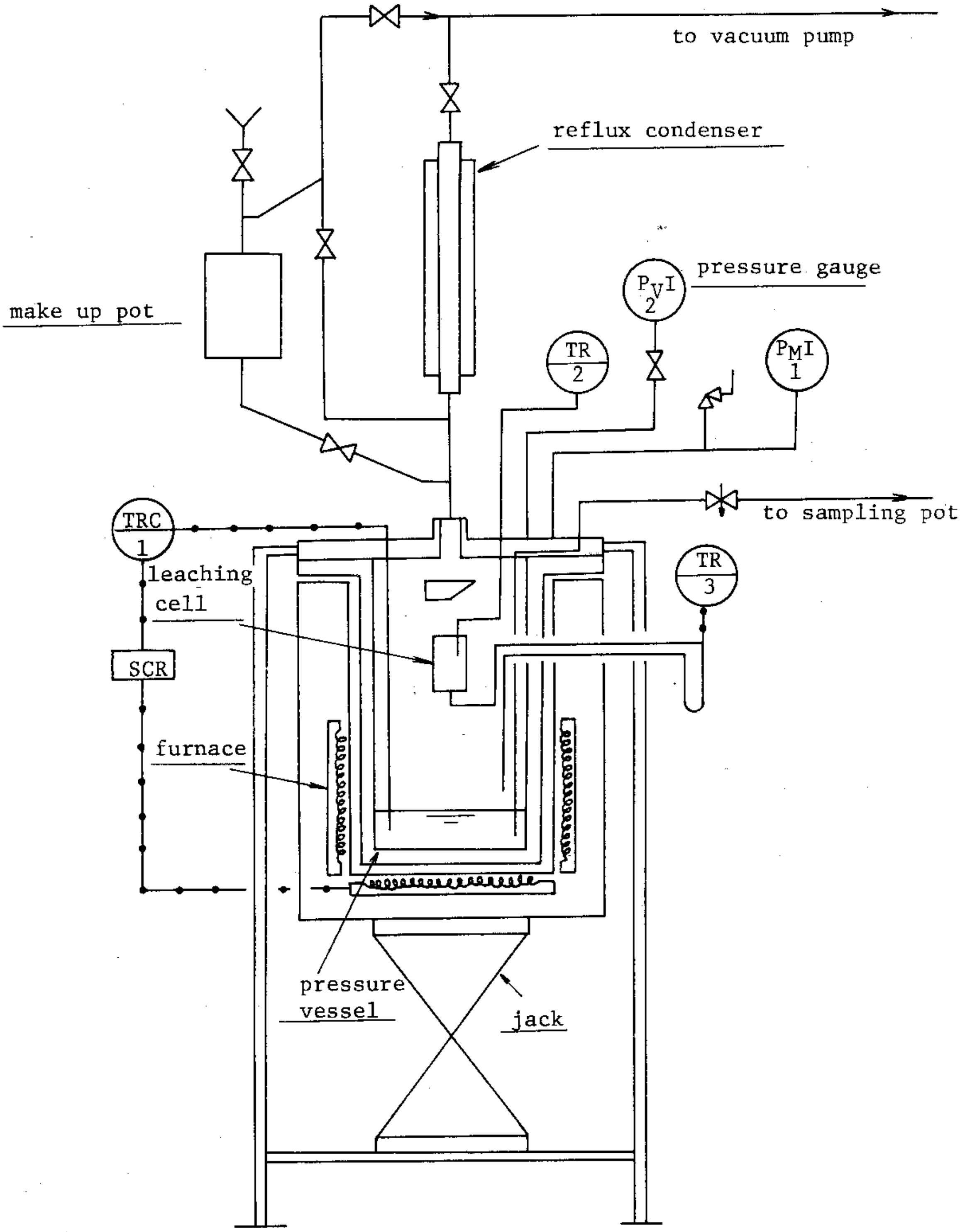


Fig. 1 Line drawing of HIPSOL

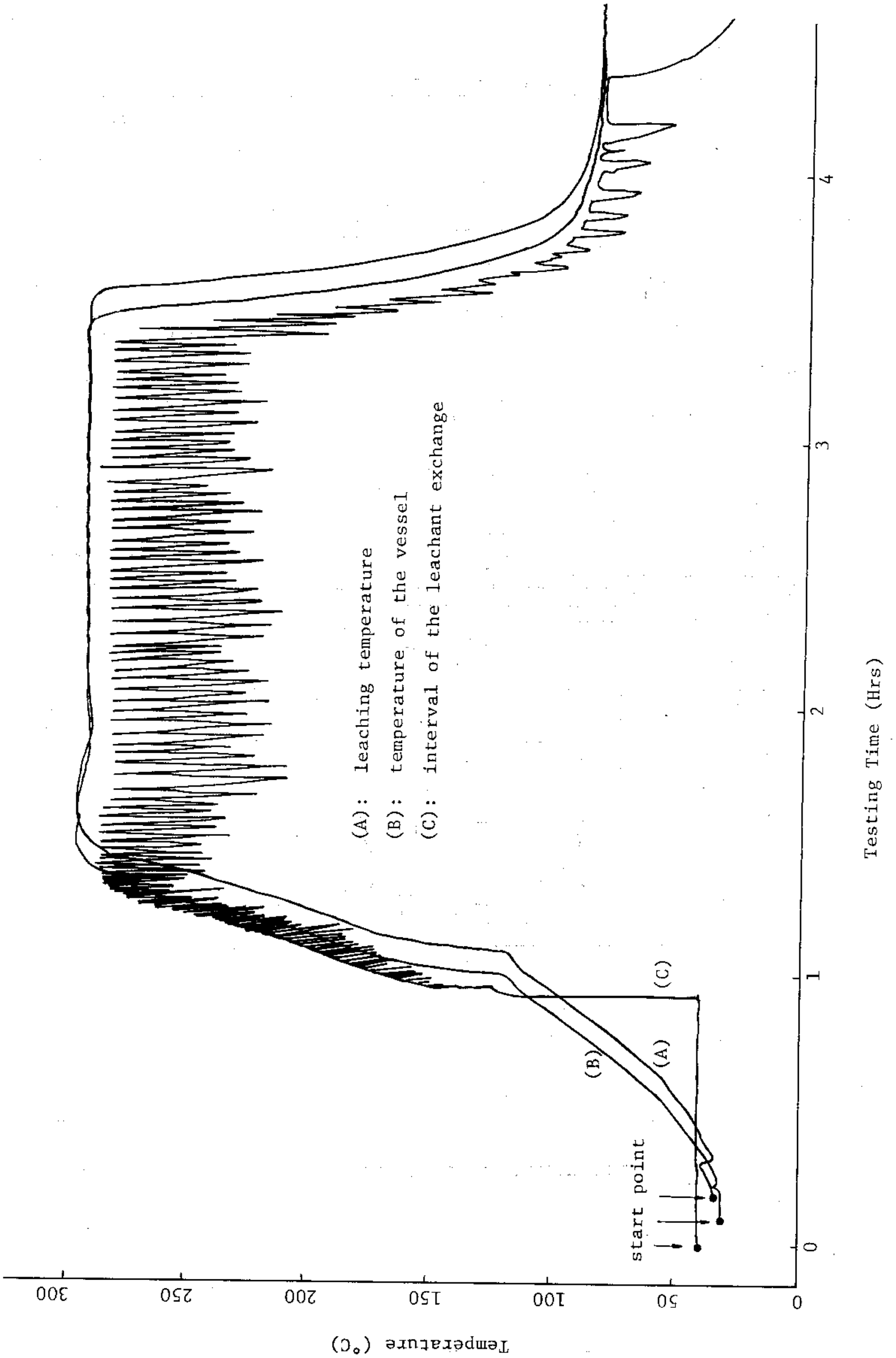
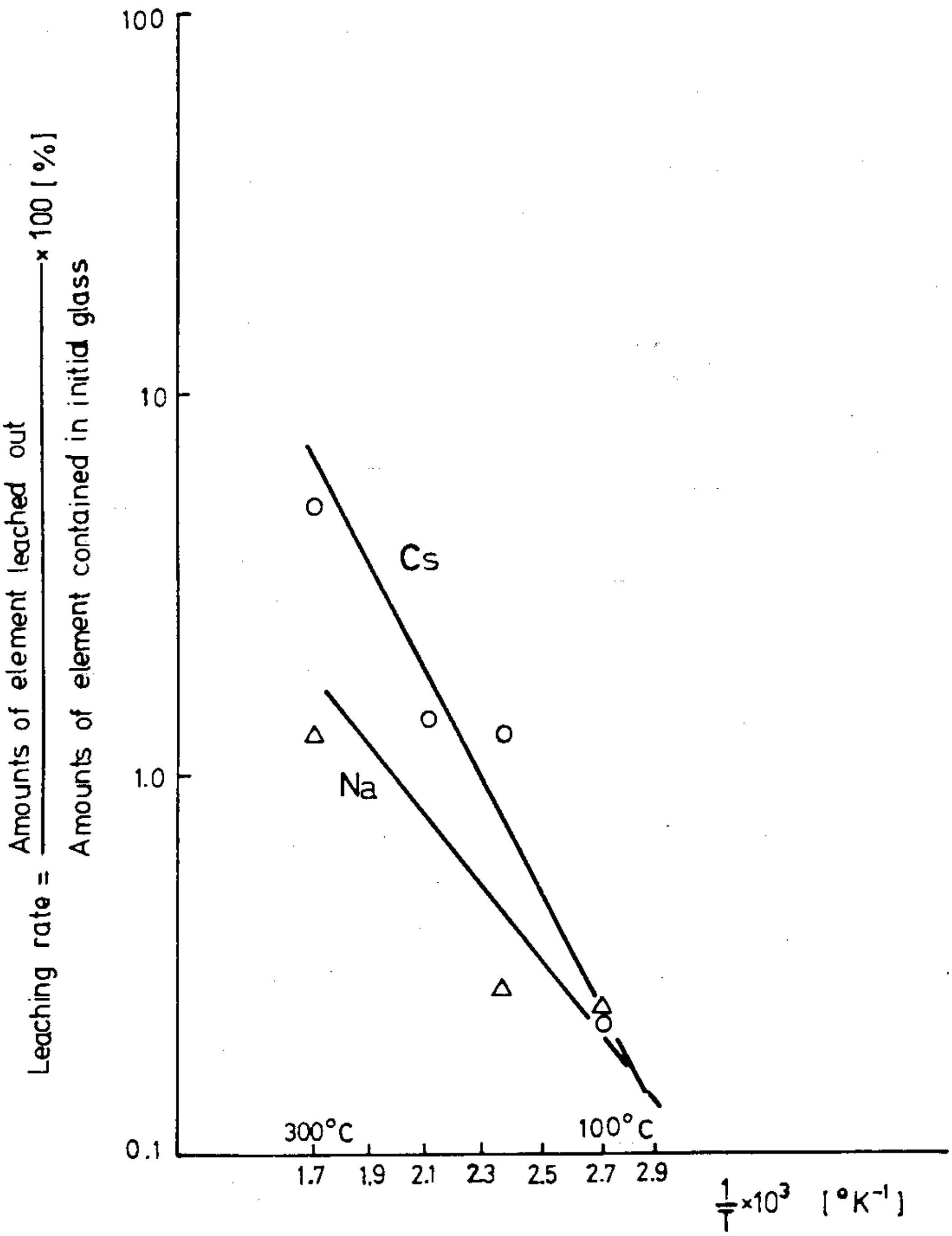


Fig. 2 Operating condition of HIPSOL



T : Leaching temerature

Sample : JG-1 35 ~ 60 mesh 6.0 g

Leachant : Distilled water 300 ml

Leaching time : 2hr

Device : High pressure Soxhlet type leachability testing device (HIPSOL)

Fig. 3 Leaching behavior of Na and Cs from glass products containing simulated HLW