Experimental Investigation of LWR-Core Material Release at Temperature Ranging from 1500-2800°C

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Summary

The release experiments are conducted at the melting plant SASCHA (Fig. 1). Instead of highly radioactive spent fuel, so-called fissium pellets are used which contain the following fission product elements with normally 6 of them as radioactive nuclides:

Se, Zr, Mo, Ru, Ag, Cd, Sb, Te, I, Cs, Ba, Ce, and Nd.

Irradiated steel and Zircaloy are used together with normal (undoped) UO_2 pellets for determination of the release fractions of Cr, Mn, Fe, Co, Zr, Sn, and Sb.

The experimental details are given below:

mass of melt material	150-200 g
fraction of UO_2 (fissium)	0.6
pressure of furnace atmosphere	0.5-2.0 bar
flow rate	10-50 l/min
max. test temperature	1800-2800°C
time at const. temperature	$\leq$ 60 min
collection-time for on-line γ-spectra	60 sec

Following the experiment, the total release of each nuclide is determined by measuring the gamma-ray activity of the deposits on the glass vessel and on all tubes of the aerosol transport line.

In order to calculate the total release as a function of time, it is assumed that a constant relationship exists between the activity found on the filters and the total activity released:

$$A_{total} = ß \cdot A_{filter}$$

with ß not depending on time or temperature. By use of this factor ß, all results of the on-line measurement are converted to total release values.

1606 313

The total release of the most relevant fission products with high and medium volatility are plotted as a function of temperature in Fig. 2.

The low release values for I, Cs, and Te at temperatures below 1800°C can be explained by two different effects:

- compound formation, i.e. chemical reactions with the UO_2 matrix and/or with the cladding material and other fission product elements,
- retention in intergranular bubbles.

For the conditions specified in Fig. 2, the release of I and Cs was found to be complete at temperatures of 2100°C and 2400°C, respectively. The release curves of Te and Sb relative to those of I and Cs are qualitatively in accord with the differences in the vapor pressure of the elements.

The percentage release as a function of time at constant temperature is plotted in Fig. 3. It can be seen that the integral release at the end of both tests is almost the same for all three elements. There is no doubt that the release is due to solid-state diffusion from the UO_2 matrix under the conditions of these experiments.

In Fig. 4 the results of two experiments with similar heating curves, but different pressure (0.5 bar/2.0 bar) of the furnace atmosphere, have been plotted as a function of time. It turned out that the release was considerably higher for all elements at the pressure of 2 bar. The same result was found from two corresponding experiments at 1800°C in which the release of some additional elements (Se, Ag, Cd, Cs besides Mo, Sb, and Te) was measured.

The first post-examinations of the fuel samples by optical microscopy revealed an increased number of cracks and larger void spaces in the samples of the 2-bar-experiments. These findings - though not quantitatively evaluated at present - are interpreted as the reason for the higher release at 2 bar.

The curves in Fig. 5 and 6 are calculated by converting the results of all release tests in air of 2 bar into release rates, expressed in units of (% release·min^{-1}).

For calculating the release at constant temperature, the following simple formula may be used:

$$\Delta m = m_o \, (1 - e^{-\alpha \cdot t})$$

$\frac{\Delta m}{m_o}$ = relative mass release, α = release fraction, t = time

1606 314

Besides the release of fission products from an overheated core, one has to expect also considerable release of the activation products contained in the cladding and structural material of a LWR. The results tabulated below have been obtained for the same conditions as for the fission products, i.e. for an atmosphere of air and a pressure of 2 bar.

T (°C)	Release Rates (% per min)						
	Cr	Mn	Fe	Co	Sb	Sn	Zr
2350	0.055	0.96	0.024	0.023	1.05	0.74	0.003
2750	7.69	80.5	4.01	2.10	29.1	23.4	0.04

For comparing the results on the fission product release with the best estimate values of the Reactor Safety Study (RSS), it may be assumed that a reactor core attains a temperature of 2600°C for 20 min. If the release during heat-up is neglected and the release rates of Fig. 5 and 6 are applied, it is found that the total release will be higher for the elements Sb and Te and lower for the elements Ba und Ru compared to the RSS. It should be mentioned, however, that (a) the release rates used here may not be valid for an atmosphere of steam and (b) the conditions of time and temperature were chosen somewhat arbitrary for this comparison.

The deposition behavior of the released material is shown in Fig. 7 for two different flow rates of air. Two results are apparent:

- the low volatile elements tend to deposit within smaller distances from the melt than the high volatile elements
- the relative amount of released products, which is transported to the filters, increases with increasing flow rate.

References:

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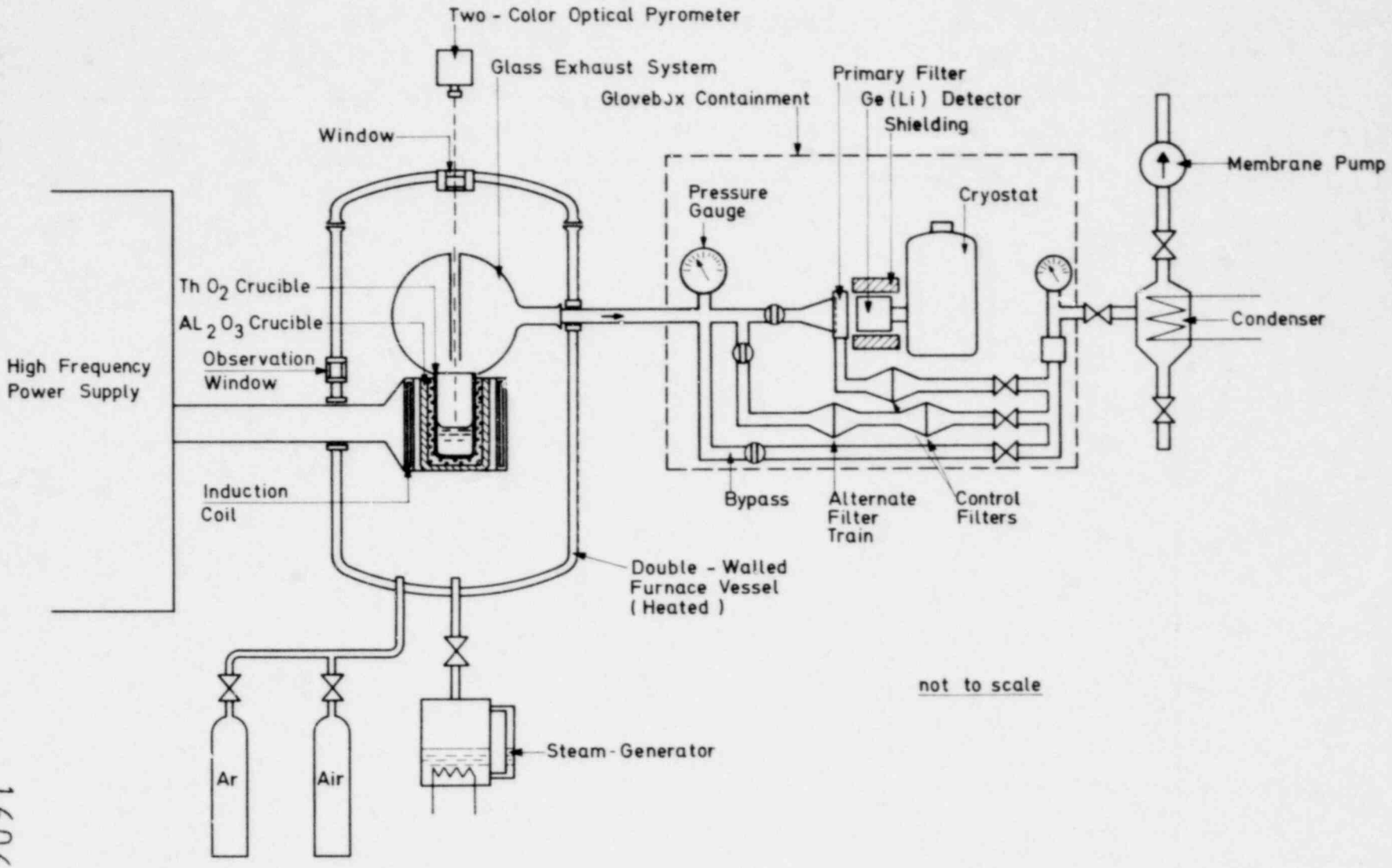

Fig. 1. Experimental Apparatus for Core Melt Release Studies

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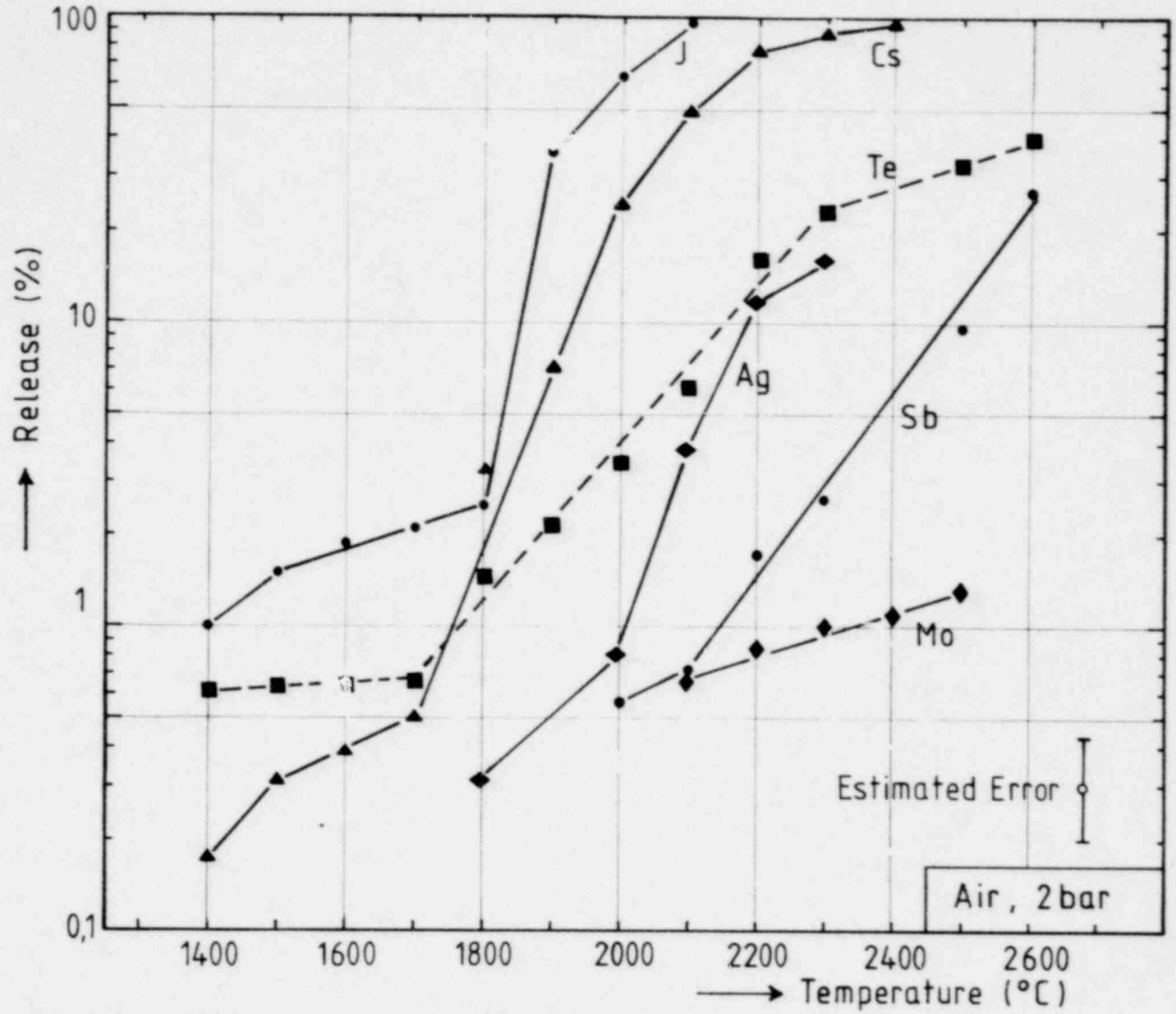

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Fig. 2. Fission Product Release During Linear Heat-up With 110 °C/min

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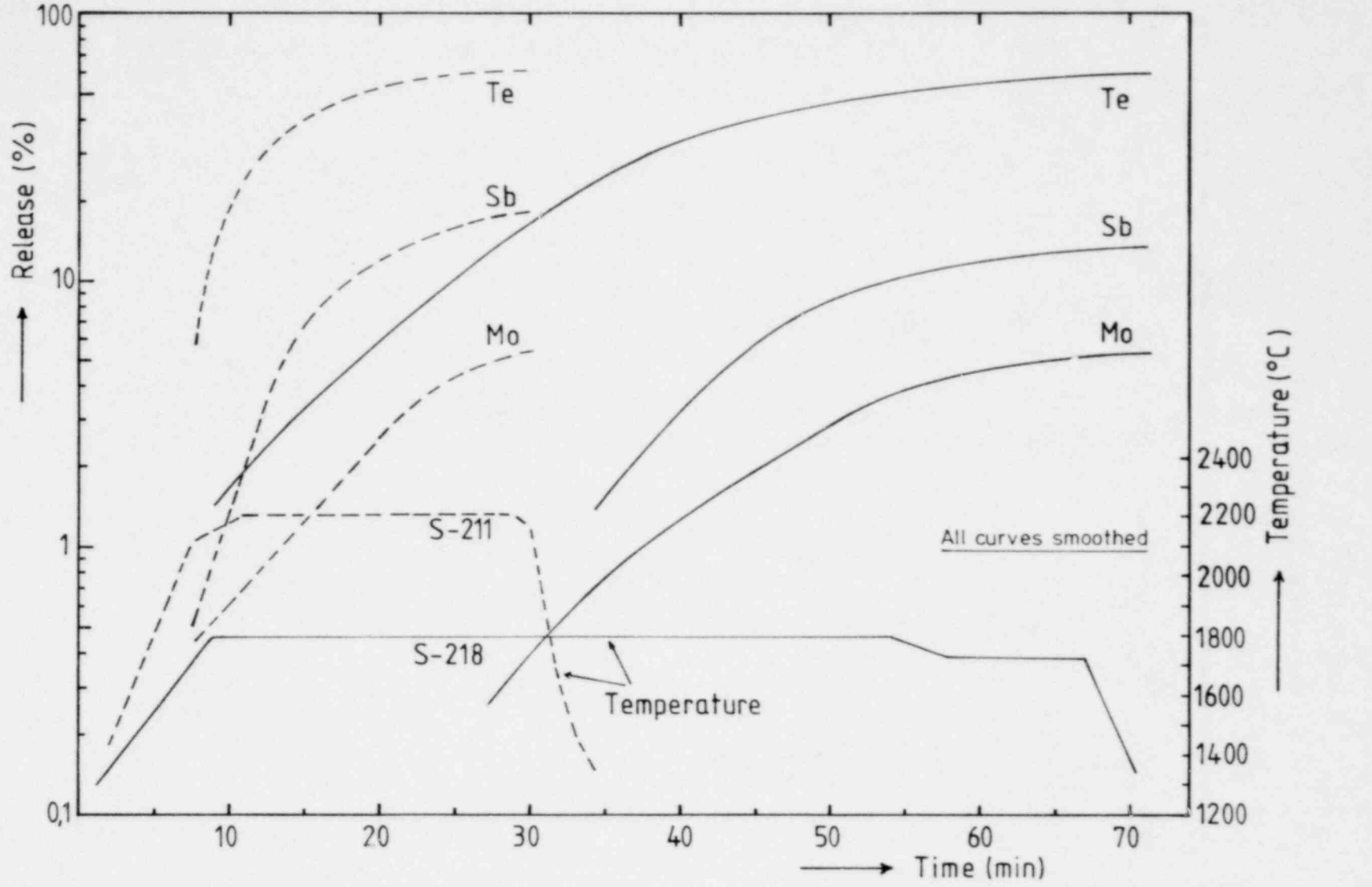

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Fig. 3. Release in Air (2 bar) as a Function of Time, Parameter: Temperature
---- 20 min at 2200 °C (Test S-211), —— 45 min at 1800 °C + 12 min at 1720 °C (Test S-218)

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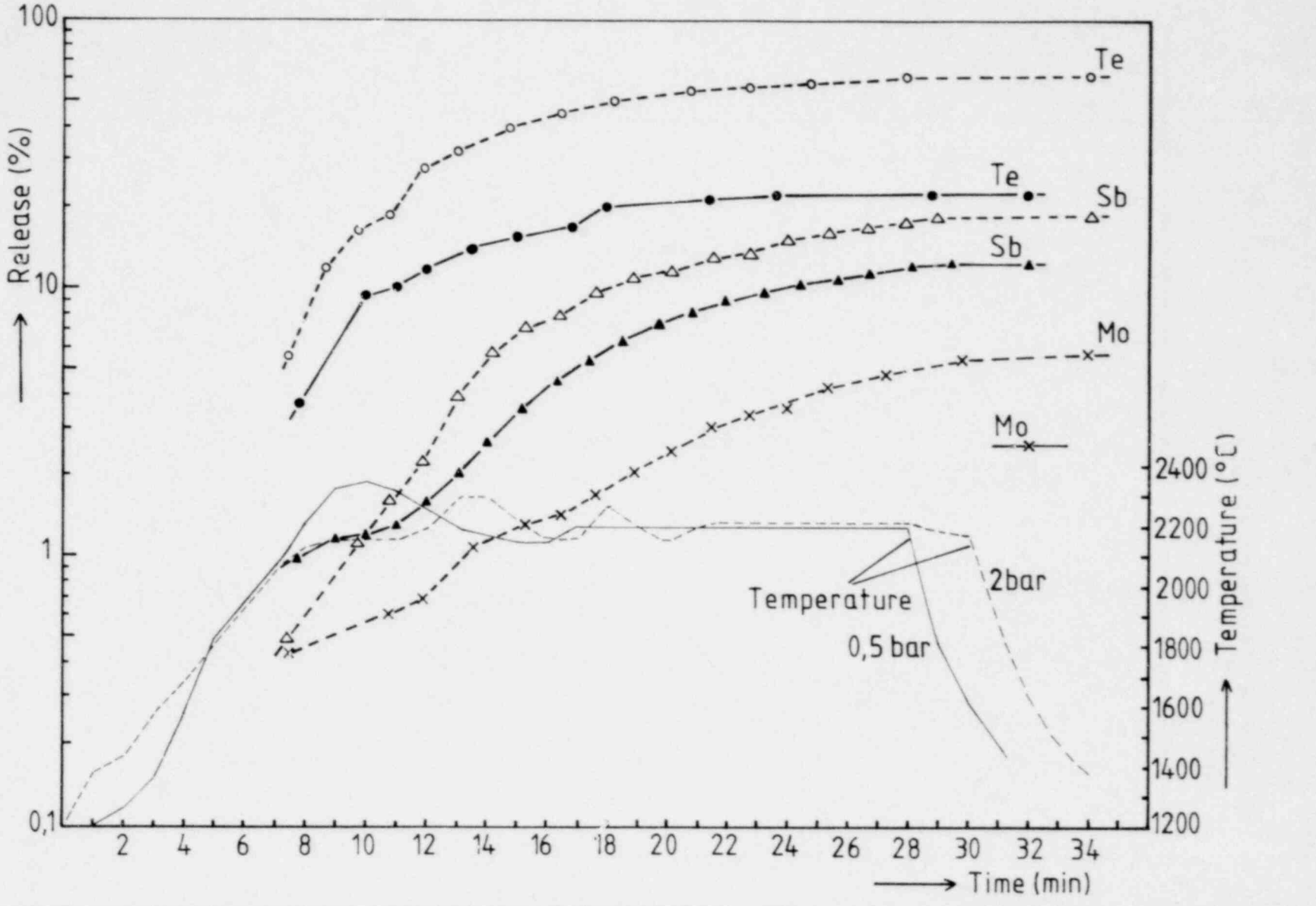

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Fig. 4. Release in Air as a Function of Time, Parameter: Pressure
——0,5 bar (Test S-210), – – – 2 bar (Test S-211)

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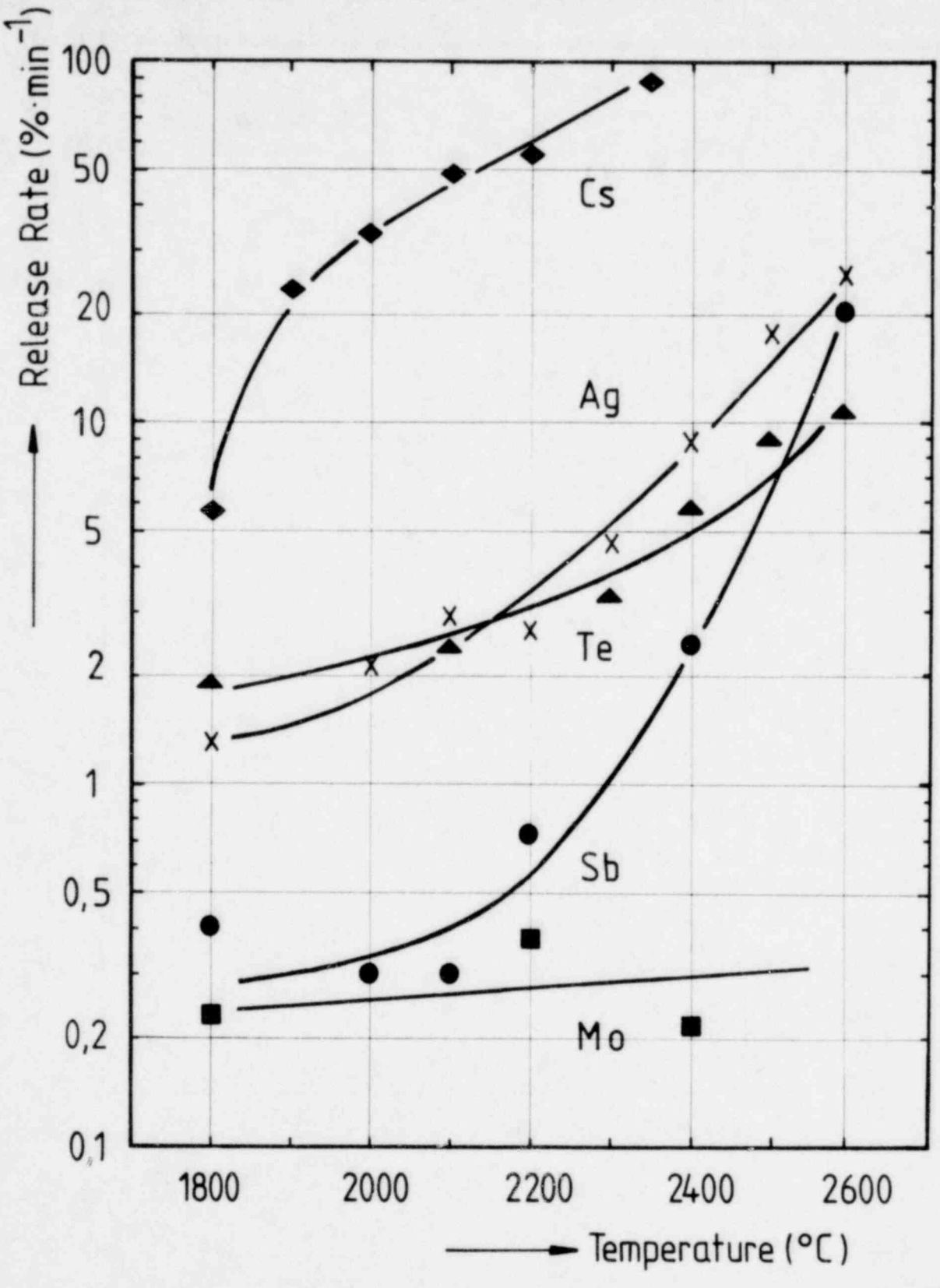

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Fig.5. Fission Product Release Rates in Air (2 bar)

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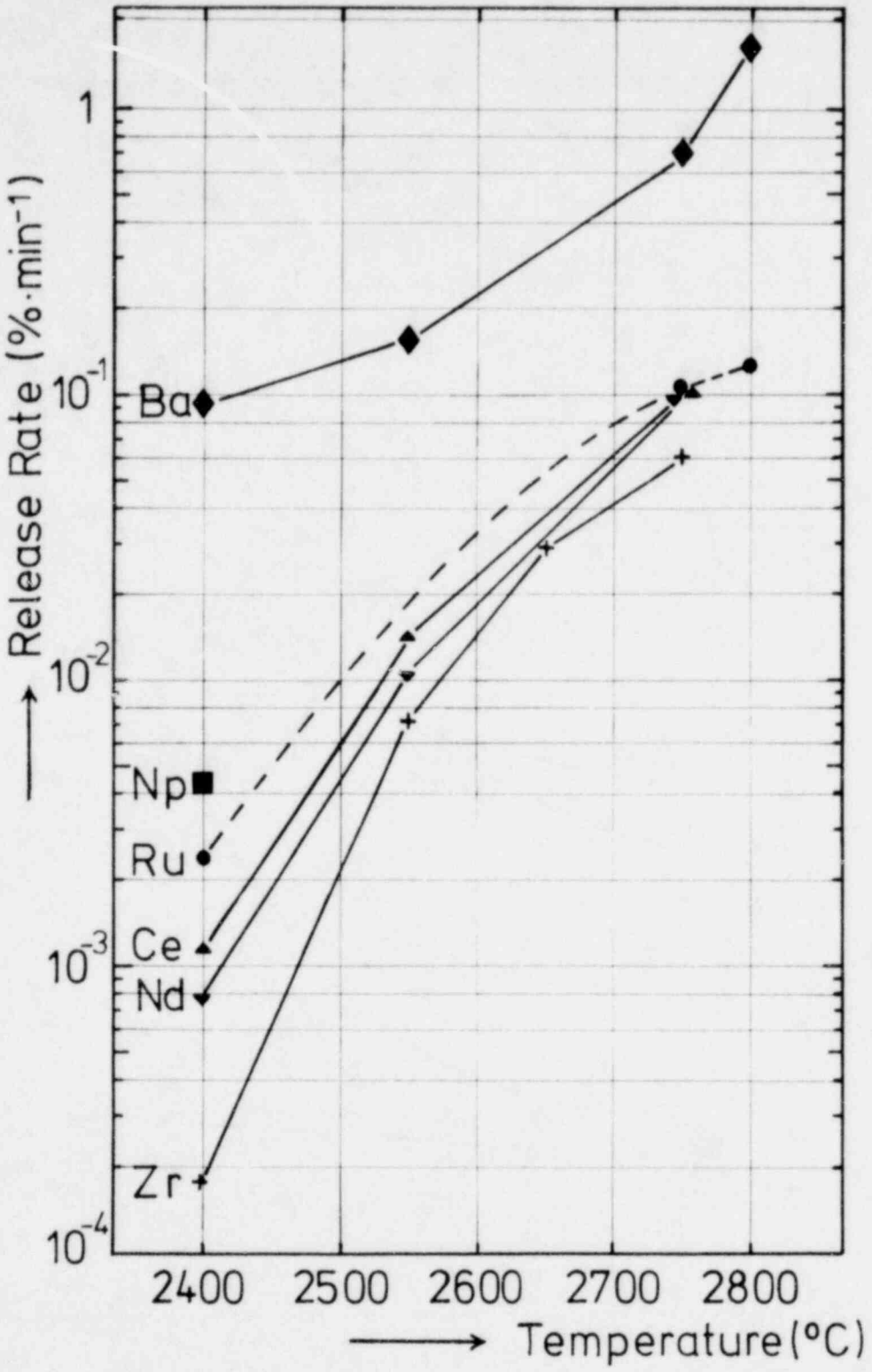

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Fig.6. Fission Product Release Rates in Air (2bar)

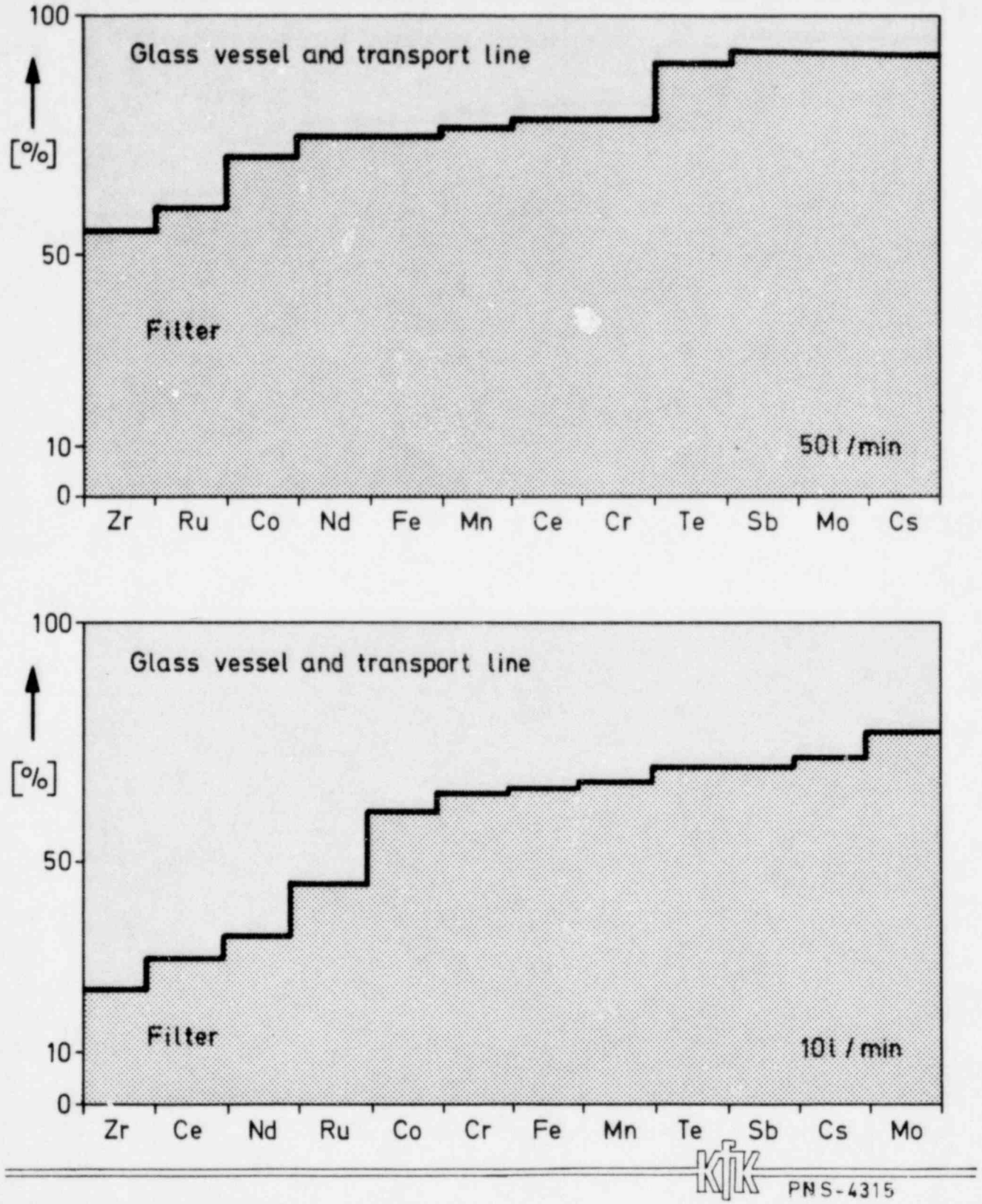

Fig. 7 Deposition of the released material for two different flow rates

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