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in Phebus Tests FPT0 and
FPT1 and the Possible
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Fission Product Speciation in Phebus Tests FPT0 and FPT1 and the Possible Influence of Boron

Abstract

The thermochemical calculations performed with a large number of elements to describe the chemical system in a Phebus FP test allow the following conclusions:

The metallic absorber materials play a significant role in fission product speciation. At around 1000°C and above, silver iodide is the dominant iodine carrier in a full steam atmosphere in both FPT0 and FPT1. At lower temperatures, cadmium iodide takes over. In a reduced environment, indium iodide becomes important. Caesium iodide does certainly appear; at certain test conditions it becomes the major iodine transporting vehicle. But its role is not greater than that of the other iodides. It has more chances in FPT1 than in FPT0.

Chlorine, which was present in FPT0 as a contaminant in relatively large amounts, does not prevent the iodine from forming CsI. Both share the caesium like partners. The evolution of caesium chloride and iodide proceeds in parallel. When CsCl is abundant, CsI is strong, too.

Should boron be added to the steam injection, the effect on fission product chemistry would be minor. It would certainly not lead to more formation of the highly volatile iodine species HI and elemental iodine. All that boron can do is to shift some of the iodine from CsI onto the absorber materials.

The effect of boron on caesium redistribution is significant only at temperatures above 1000°C - which are virtually not determining for the final fission product speciation in tests with spent fuel pins - or in a strongly reduced atmosphere, which is not available for more than some five to 10 minutes during a test.

An important role in the formation of caesium species is played by molybdenum. For the results of code calculations it is therefore important what release rate is attributed to this fission product, especially if low values are involved.

Spaltproduktverhalten in den Phebus Versuchen FPT0 und FPT1 und der mögliche Einfluß des Bors

Die thermochemischen Rechnungen, die mit einem Großteil der Elemente, die sich im chemischen System eines Phebus FP-Versuchs befinden, durchgeführt wurden, erlauben folgende Schlußfolgerungen:

Die metallischen Absorbermaterialien, Silber, Indium und Kadmium, spielen eine beträchtliche Rolle in der Spaltproduktchemie. Bei über 1000°C ist Silberjodid der Hauptträger des Jods in Dampfatmosfera sowohl bei FPT0 wie auch bei FPT1. Bei tieferen Temperaturen übernimmt diese Rolle das Kadmiumjodid. Indiumjodid wird in einer reduzierten Atmosphäre dominant. Cäsiumjodid ist selbstverständlich auch vorhanden. Bei gewissen Versuchsbedingungen wird es zum vorherrschenden Jodträger. Im ganzen gesehen ist seine Rolle aber nicht bedeutender als die der anderen Jodide. In FPT1 hat es mehr Chancen als in FPT0.

Das Chlor, das in FPT0 in vergleichsweise großen Mengen als Verunreinigung vorhanden war, verhindert nicht das Jod daran, CsI zu bilden. Beide teilen sich das Cäsium wie Partner. Die Entwicklung von Cäsium-Chlorid und Jodid verläuft in FPT0 parallel. Dort wo CsCl reichlich vorhanden ist, ist auch CsI stark vertreten.

Sollte Bor dem Wasserdampf zugesetzt werden, würde die Spaltproduktchemie davon wenig beeinflußt werden. Vor allem würde es nicht zu einer Erhöhung der sehr flüchtigen Jodspezien Jodwasserstoff und elementares Jod kommen. In der Jodchemie bewirkt das Bor bestenfalls einer Verlagerung des Jods von CsI auf die Absorbermaterialien.

Der Einfluß des Bors auf die Cäsiumverteilung ist nur von Bedeutung bei Temperaturen über 1000°C - die für die Endverteilung im Fall von Versuchen mit abgebrannten Brennstäben eigentlich nicht ausschlaggebend sind - oder in stark reduzierter Atmosphäre, die nur während einiger Minuten eines Tests vorhanden ist.

Eine wichtige Rolle in der Cäsiumchemie spielt das Molybdän. Für die Rechenergebnisse ist daher von Bedeutung, welche Freisetzungsrates für dieses Spaltprodukt angenommen wird, vor allem, wenn es sich um niedere Werte handeln sollte.

Introduction and computational procedure

In a contribution to the 22nd TG Meeting at Brussels in March of 1995, which was later registered as TG 257, it was shown how the absorber materials silver and indium influence the iodine speciation in the gas stream of PHEBUS FP tests. The calculations for that presentation were done with a limited number of elements for the representation of the chemical system of such tests. Since the outcome of this thermochemical exercise was not in line with code calculations performed previously, in particular those with the code GEMINI, it appeared useful to do new calculations with a larger number of elements which represent more adequately the chemical system in PHEBUS FP tests. So, besides caesium and iodine, tellurium and molybdenum were added to represent the volatile fission products. Especially molybdenum is important since it has a high affinity for caesium when oxygen is available. Cadmium was added to complete the absorber material inventory. Also added was uranium for the fuel, zirconium and tin for the cladding material, and, finally, chlorine which is present in FP tests as a contaminant in the fuel and the shroud.

The volatile fission products rubidium, selenium and bromine were not separately included. The masses were added to their respective chemical homologues caesium, tellurium and iodine. It does not make sense to treat them separately since the data for rubidium compounds, e.g., are not as complete as those for caesium. Even though rubidium forms the same molybdates as caesium (see Appendix) there are no useful data available for rubidium molybdate (Rb_2MoO_4) in the open literature, whereas those for Cs_2MoO_4 have been known from the early days of reactor fuel research. Treating rubidium separately from caesium can thus easily lead to erroneous results. The surprising outcome of the GEMINI calculations presented by P. Dumaz at the 19th SAWG meeting at Ispra in July 1994 predicting RbI as the predominant iodine carrying species is very likely the consequence of neglecting the rubidium molybdate formation probability. One could, of course, fabricate one's own data for Rb_2MoO_4 . These should result in a stability for Rb_2MoO_4 not significantly different from that of Cs_2MoO_4 . This would not make RbI to be dominant over CsI since RbI is a little bit less stable than CsI . However, instead of searching for Rb-Mo-O data, it is easier and equally adequate to just add the rubidium masses to those of caesium.

As in the previous study (TG 257) the volatile fission products were assumed to be released 100% during a period of 6000 s. The release of silver was taken as 15%, that of indium as 30%, and that for cadmium as 50%. These release numbers lead

to the amount of absorber material given by the analysts as transported away from the test train in FPT0. The amount of fuel and zirconium was also taken from the FPT0 results. Tin was assumed to be released by about 80%. Chlorine, finally, was assumed as four times more in weight units than iodine in the FPT0 test. This number was deduced from the analysis of the sump water in FPT0. Which means that in FPT0 we have had almost 15 times more moles of chlorine than iodine (and about three times more than caesium).

The equilibrium calculations were performed with the latest version (1995) of the computer programme "Thermo Calc" developed by the Division of Physical Metallurgy, KTH, Stockholm, Sweden, which is equipped with the SGTE substance data base of 1994 (SGTE for Scientific Group Thermodata Europe). It does not contain data on the Rb-Mo-O system, which does not cause a problem. However, there are also no data available on gaseous caesium telluride, which, according to our experience in fast reactor fuel testing, must exist, and which could have an influence on fission product speciation. As in other data listings, only values for condensed Cs_2Te are available here, too.

The results of the calculations are given in diagrams and tables. The diagrams cover a temperature range from the melting point of UO_2 down to that of Point C in the circuit (700°C). The injected steam flow rate was in all cases 3 g/s. Examples for a full steam atmosphere as well as for a 90% steam reduction are given for FPT0 (Figs. 1 and 2) and FPT1 (Figs 3 and 4). The FPT1 example is also shown in case 1000 ppm boron is added to the injected steam (Figs. 5 and 6). The diagrams give values for the mass flow in mole/s by the scale of the left hand side and for the partial pressures of the species by the scale on the right hand side. Moles of the species in the diagrams mean true moles in the chemical sense and not "computer moles". In comparing the mass flow curves of the different caesium species to assess their caesium carrying capacity, one has to bear in mind that one mole of Cs_2MoO_4 carries twice as much caesium as the other caesium species shown in the diagrams. The representation of true moles was chosen to keep a direct relationship between mass flow and partial pressure. The same holds for elemental tellurium. The curve marked "Te" represents the total mass of all "non-aligned" tellurium species, i.e., atomic and molecular Te as well as the tellurium hydrides and oxides. The direct correlation between mass flow and partial pressure does no more exist when condensation takes place. In such an event, the partial pressure is the vapour pressure of the respective substance. Involved in such an evolution above 700°C were the species Cs_2MoO_4 , Ag_2Te and CsBO_2 to various

degrees depending on test conditions. The traces of their vapour pressures were not outlined in all diagrams just to avoid confusion of lines in a crowded place.

The tables list the redistribution of the three volatile fission products caesium, iodine, and tellurium over the possible species contributing to their transport. The numbers given are in percent of the total amount of the respective fission product element (including the chemical homologue) present in the test. The tables include, besides the results for conditions of full steam and 90 % reduction, also those for 50% steam reduction. The temperatures covered are those believed to be representative for the final speciation. For the FPT0 test, this temperature is considered to be around 1000°C, So, we find tables for equilibrium species at 1200 K and 1300 K (Tables 1 and 2).

In FPT1, the reaction velocities should be higher than those in FPT0 by two to three orders of magnitude. The temperature representative of the final speciation is therefore lower in FPT1 than in FPT0. It could be 700°C, which is the temperature at Point C (see Table 3). Tables with still lower temperatures are given to show the direction which reactions will be taking at those temperatures (Tables 4,5 and 6).

In considering the possible effect of reaction rates on the final speciation one has to be aware of the fact that it takes the fission products and the other materials from their release in the test train to the outlet of the circuit not much more than one second (unless they get trapped or settle out along their path).

Reactions with the piping surface of the circuit were also considered. Such reactions do not interfere in fission product speciation. Neither Cs_2MoO_4 nor CsI nor $\text{Cs}_2\text{U}_4\text{O}_{12}$ are being decomposed in favour of caesium chromate formation. Four chromate species, including Cs_2CrO_2 , were considered. And for the FeI_2 , there is no chance to be formed even in minor amounts in any Phebus atmosphere at circuit temperatures. Anyway, there is sufficient Fe, Cr and Ni carried along in the gas stream ready to react with the fission products should this be possible. There is no need to wait for the piping surfaces to provide this opportunity.

Discussion and conclusions

The diagrams and tables are the results of equilibrium calculations and, thus, give the speciations under the assumption that equilibrium among the species in the gas stream is attained. This will likely be the case at high temperatures ($T \geq 1000^\circ\text{C}$) but not necessarily at lower temperatures, especially if the partial pressures of the species (i.e., their concentrations in the gas stream) are low. The diagrams show

the evolution of the equilibria in going from high to lower temperatures (as the gas stream does). So, everybody may select the equilibrium situation he thinks will remain more or less unchanged in going to lower temperatures. And anybody who wishes to know what the situation will be below 700°C, in case reactions are still going on, is referred to the respective tables.

From the results, as they are outlined in the diagrams and tables, the following main conclusions can be drawn:

The metallic absorber materials play a significant role in fission product speciation. At around 1000°C and above, silver iodide is the dominant iodine carrier in a full steam atmosphere in both FPT0 and FPT1. At lower temperatures, cadmium iodide takes over. In a reduced environment, indium iodide becomes important. Caesium iodide does certainly appear as iodine transporting vehicle. In some test conditions it becomes the major iodine carrier. But its role is not greater than that of the other iodides, like AgI, InI and CdI. It has more chances to appear in FPT1 than in FPT0. One reason for this is also the effect of chlorine, which is more important in FPT0 than in FPT1. Chlorine, however, does not prevent the iodine from forming CsI. Both share the caesium like partners, at least at higher temperatures ($T \geq 700^\circ\text{C}$). The evolution of caesium chloride and iodide in FPT0 proceeds in parallel. For conditions that favour CsCl formation, CsI is strong, too. Until, at lower temperatures, CsCl condenses. Then, of course, all the available caesium is used up for CsCl, and CsI practically disappears, should equilibrium be reached. Tables 7 through 11 were produced just to show the effect of chlorine in FPT0. The numbers on the left hand side are those for an FPT0 test without any chlorine present. The right hand numbers are those for the "real" FPT0 test with chlorine contamination. It appears as if chlorine affects rather caesium than iodine speciation.

The effect of boron on fission product speciation in general and on that of iodine in particular is surprisingly small, according to these calculations. Its effect is most perceivable in reduced steam and at temperatures above 700°C, i.e., in the test circuit before Point C. And there it affects more the caesium than the iodine. At temperatures lower than 700°C, a boron effect is practically non-existent for any atmosphere condition, even if there is only little molybdenum available. There is absolute no sound foundation for the contention that boron does increase the presence of the highly volatile iodine species, viz., hydrogen iodide and elemental iodine. All that boron can do is to shift some of the iodine from CsI onto the absorber material iodides.

An increase in boron addition over the 1000 ppm to 2000 or even 3000 ppm would not change anything in this situation. The bulk of the boron is carried away by boric acid and other boron hydroxides. The highest boron potential in the chemical system of a Phebus FP test would be that of boron oxide (B_2O_3) as long as a trace of oxygen were still present. And even this could only be attained in a reduced atmosphere. More boron addition to the steam would not and could not raise the boron potential above that of B_2O_3 . And no matter how much boron oxide or other boron material condensed on the wall of the circuit, it would not influence the iodine speciation more than described in this report.

The most important caesium carrier is Cs_2MoO_4 . It shows its strength especially at lower temperatures. It gets weakened with steam reduction. For a full steam atmosphere, it is dominant from temperatures of well above $1000^\circ C$ downwards. In largely reduced atmospheres, its dominance starts at somewhat above $700^\circ C$ (Point C). It is the caesium molybdate formation that would prevent the boron from playing a noticeable role in fission product speciation. The importance of Cs_2MoO_4 was reduced in FPT0 also by the presence of chlorine.

The tellurium carrier the most likely to appear in FPT tests is silver telluride ($AgTe$ or Ag_2Te). It is the dominant tellurium species in a full steam atmosphere and at temperatures above $700^\circ C$. Only in a reduced atmosphere does tin telluride ($SnTe$) prevail. Indium and cadmium telluride get their chances at lower temperatures (see Table 4).

Finally, Table 12 lists for various test conditions the calculated condensation temperatures of fission product species. As pointed out in the former report, these are no fixed values. They change with conditions that change the partial pressures of the species: they decrease with increasing steam flow rate, and increase with increasing system pressure. It becomes evident from Table 12 that iodine-containing species had hardly any chance to condense at temperatures around $700^\circ C$ in FPT0 and, hence, could not appear "predominantly in aerosol form at Point C", as was predicted for FPT0. They will not be doing so in FPT1, either. The boron species in the two lowest lines in Table 12 were included (even though they are evidently no fission product species) just to show how little boron condensation takes place and that a high B_2O_3 -activity is reached only for strongly reduced atmospheres.

Additional remarks

The presence of molybdenum in the gas stream of a Phebus test plays an important role in fission product speciation. It may therefore appear that its release rate chosen as input for the calculations has a significant effect on the results. This effect is indeed important up to a value of about 40 % of total release. For higher values up to the 100% used in the calculations here, the actual value of the Mo-fraction is not important. The effect is much less visible in reduced atmospheres. Lower values than 30 %, however, bring about new caesium containing species. In a steam atmosphere, it is caesium uranate that shows up as $Cs_2U_4O_{12}$ (Table 13). In a reduced atmosphere it is Cs_2Te that settles out (Table 14). For an exceptionally low fission product inventory, like in FPT0, the effect of the Mo-fraction on speciation is perhaps more important. This is not so evident at higher temperatures (Table 15), but at lower temperatures (Table 16), especially if chlorine contamination is not taken into account.

The release of molybdenum was chosen in these calculations as equal to that of the volatile fission products, because in a steam atmosphere the total pressure of molybdenum bearing species in equilibrium with an irradiated fuel can reach a partial pressure that ensures the total release of molybdenum if the fuel is at a temperature of 1000°C or more. In a reduced atmosphere, we get theoretically less release and the fuel temperature would therefore have to be higher to fulfill the input conditions for these calculations. But, as said above, in a reduced atmosphere, the chosen release value for molybdenum could even be lower than 40 % without significant consequences for the results shown in this report.

The molybdenum is often, and this is probably an error, not regarded as a fission product easy to be released. In FPT0, though, it showed up as the only fission product element (except for zirconium, which does not count) in sufficient amounts to be detected in PTA with analytical methods other than γ -spectroscopy. The deposits in the filters and impactors at Points C and G in the circuit contained about 1 wt % Mo and those at Point H in the containment about 2 wt % Mo (see C. Pagano, B. Rabu, Note Technique DTP/SECC no 95.037A). According to the Data Book, there was no molybdenum as structural material available in FPT0. However, it cannot be excluded that some of the austenitic materials used in the test section (in spacer grids, thermocouple claddings) contained some molybdenum. But if it did, it was very little. And the release from the metallic or ceramic melt or crust was certainly not easier than from the hot, overstoichiometric fuel.

Considering the effect of boron on fission product speciation and its counterbalance by molybdenum, it was argued that there exist other borates that have a higher gettering capability for caesium than CsBO_2 , the only caesium borate for which adequate thermochemical data are available and which, therefore, was the only one considered in the calculations. Hence, if the other (poly-)borates had been taken into account, a true effect of boron would have probably become visible. This assumption is pure speculation and not necessarily true. From the phase diagrams of the Cs-B-O system shown in the Appendix, and assuming that CsBO_2 is the ternary compound in the system with the lowest level for the integral free enthalpy (a plausible assumption) we can deduce that the polyborates need, for their formation, considerable higher chemical activities of B_2O_3 than what CsBO_2 requires. A high B_2O_3 availability or chemical activity is hard to get in a steam atmosphere due to the competition by the boric acids. This competition becomes stronger with decreasing temperature and makes that in the temperature range where the polyborates appear as separate phases no borate with a higher boron content than in triborate can be formed, even though, theoretically, there is sufficient material available to form the nanoborate, if a 1000 ppm boron addition to the steam injection is adopted as test procedure.

In the Cs-Mo-O system, there are also six ternary compounds (see Appendix). Due to the limited fission Mo-inventory, the highest molybdate that can be formed in a FP test is the trimolybdate. But, contrary to the conditions for the borates, the accompanying Mo-species in the gas stream (as long as it is mainly composed of steam) do not compete with but support the caesium molybdate formation, with an increasing support in going to lower temperatures. So what we would get by employing a more complete set of thermochemical data is, instead of only the competition between the monoborate and the monomolybdate of caesium, also, at lower temperatures, a competition between the respective di- and tricompounds. It is pretty unlikely that this would change much in the outcome of the calculations. In particular, since the big drawback for boron in the competition with molybdenum for the caesium is, and this may sound paradoxical, the relative high volatility of CsBO_2 . The vapour pressure of CsBO_2 is several orders of magnitude higher than that of Cs_2MoO_4 in the temperature range of interest. This means, the molybdate needs a much lower partial pressure than the borate to condense. But once condensation takes place, the respective species acts as a getter for its constituents and competing gaseous products have little chances to carry much material. So, in the gas stream of an FP test on its way to lower temperatures, the molybdate gathers practically all the caesium as soon as its condensation (or saturation) temperature is reached and enough molybdenum is available (for which

less than 40 % of the Mo-inventory suffice). The possibility of polyborate formation (or its consideration in the calculations) would not change anything since nothing would be changed with respect to the partial pressure of CsBO_2 nor its saturation temperature.

The chances for a high B_2O_3 availability and thus for polyborate formation increase, however, with the degree of steam reduction. But it needs considerable reduction ($p_{\text{H}_2} > 50\%$) to get a sufficiently high B_2O_3 -activity to form polyborates with low caesium content. It must be remembered, though, that a reduced atmosphere, which not only favours the borates but also reduces the molybdate formation, exists only during some 10 to 15 min, i.e. about 10 % of the test time when fission product release takes place.

So, except for this short event, we can forget about the polyborates. And if they actually appeared during the major part of the test period, what could they effect? They would not change much in the iodine speciation. A caesium potential reduced to somewhat below the level in equilibrium with molybdenum could not liberate much iodine. At most there would be a shift from the CsI onto the absorber material iodides. And concerning the Cs-species, there could be some more condensed caesium borate instead of molybdate shown for the boron cases in the diagrams and tables. This would, however, not alter much in the already low availability of CsOH in the circuit.

There are some uncertainties about the influence the thermocouple materials could have on the fission product chemistry. It concerns the rhenium and the tungsten. We have had roughly 700 g Re and about 30 g of W in FPT0, and we will have about the same amount of these elements in FPT1. Most of it will be carried away; this is certainly true for the tungsten. It needs high temperatures to vaporize the rhenium even in a steam atmosphere. However, the oxides formed will practically all be decomposed before they enter the circuit at temperatures below 1000°C (including the Re_2O_7). At point C at 700°C , practically all the rhenium will be carried as solid metallic material. The little hydrogen generated even in a full steam atmosphere at high temperature by thermal decomposition and the reactions with the thermocouple materials, but carried along in the gas stream, claims back the oxygen from the Re-oxides at lower temperatures.

It is different with the tungsten. The tungsten oxides may decompose but the tungsten hydroxide (WOH) survives in the gas stream down to the lowest temperatures in the circuit even in a strongly reduced (90 % H_2) atmosphere.

Even though both rhenium and tungsten can form complex compounds with caesium and oxygen, it is difficult to say whether one of them is capable of interfering in fission product chemistry. Data on CsReO_4 and Cs_2WO_4 are totally lacking. The chemical similarity of W and Mo could suggest a stability for Cs_2WO_4 not far different from that of Cs_2MoO_4 . However, the formation of Cs_2WO_4 would need the presence of some WO_3 , which is not in sight in a steam environment. The WOH consumes all the tungsten. The CsReO_4 could have more chances in this respect. There are no known data for a gaseous rhenium hydroxide which could lower the availability of Re_2O_7 in a steam environment. But the rhenium behaves more nobly than tungsten. So it appears unlikely that either Cs_2WO_4 or CsReO_4 will be formed.

The conclusion that reactions with the piping surfaces do not interfere in fission product speciation, may be hard to digest for some people. Equilibrium calculations with the Phebus atmosphere and the material carried along in the gas stream (notably Mo, U, Zr as oxidised species) do not predict the formation of compounds between fission products and piping material components. And anyone who believes that thermodynamics are not predicting the real situation or that the thermodynamics applied here were incomplete because "experimental evidence" (but under what conditions?) proves the contrary, is referred to the FPT0 results that show that sufficient Cr-Fe-Ni material is carried along in the gas stream to react with all the possible fission products, if such reactions could take place. It is hard to see why the fission products should not react with these materials in the gas stream but wait to get a chance to react with the same materials on the piping surfaces. So it is very unlikely that noticeable masses of fission products will react with the piping surfaces of the circuit.

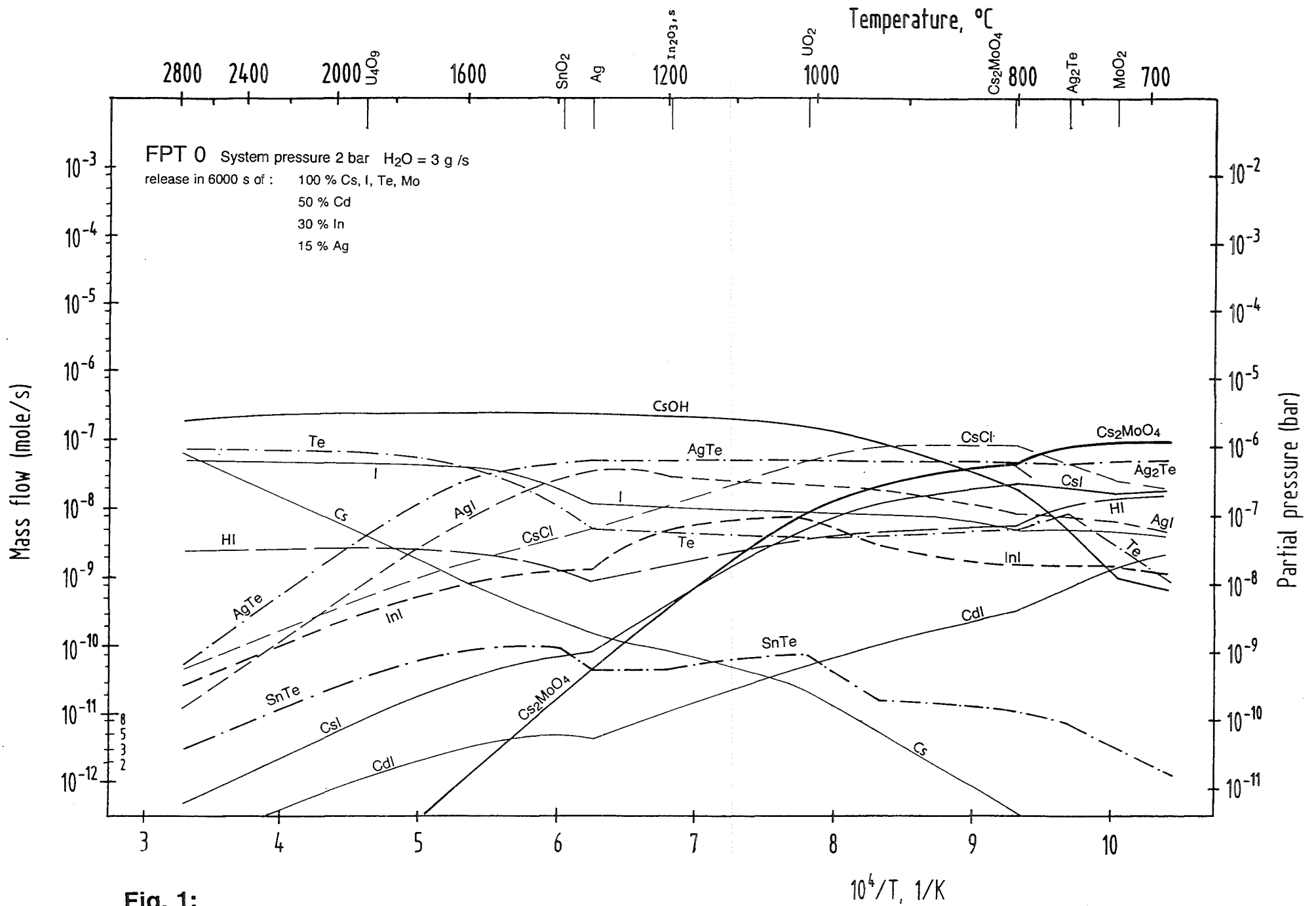


Fig. 1:

Temperature, °C

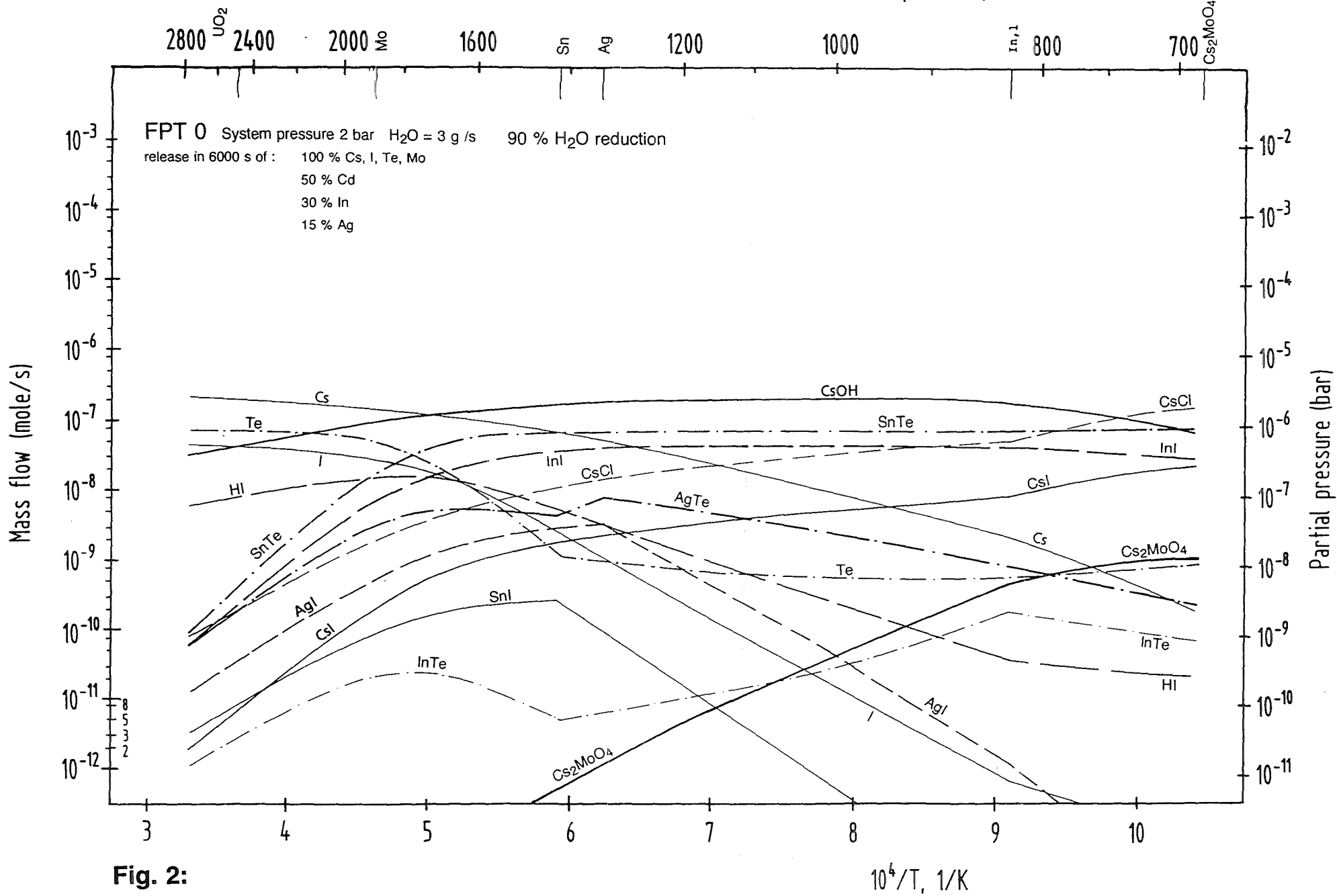


Fig. 2:

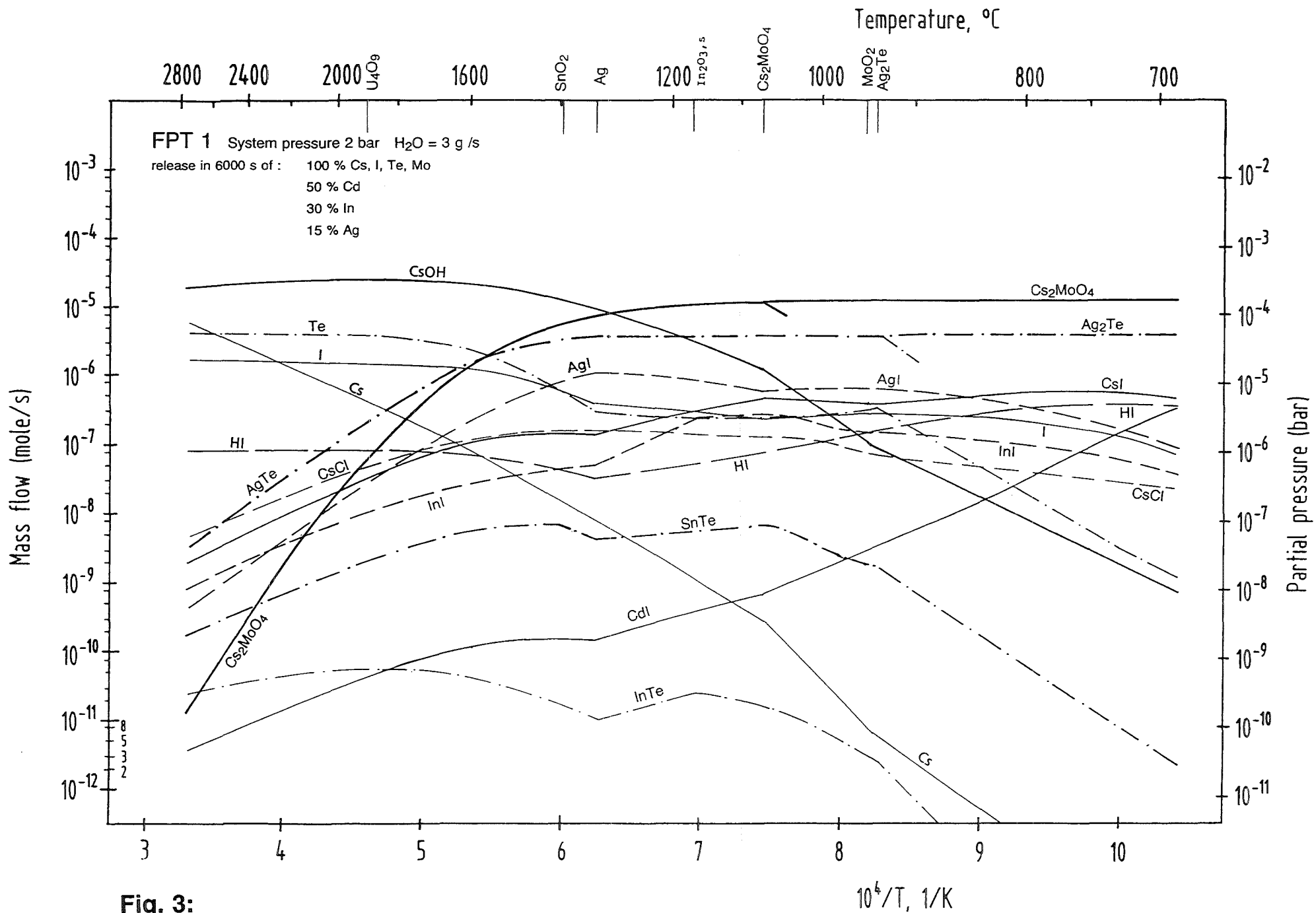


Fig. 3:

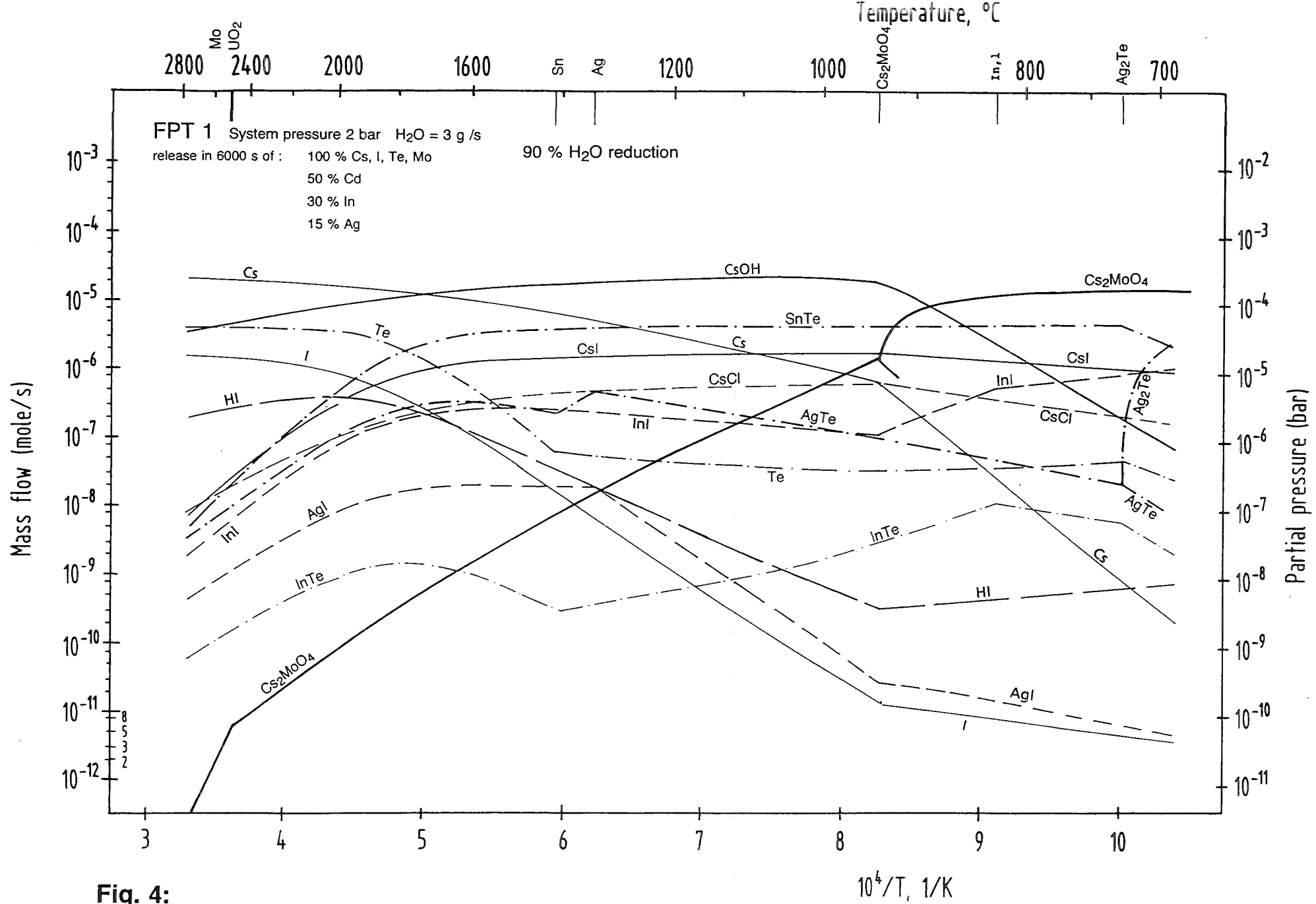


Fig. 4:

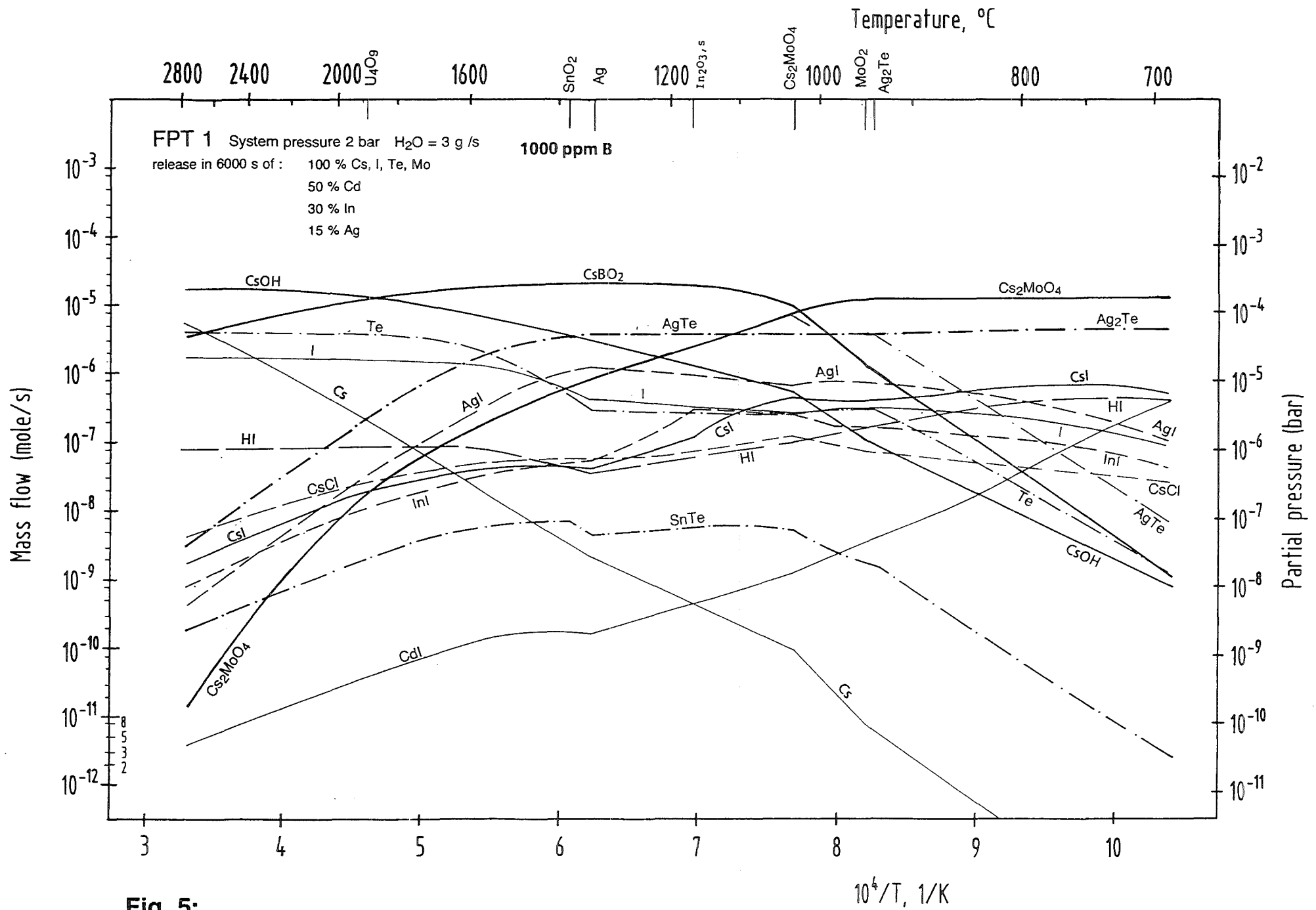


Fig. 5:

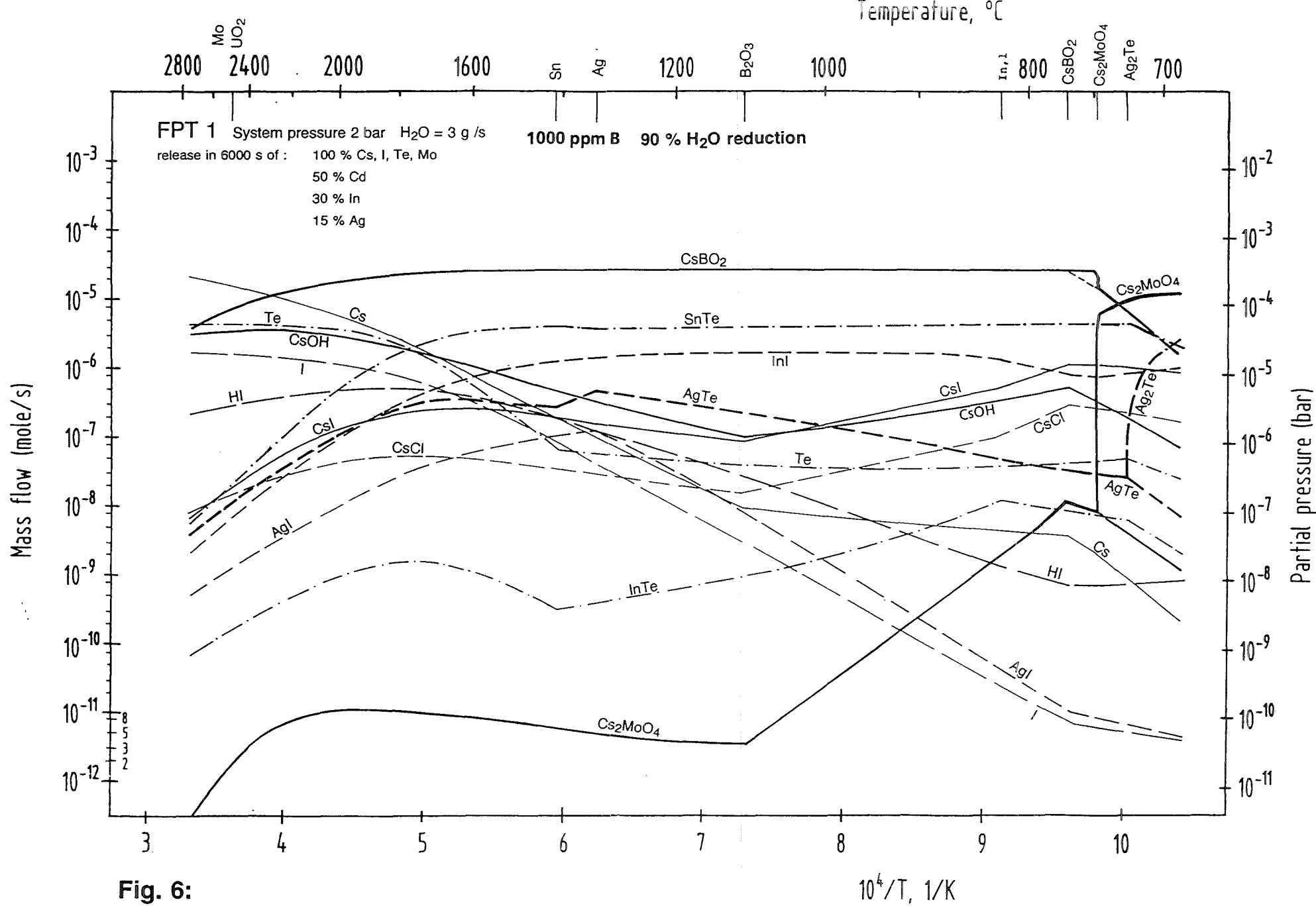


Fig. 6:

Table 1: Distribution of Volatile Fission Products over Species in the Gas Stream at 1300 K / 1027°C in %

		FPT 0			FPT 1			with Boron		
		H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂
CS	CS2MOO4	3.11	1.36	0.02	95.15	57.51	1.62	57.67	0.12	-
	CSOH	72.90	83.29	79.79	2.43	34.67	83.00	2.23	2.39	0.52
	CSI	1.96	1.98	2.08	1.89	5.76	6.47	1.78	1.72	0.51
	CSCL	18.92	11.23	13.27	0.53	1.75	2.30	0.49	0.31	0.09
	CS	0.01	0.75	4.83	-	0.31	5.00	-	0.02	0.03
	CSBO2	-	-	-	-	-	-	37.82	95.32	98.85
I	CSI	9.16	9.26	9.72	26.93	82.06	92.27	25.33	24.53	7.29
	AGI	48.3	0.89	0.15	37.65	0.18	0.01	38.47	0.76	0.15
	INI	16.25	86.25	89.37	14.65	17.05	7.65	14.96	71.64	91.74
	CDI	0.1	-	-	0.08	-	-	0.08	-	-
	HI+I	26.18	3.58	0.76	20.69	0.71	0.06	21.14	3.05	0.81
TE	AGTE	93.79	4.02	4.01	92.59	4.02	4.01	92.59	4.02	4.01
	SNTE	0.12	95.29	95.08	0.14	95.30	95.08	0.14	95.3	95.08
	INTE	-	-	0.04	-	-	0.04	-	-	0.03
	CDTE	-	-	-	-	-	-	-	-	-
	TE	6.09	0.67	0.87	7.27	0.68	0.87	7.27	0.68	0.87

Table 2: Distribution of Volatile Fission Products over Species in the Gas Stream at 1200 K / 927°C in %

	FPT 0			FPT 1			with Boron			
	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	
CS	CS2MOO4	20.55	4.86	0.01	97.72	84.5	19.86	93.96	1.07	-
	CSOH	39.82	77.64	78.28	0.34	9.78	69.17	0.34	3.51	0.78
	CSI	5.10	2.52	2.68	1.65	4.54	6.53	1.65	2.80	0.96
	CSCL	34.53	14.66	16.62	0.29	1.14	2.33	0.29	0.56	0.17
	CS	-	0.35	1.63	-	0.04	2.08	-	0.02	0.02
	CSBO2	-	-	-	-	-	-	3.77	92.06	98.08
I	CSI	23.81	11.78	12.52	23.53	64.65	93.08	23.50	39.96	13.74
	AGI	41.34	0.16	0.02	39.06	0.06	-	39.11	0.11	0.03
	INI	6.60	86.78	87.21	9.34	34.74	6.88	9.31	59.03	85.94
	CDI	0.25	-	-	0.26	-	-	0.28	0.01	-
	HI+I	27.98	1.31	0.27	27.77	0.53	0.02	27.80	0.91	0.28
TE	AGTE	93.67	2.40	2.39	89.31	2.40	2.39	89.36	2.40	2.39
	SNTE	0.03	97.04	96.75	0.04	97.00	96.71	0.04	97.00	96.70
	INTE	-	0.01	0.01	-	0.01	0.01	-	-	0.01
	CDTE	-	-	-	-	-	-	-	-	-
	TE	6.30	0.58	0.81	10.64	0.59	0.82	10.64	0.60	0.82

Table 3: Distribution of Volatile Fission Products over Species in the Gas Stream at 973 K / 700°C Point C in %

		FPT 0			FPT 1			with Boron		
		H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂
CS	CS2MOO4	81.47	57.56	0.46	97.70	98.25	95.51	97.69	97.64	85.58
	CSOH	0.42	15.46	31.63	-	0.14	0.39	-	0.14	0.39
	CSI	8.15	4.25	8.94	2.18	1.40	3.43	2.18	1.39	3.46
	CSCL	9.96	22.72	58.38	0.11	0.21	0.67	0.11	0.21	0.68
	CS	-	0.01	-	-	-	-	-	-	-
	CSBO2	-	-	-	-	-	-	0.01	0.61	9.87
I	CSI	38.07	19.89	41.80	31.13	19.89	48.86	31.14	19.86	49.35
	AGI	11.89	-	-	7.15	-	-	7.13	-	-
	INI	2.97	79.87	58.15	2.83	79.87	51.10	2.92	79.89	50.63
	CDI	5.23	-	-	30.41	-	-	30.41	0.08	-
	HI+I	41.83	0.23	0.05	28.37	0.23	0.04	28.35	0.23	0.04
TE	AGTE	96.05	0.41	0.40	99.93	39.23	38.88	99.93	39.26	38.9
	SNTE	3	98.85	98.28	-	60.27	60.27	-	60.24	60.25
	INTE	-	0.01	0.12	-	-	0.07	-	-	0.07
	CDTE	-	-	-	-	-	-	-	-	-
	TE	3.95	0.74	1.20	0.07	0.50	0.78	0.07	0.5	0.78

Table 4: Distribution of Volatile Fission Products over Species in the Gas Stream at 800 K / 527°C in %

	FPT 0			FPT 1			with Boron			
	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	
CS	CS2MOO4	96.15	38.13	66.45	99.89	94.89	97.73	99.86	94.96	97.73
	CSOH	-	0.01	0.14	-	-	-	-	-	-
	CSI	1.45	11.32	5.94	0.12	4.57	2.02	0.12	4.57	2.02
	CSCL	2.42	50.44	27.47	-	0.47	0.25	0.03	0.47	0.25
	CS	-	-	-	-	-	-	-	-	-
	CSBO2	-	-	-	-	-	-	-	-	-
I	CSI	6.73	52.91	27.75	1.67	65.16	28.74	1.67	65.15	28.63
	AGI	0.21	-	-	0.04	-	-	0.04	-	-
	INI	0.12	45.84	72.14	0.03	33.90	71.16	0.03	33.96	71.29
	CDI	81.40	0.02	-	95.82	0.02	-	95.82	0.03	-
	HI+I	11.54	1.22	0.10	2.44	0.90	0.10	2.44	0.90	0.08
TE	AGTE	99.99	0.02	0.01	100	-	-	100	-	-
	SNTE	-	21.09	25.25	-	0.45	0.44	-	0.45	0.43
	INTE	-	-	23.77	-	-	99.54	-	-	99.55
	CDTE	-	78.24	-	-	99.54	-	-	99.53	-
	TE	0.01	0.66	0.96	-	-	0.02	-	0.01	0.02

Table 5: Distribution of Volatile Fission Products over Species in the Gas Stream at 600 K / 327°C in %

	FPT 0			FPT 1			with Boron			
	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	
CS	CS2MOO4	-	-	-	90.75	90.25	90.25	90.74	90.25	90.24
	CSOH	-	-	-	-	-	-	-	-	-
	CSI	-	0.01	0.01	6.75	7.02	7.02	6.75	7.02	7.02
	CSCL	100	99.99	99.99	2.5	2.75	2.75	2.5	2.74	2.74
	CS	-	-	-	-	-	-	-	-	-
	CSBO2	-	-	-	-	-	-	-	-	-
I	CSI	0.01	0.03	0.05	96.21	100	100	96.22	100	100
	AGI	-	-	-	-	-	-	-	-	-
	INI	-	0.82	19.83	-	-	-	-	-	-
	CDI	99.34	97.34	77.95	3.77	-	-	3.76	-	-
	HI+I	0.65	1.82	2.18	0.02	-	-	0.13	-	-
TE	AGTE	-	-	-	-	-	-	-	-	-
	SNTE	-	-	-	-	-	-	-	-	-
	INTE	-	-	-	-	-	-	-	-	-
	CDTE	100	100	100	100	100	100	100	100	100
	TE	-	-	-	-	-	-	-	-	-

Table 6: Distribution of Volatile Fission Products over Species in the Gas Stream at 450 K / 177°C in %

	FPT 0			FPT 1			with Boron			
	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂	
CS	CS2MOO4	-	-	-	90.24	90.24	90.24	90.24	90.24	90.24
	CSOH	-	-	-	-	-	-	-	-	-
	CSI	-	-	-	7.02	7.02	7.02	7.02	7.02	7.02
	CSCL	100	100	100	2.74	2.74	2.74	2.74	2.74	2.74
	CS	-	-	-	-	-	-	-	-	-
	CSBO2	-	-	-	-	-	-	-	-	-
I	CSI	-	-	-	100	100	100	100	100	100
	AGI	-	-	-	-	-	-	-	-	-
	INI	-	-	-	-	-	-	-	-	-
	CDI	100	99.9	99.99	-	-	-	-	-	-
	HI+I	-	0.01	0.01	-	-	-	-	-	-
TE	AGTE	-	-	-	-	-	-	-	-	-
	SNTE	-	-	-	-	-	-	-	-	-
	INTE	-	-	-	-	-	-	-	-	-
	CDTE	100	100	100	100	100	100	100	100	100
	TE	-	-	-	-	-	-	-	-	-

Table 7: Effect of Chlorine on Fission Product Speciation in FPTO at 1300 K / 1027°C

	FPTO CL absent		FPTO		
	H ₂ O	90 % H ₂	H ₂ O	90 % H ₂	
CS	CS ₂ MOO ₄	9.07	0.02	3.11	0.02
	CSOH	88.58	92.05	72.90	79.79
	CSI	2.34	2.33	1.96	2.08
	CSCL	-	-	18.92	13.27
	CS	0.02	5.57	0.01	4.83
I	CSI	10.92	10.90	9.16	9.72
	AGI	47.31	0.14	48.30	0.15
	INI	16.00	88.12	16.25	89.37
	CDI	0.10	-	0.10	-
	HI+I	25.66	0.74	26.18	0.76

Table 8: Effect of Chlorine on Fission Product Speciation in FPTO at 1200 K / 927°C

	FPTO CL absent		FPTO		
	H ₂ O	90 % H ₂	H ₂ O	90 % H ₂	
CS	CS ₂ MOO ₄	37.64	0.14	20.55	0.01
	CSOH	55.84	93.92	39.82	78.28
	CSI	6.52	3.10	5.10	2.68
	CSCL	-	-	34.53	16.62
	CS	-	2.83	-	1.63
I	CSI	30.47	14.47	23.81	12.52
	AGI	37.73	0.02	41.34	0.02
	INI	6.02	85.28	6.60	87.21
	CDI	0.23	-	0.25	-
	HI+I	25.54	0.26	27.98	0.27

Table 9: Effect of Chlorine on Fission Product Speciation in FPT0 at 973 K/ 700°C Point C

		FPT0 CL absent		FPT0	
		H ₂ O	90 % H ₂	H ₂ O	90 % H ₂
CS	CS2MOO4	91.41	47.28	81.47	0.46
	CSOH	0.43	42.10	0.42	31.63
	CSI	8.16	10.46	8.15	8.94
	CSCL	-	-	9.96	58.38
	CS	-	0.15	-	-
I	CSI	38.16	48.88	38.07	41.80
	AGI	11.85	-	11.89	-
	INI	3.00	51.08	2.97	58.15
	CDI	5.21	-	5.23	-
	HI+I	41.78	0.04	41.83	0.05

Table 10: Effect of Chlorine on Fission Product Speciation in FPT0 at 800 K /527°C

		FPT0 CL absent		FPT0	
		H ₂ O	90 % H ₂	H ₂ O	90 % H ₂
CS	CS2MOO4	98.55	93.92	96.15	66.45
	CSOH	-	0.14	-	0.14
	CSI	1.45	5.94	1.45	5.94
	CSCL	-	-	2.42	27.47
	CS	-	-	-	-
I	CSI	6.75	27.75	6.73	27.75
	AGI	0.21	-	0.21	-
	INI	0.12	72.14	0.12	72.14
	CDI	81.35	-	81.40	-
	HI+I	11.56	0.10	11.54	0.10

**Table 11: Effect of Chlorine on Fission Product Speciation
in FPTO at 600 K/ 327°C**

		FPTO CL absent		FPTO	
		H ₂ O	90 % H ₂	H ₂ O	90 % H ₂
CS	CS ₂ MOO ₄	99.98	78.61	-	-
	CSOH	-	-	-	-
	CSI	0.02	21.39	-	0.01
	CSCL	-	-	100	99.99
	CS	-	-	-	-
I	CSI	0.08	99.98	0.01	0.05
	AGI	-	-	-	-
	INI	-	0.02	-	19.83
	CDI	99.27	-	99.34	77.95
	HI+I	0.65	-	0.65	2.18

Condensation Temperatures for Fission Product Species in Phebus Tests [°C]

	FPT 0				FPT 1						
	1.5 g/s	3g / s			1.5 g/s	3g / s			with Boron		
	H ₂ O	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	H ₂ O	50 % H ₂	90 % H ₂	H ₂ O	50 % H ₂	90 % H ₂
CS2Mo04	827	801	764	683	1115	1066	1032	934	1031	878	745
CSI	-	-	425	-	400	377	533	518	377	533	518
CSCL	415	385	492	485	418	384	492	485	388	493	485
AG2TE	784	759	558	-	971	936	723	723	936	723	723
INTE	-	-	-	578	-	-	-	658	-	-	658
CDTE	548	523	533	522	548	524	533	522	524	533	522
CSBO2	-	-	-	-	-	-	-	-	-	-	767
B2O3	-	-	-	-	-	-	-	-	-	-	1094
HBO2	-	-	-	-	-	-	-	-	238	282	316

**Table 13: Redistribution of volatile F. P. over species in FPT1 at 973K/700°C,
Effect of Mo-fraction in steam atmosphere**

	Mo-release %					
		20	30	35	40	100
Cs	Cs ₂ MoO ₄	55.06	82.59	96.19	97.70	97.70
	CsOH	17.60	7.70	0.01	0.005	0.005
	CsI	7.01	7.01	3.58	2.19	2.19
	CsCl	2.72	2.70	0.21	0.11	0.11
	Cs	-	-	-	-	-
	Cs ₂ U ₄ O ₁₂	17.6	-	-	-	-
I	CsI	99.97	99.93	51.03	31.15	31.15
	AgI	0.01	0.01	5.58	7.15	7.15
	InI	0.002	0.01	2.25	2.92	2.92
	CdI	0.004	0.004	19.00	30.41	30.41
	HI+I	0.02	0.05	22.14	28.37	28.37

**Table 14: Redistribution of volatile F.P. over species in FPT1 at 973 K / 700 °C,
Effect of Mo-fraction in reduced atmosphere**

	Mo-release %	50 % H ₂			90 % H ₂					
		20	40	100	20	30	35	40	60	100
Cs	Cs ₂ MoO ₄	55.06	98.25	98.25	55.06	82.59	95.47	95.47	95.47	95.47
	CsOH	35.42	0.14	0.14	25.12	8.29	0.40	0.40	0.40	0.40
	CsI	6.89	1.40	1.40	6.90	6.69	3.45	3.45	3.45	3.45
	CsCl	2.60	0.21	0.21	2.62	2.40	0.68	0.68	0.68	0.68
	Cs	0.02	-	-	0.09	0.03	0.001	0.001	0.001	0.001
	Cs ₂ Te	-	-	-	10.21	-	-	-	-	-
I Cd HI+	CsI	98.26	19.88	19.88	98.38	95.32	49.18	49.17	49.12	49.04
	AgI	-	0.002	0.002	-	-	-	-	-	-
	InI	1.72	79.90	79.90	1.62	4.68	50.78	50.79	50.84	50.92
	CdI	-	0.001	0.001	-	-	-	-	-	-
	HI+I	0.005	0.23	0.23	0.001	0.004	0.04	0.04	0.04	0.04

**Table 15: Redistribution of volatile F.P. over species in FPTO at 1200 K / 927°C ,
Effect of Mo-fraction in steam atmosphere**

		20	30	35	40	60	100
CS	Cs ₂ MoO ₄	5.74	8.19	9.36	10.41	14.31	20.55
	CsOH	47.87	46.53	45.89	45.31	43.19	39.83
	CsI	5.84	5.72	5.67	5.61	5.42	5.10
	CsCl	40.55	39.57	39.09	38.67	37.08	34.53
	Cs	0.003	0.003	0.003	0.003	0.003	0.003
I	CsI	27.31	26.75	26.48	26.24	25.32	23.82
	AgI	39.44	39.75	39.89	40.03	40.52	41.34
	InI	6.29	6.34	6.37	6.38	6.47	6.60
	CdI	0.24	0.24	0.25	0.25	0.25	0.25
	HI+I	26.72	26.72	27.02	27.11	27.45	28.00

**Table 16: Redistribution of volatile F.P. over species in FPTO at 973 K / 700 °C;
Effect of Mo-fraction in steam atmosphere**

Mo-release %		Cl included						no Cl		
		20	30	35	40	60	100	20	40	100
Cs	CsMoO ₄	47.43	63.25	66.75	71.87	79.66	81.47	54.15	86.70	91.40
	CsOH	1.66	1.02	0.84	0.72	0.48	0.42	25.00	0.90	0.45
	CsI	15.22	12.83	11.77	10.94	8.75	8.15	20.85	12.35	8.15
	CsCl	35.70	22.90	19.00	16.46	11.12	9.96	-	-	-
	Cs	-	-	-	-	-	-	-	-	-
I	CsI	71.15	59.95	55.00	51.15	40.85	38.10	97.40	57.70	38.15
	AgI	5.65	7.80	8.70	9.45	11.38	11.90	0.50	8.20	11.85
	InI	1.40	1.94	2.18	2.36	2.85	2.95	0.13	2.10	3.00
	CdI	1.95	2.94	3.42	3.80	4.92	5.23	0.14	3.15	5.26
	HI+I	19.86	27.37	30.70	33.25	40.00	41.82	1.80	28.80	41.80

**Table 17: Redistribution of volatile F. P. over species in FPT1 at 973 K/700°C,
Effect of Mo-fraction in steam atmosphere with Boron addition**

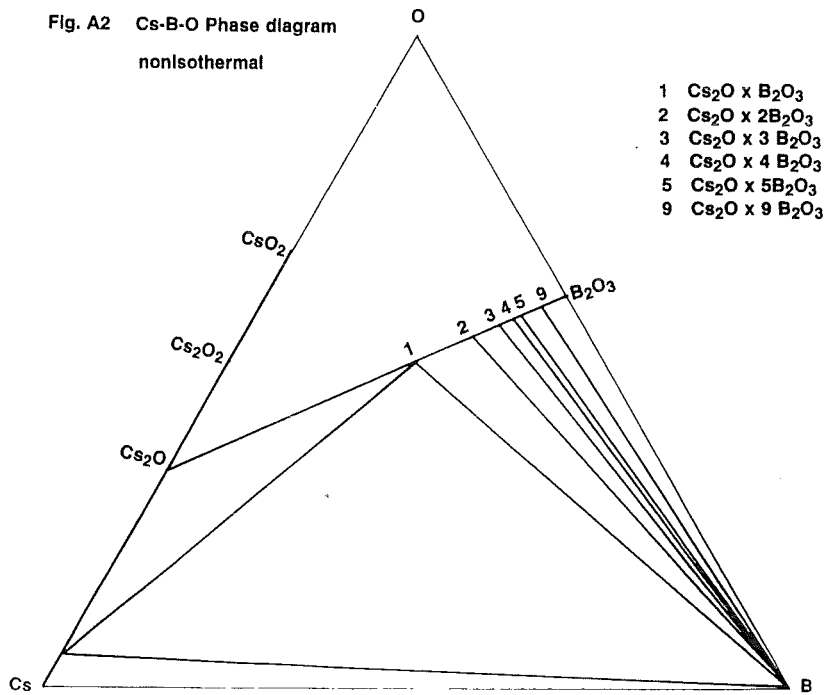
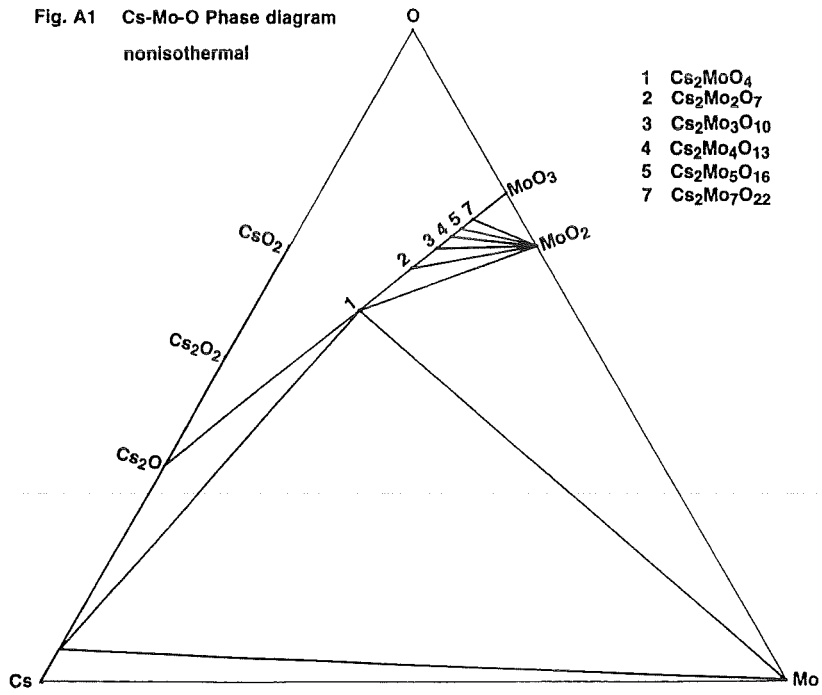
	Mo-release %						
		20	30	35	40	60	100
Cs	Cs ₂ MoO ₄	55.06	82.60	96.19	97.49	97.69	97.69
	CsOH	8.16	2.90	0.01	0.005	0.005	0.005
	CsI	7.01	7.01	3.57	2.38	2.19	2.19
	CsCl	2.70	2.64	0.21	0.11	0.11	0.11
	Cs	-	-	-	-	-	-
	CsBO ₂	27.08	4.86	0.02	0.01	0.01	0.01
I	CsI	99.94	99.82	50.87	33.89	31.18	31.18
	AgI	0.01	0.04	5.60	6.73	7.14	7.14
	InI	0.005	0.01	2.26	3.41	2.92	2.92
	CdI	0.004	0.01	19.11	27.70	30.41	30.41
	HI+I	0.05	0.14	22.64	28.30	28.36	28.36

**Table 18: edistribution of volatile F. P. over species in FPT1 at 973/700°C,
Effect of Mo-fraction in reduced atmosphere with Boron addition**

	% Mo-release	50 % H2			90 % H2			
		20	40	100	20	30	35	100
Cs	CsMoO4	55.06	97.64	97.64	55.06	85.41	85.42	85.49
	CsOH	3.21	0.14	0.14	0.54	0.40	0.40	0.40
	CsI	5.95	1.39	1.39	4.01	3.49	3.48	3.47
	CsCl	1.78	0.21	0.21	0.86	0.69	0.69	0.69
	Cs	0.002	-	-	0.002	0.001	0.001	0.001
	CsBO2	34.00	0.61	0.61	39.53	10.01	10.0	9.94
I	CsI	84.74	19.86	19.86	57.18	49.68	49.67	49.52
	AgI	-	0.002	0.002	-	-	-	-
	InI	15.22	79.90	79.90	42.78	50.28	50.29	50.44
	CdI	-	0.001	0.001	-	-	-	-
	HI+I	0.04	0.23	0.23	0.04	0.04	0.04	0.04

Appendix

The appendix contains several phase diagrams in support of arguments used in the report.



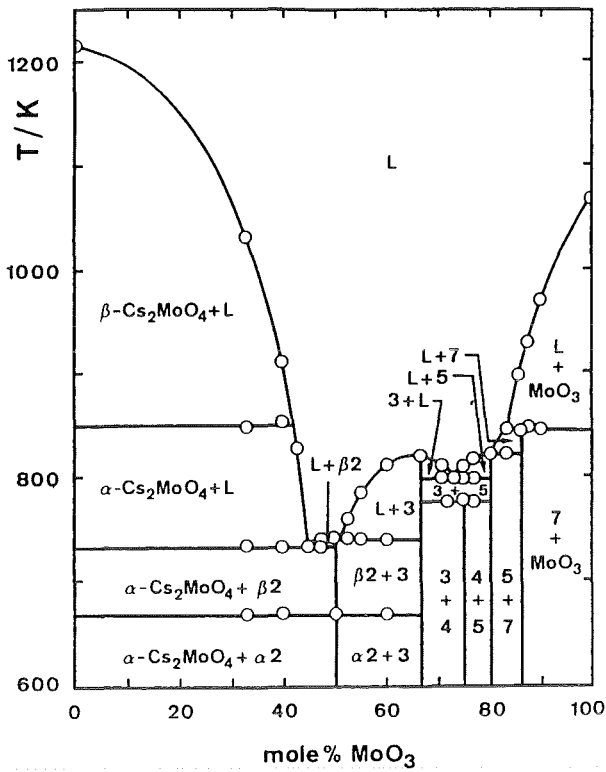


Fig. A3 System $\text{Cs}_2\text{MoO}_4\text{-MoO}_3$ T/K

the numbers refer to the corresponding polymolybdates.

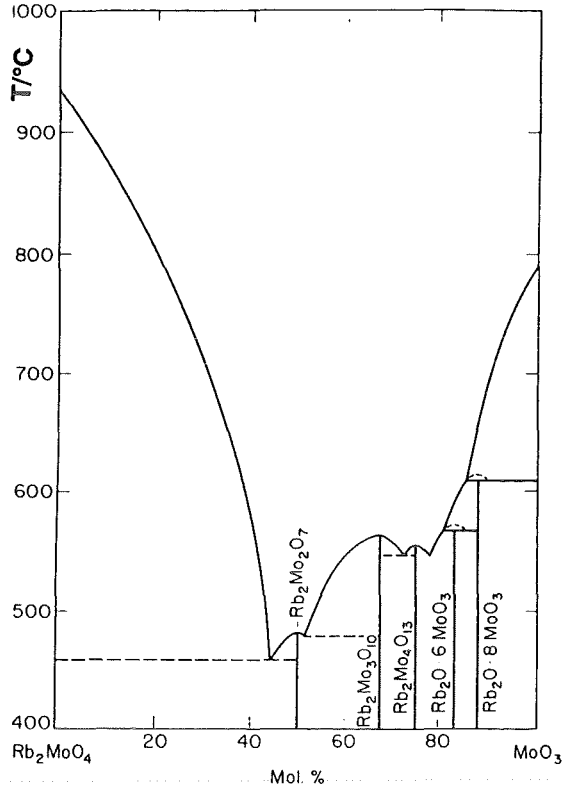


Fig. A4 System $\text{Rb}_2\text{MoO}_4\text{-MoO}_3$ T/°C

Viktor Spitzyn and I. M. Kuleshov, *Zhur. Obshchei Khim.*, 21, 1370 (1951).

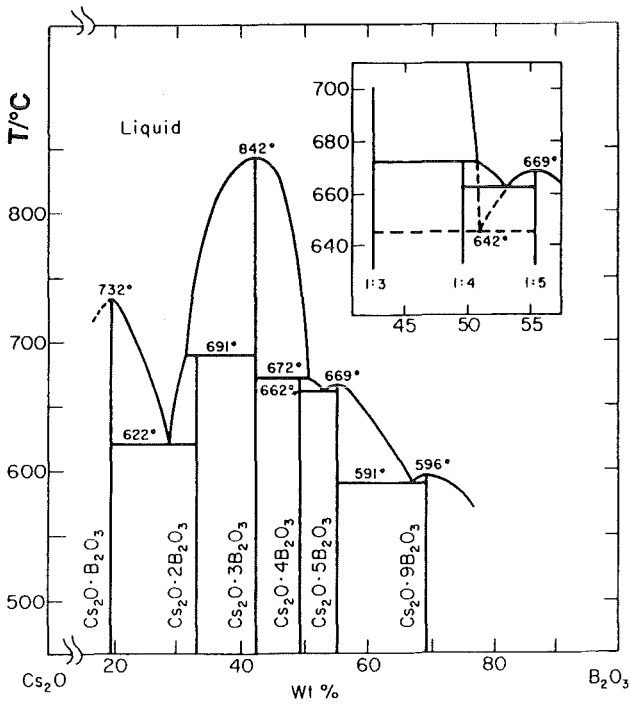


Fig. A5 System $\text{Cs}_2\text{O-B}_2\text{O}_3$ T/°C

FIG. 4264.—System $\text{Cs}_2\text{O-B}_2\text{O}_3$. Inset shows metastable eutectic melting at 642° between $\text{Cs}_2\text{O} \cdot 3\text{B}_2\text{O}_3$ and $\text{Cs}_2\text{O} \cdot 5\text{B}_2\text{O}_3$.
J. Kocher, *Rev. Chim. Miner.*, 3 [2] 221 (1966).

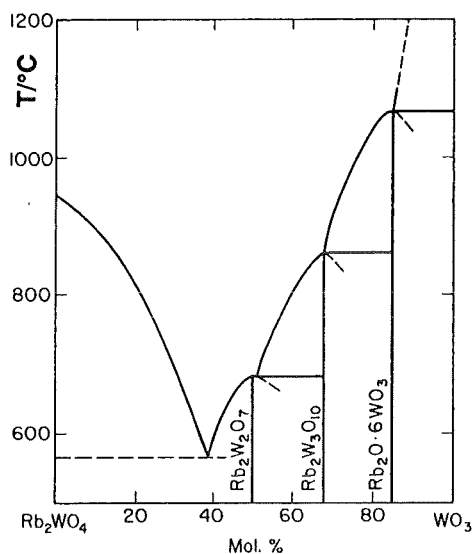


Fig. A6 System Rb_2WO_4 - WO_3 $T/^\circ\text{C}$

V. I. Spitsyn and I. M. Kuleshov, *J. Phys. Chem. U.S.S.R.*, 24 [10] 1197 (1950).

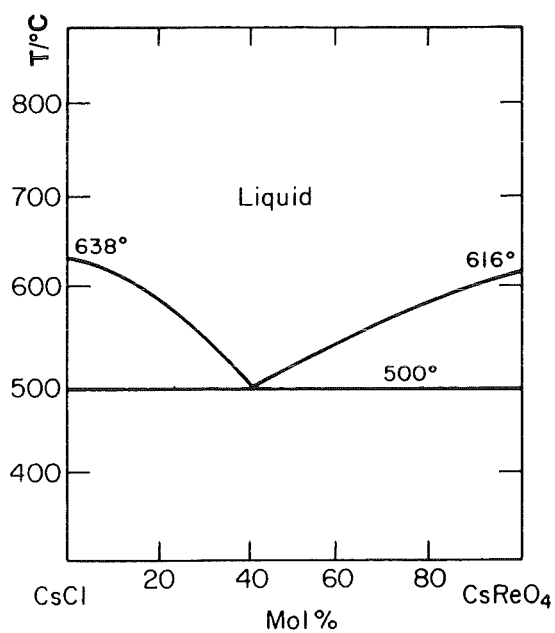


Fig. A7 System CsCl - CsReO_4 $T/^\circ\text{C}$

N. M. Igumnova, D. V. Drobot, and V. V. Tolokonnikova, *Zh. Neorg. Khim.*, 20 [2] 556 (1975); *Russ. J. Inorg. Chem. (Engl. Transl.)*, 20 [2] 310 (1975).