

Gaseous Fission Products in the Stable Salt Reactor

Perhaps the defining advantage of molten salt fuel over solid fuels is the form that fission products take. Fission products are inevitable products in nuclear reactors but the chemical form they take matters a great deal to the risks associated with such reactors.

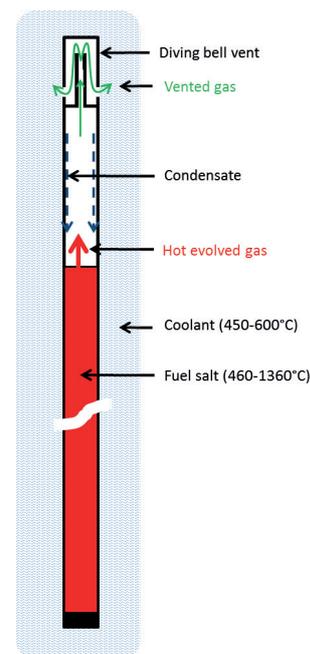
This was dramatically shown at Chernobyl where a large fraction of the core iodine, cesium and tellurium were released, largely in airborne form which spread for thousands of miles, while a far smaller fraction of other non-volatile fission products and actinides were released mostly as larger particles concentrated around the reactor area..

In any molten salt fuel, caesium is present not as the element (as it predominantly is in solid fuels) but as caesium chloride or fluoride. Caesium metal has a boiling point of just 670°C and is highly reactive with air forming oxide fumes. Caesium halides are chemically stable and have boiling points above 1250°C and are diluted in the bulk molten salt where their vapour pressure is greatly reduced until temperatures are even higher than this. From the perspective of caesium, any molten salt fuel is thus far safer than a solid fuel under core damage conditions.

The form of iodine and tellurium in molten salt reactors can be less favourable. While sodium iodide has a high boiling point, like the caesium salts, iodine can form volatile species in molten salts of which the most important are the tellurium iodides.

Since the fuel tubes in Stable Salt Reactors are vented, it is vital to know what the composition of the gasses being vented are, and whether any special systems will be required to manage any hazard.

To establish that composition, thermodynamic equilibrium calculations were carried out on a model partly fissioned molten salt fuel containing 60% NaCl, 20% UCl₃ and 20% PuCl₃ in which a quarter of the Pu had been fissioned producing 10% fission products and 15% liberated chlorine. HSC Chemistry v8 software was used which enabled modelling of the entire chemically complex system with all fission products. The elemental composition is shown in the table below.



Mol % of elements and NaCl in fuel salt

Ag	0.065	In	0.004	Rh	0.295	Xe	1.04
Ba	0.336	Kr	0.113	Ru	0.994	Y	0.115
Br	0.009	La	0.28	Sb	0.014	Zr	1.076
Cd	0.038	Mo	1.129	Se	0.024	Pu	15
Ce	0.612	Nb	0.007	Sm	0.217	Am	0.334
Cs	0.935	Nd	0.8	Sn	0.037	Np	1.113
Dy	0.001	Pd	0.671	Sr	0.206	Cm	0.093
Eu	0.031	Pm	0.075	Tb	0.002	U	18.46
Gd	0.022	Pr	0.231	Tc	0.303	Cl	15
I	0.075	Rb	0.096	Te	0.147	NaCl	60

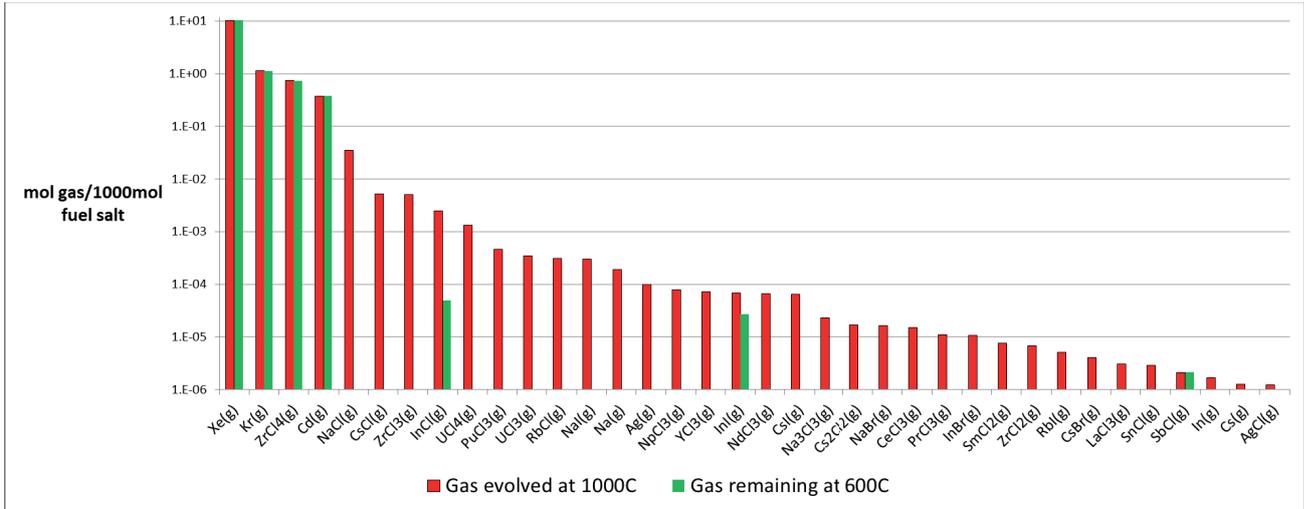
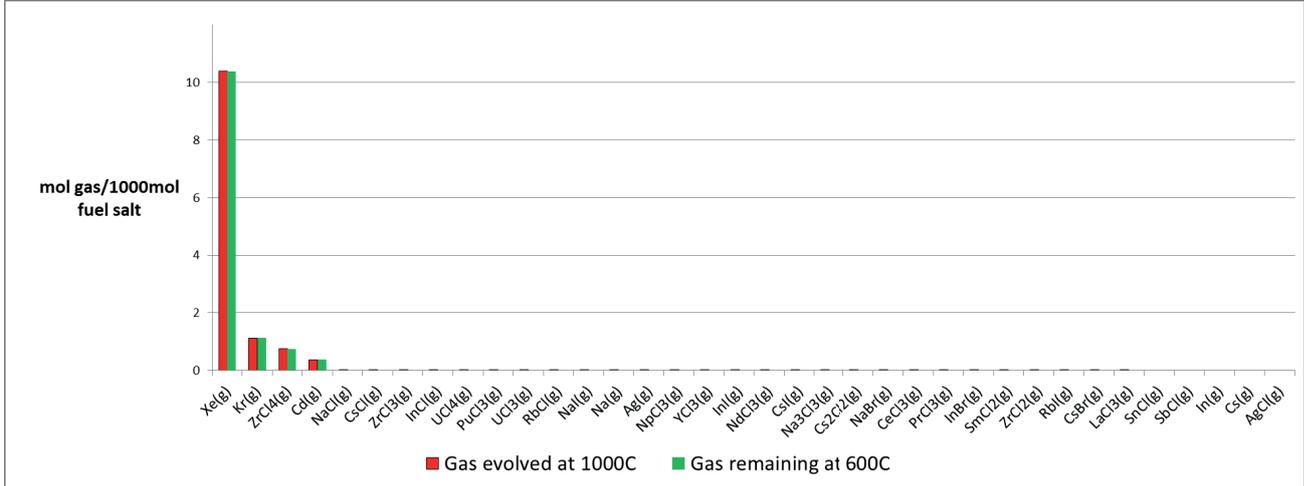
To the right is shown a diagram of a fuel tube. The top of the tube is closed with a “diving bell” assembly which allows gas to escape but prevents coolant entering the fuel tube. There is a gas space in the tube above the hot fuel salt where the tube walls are cooled to the temperature of the coolant, about 600°C. Calculating the gas phase composition is therefore a two stage process.

First the composition of the gas actually released from the hot fuel salt is calculated. This gas rises in the form of bubbles formed in the fuel salt, primarily from the noble gasses xenon and krypton. The temperature selected to define the composition of this gas was 1000°C. This is a pessimistic (high) temperature estimate since the noble gasses have lower solubility at lower temperatures in molten salts and will therefore form bubbles in the cooler regions of the fuel salt. The high temperature and the assumption

that the volatile fission products in the molten salt fully equilibrate with the gas bubbles both represent very conservative assumptions that will overestimate the amount of volatile fission products in the gas.

The second stage is to allow this hot gas to condense on the cooler walls of the fuel tube. The mean residence time of the gas in this space is many days (see below) so full equilibration of the gas with the cooler tube wall is expected. This final gas phase is what is vented from the “diving bell” gas venting system.

The graphs below show the composition of the gas evolved from the fuel salt and its composition after condensing at 600°C. The upper graph is a linear scale showing just how the composition is dominated by the noble gasses. The lower graph has a logarithmic scale so that the contributions of minor gasses can be more easily seen



The fuel salt was in contact with sacrificial zirconium in this simulation, an invention which not only controls the level of dangerous volatile gasses but also prevents corrosion of the tube wall (See "Materials for building the SSR"). Without the sacrificial zirconium there were high levels of tellurium iodide and uranium tetrachloride in the gas phase showing that the sacrificial zirconium has very positive effects on the vapour produced from the fuel salt in addition to its other benefits.

The gas stream from the fuel salt thus contains principally the noble gasses with substantial amounts of $ZrCl_4$ and cadmium and only traces of other compounds. Of particular importance is the very low level of iodine and caesium gasses, representing just 0.001% of the total in the salt.

The substantial amounts of $ZrCl_4$ in the gas stream will contain radioactive isotopes of zirconium since, although much of the zirconium comes from the sacrificial metal, some is formed as fission products. The preferred molten coolant salt contains ZrF_4 and KF which undergo neutron activation while passing through the reactor core. The $ZrCl_4$ will readily dissolve in this coolant and the additional radioactivity created in the coolant salt makes minimal difference to the safety or disposal route for used coolant salt, given that it already contains radioactive zirconium and chlorine (from activation of K) isotopes.

This leaves just the fate of the xenon, krypton and cadmium to be considered. Both noble

gasses contain radioactive isotopes and will not dissolve significantly in the coolant salt, instead accumulating in the gas space within the reactor. Noble gasses have rather low radiological hazard levels since they readily disperse and are rapidly eliminated from the human body even if inhaled. Indeed, ^{85}Kr which is the main radioactive gas remaining in spent conventional nuclear fuel is routinely discharged to the atmosphere during fuel reprocessing operations. However, xenon isotopes can decay to far more hazardous caesium isotopes and it is therefore a legitimate concern that such caesium isotopes should not escape from the fuel tube indirectly via the xenon precursor.

The fuel tube can be considered to contain three separate reservoirs of gas – that dissolved in the fuel salt, that in the gas space above the fuel salt and that in the diving bell apparatus. Flow between these reservoirs is essentially unidirectional, even allowing for diffusion in gasses, due to the very small surface area of the fuel salt in contact with the first gas space and the narrow long tube separating the first gas space from the diving bell gas space. These gas spaces can individually be considered to be well mixed since the mean time spent by gas in each space before passing on is relatively long.

Flow of radioactive xenon and krypton through this series of gas spaces can therefore be calculated based on the gas flow rate, the volume of the three gas spaces and the half life of the isotope. The results are shown in this table.

Xe isotope	Isotope half life (hr)	% emerging from fuel salt	% emerging from main tube	% emerging from diving bell	Overall % released
133	125.94	99.55193	39.42738	72.25031	28.3588
135	9.14	93.41306	4.682505	16.423	0.7184
137	0.063333	8.948511	0.033793	0.135036	4.08E-06
138	0.233333	26.58122	0.124362	0.495598	1.64E-04

Significant amounts of ^{133}Xe , decaying to non-radioactive ^{133}Cs , just 0.2% of ^{135}Xe , decaying to the very mildly radioactive ^{135}Cs and virtually none of the ^{137}Xe decaying to the dangerous isotope ^{137}Cs are released from the fuel tube. The only significant radioactive species in the gasses are therefore the noble gasses themselves.

The cadmium vapour released is more of a chemical toxicity issue than radiochemical issue since the isotopes surviving long enough to escape the fuel tube are of low radioactivity. Cadmium condenses and freezes at $321^{\circ}C$ and is therefore easily filtered

out in the argon cooling and circulation system built into the argon containment dome.

It can be concluded therefore that it will be acceptable to allow those gasses to be accumulated and decay within the reactor containment with periodic releases to the atmosphere as the airlock into the reactor containment is operated. The radiological hazard of these releases would be substantially lower than the unavoidable radioactive releases from current reactors.