ISOTOPIC COMPOSITION OF FISSION GASES IN LWR FUEL

T. JONSSON
Studsvik Nuclear AB,
Hot Cell Laboratory,
Nyköping, Sweden

Abstract

Many fuel rods from power reactors and test reactors have been punctured during past years for determination of fission gas release. In many cases the released gas was also analysed by mass spectrometry. The isotopic composition shows systematic variations between different rods, which are much larger than the uncertainties in the analysis. This paper discusses some possibilities and problems with use of the isotopic composition to decide from which part of the fuel the gas was released. In high burnup fuel from thermal reactors loaded with uranium fuel a significant part of the fissions occur in plutonium isotopes. The ratio Xe/Kr generated in the fuel is strongly dependent on the fissioning species. In addition, the isotopic composition of Kr and Xe shows a well detectable difference between fissions in different fissile nuclides.

1. INTRODUCTION

Most LWRs use low enriched uranium oxide as fuel. Thermal fissions in U-235 dominate during the earlier part of the irradiation. Due to the build-up of heavier actinides during the irradiation fissions in Pu-239 and Pu-241 increase in importance as the burnup of the fuel increases. The composition of the fission products varies with the composition of the fuel and the irradiation conditions. The isotopic composition of fission gases is often determined in connection with measurement of gases in the plenum of punctured fuel rods. It can be of interest to discuss how more information on the fuel behaviour can be obtained by use of information available from already performed determinations of gas compositions.

2. FISSION YIELDS

The fission gases are dominated by Xe and Kr. He is also produced in quite significant amounts but is not discussed in this paper.

2.1. Xenon

Xe-131, Xe-132, Xe-133, Xe-134, Xe-135 and Xe-136 are the six dominant xenon isotopes formed by the fission process in nuclear fuel. Xe-133 and Xe-135 are radioactive with half-lives of 5.2 days and 9.1 hours respectively.

Xe-135 has a very large capture cross-section and is transformed to Xe-136 to a large extent depending on the neutron flux. Xe-131 also has a large capture cross-section and is transformed to Xe-132 to a significant extent.

The total yield of the xenon isotopes is rather similar for different uranium and plutonium isotopes. However, the relative amounts of the xenon isotopes are somewhat dependent upon the kind of fission. The quotient (Xe-131 + Xe-132)/Xe-134 is tentatively used to describe this variation. This quotient was chosen, as it will change only slightly upon continued irradiation due to small neutron cross sections for isotopes influencing this quotient. The correctness of this assumption was also checked by ORIGEN calculations.

2.2. Krypton

Kr-83, Kr-84, Kr-85 and Kr-86 are the four dominant krypton isotopes formed by the fission process in nuclear fuel. Kr-85 is radioactive with a half-life of 10.7 years. Kr-83 has a large capture cross-section and is transformed to Kr-84 to a significant extent.

The yield of the krypton isotopes is very different between the different uranium and plutonium isotopes. The Kr yield is generally smaller for fission of the heavier isotopes. The relative amounts of the krypton isotopes are also somewhat dependent upon the kind of fission. The quotient (Kr-83 + Kr-84)/Kr-86 is tentatively used to describe this variation. This quotient was chosen, as it will change only slightly upon continued irradiation due to small neutron cross sections for isotopes influencing this quotient. The correctness of this assumption was also checked by ORIGEN calculations.

2.3. Comparison between yields

The yields of the chosen isotopes are illustrated in Figure 1. It can be observed that the yields of the krypton isotopes decline with increasing weight of the fissile isotopes. It should also be observed that the yield of Xe-134 is fairly independent of the origin. As a test of the usefulness 3 different quotients are plotted against each other in Figure 2. The quotient Kr-86/Xe-134 decreases with increasing weight of the fissioning isotope whereas the use of the quotients (Kr-83 + Kr84)/Kr-86 and (Xe-131 + Xe-132)/Xe-134can be disturbed by the small values of these quotients for Pu-241.

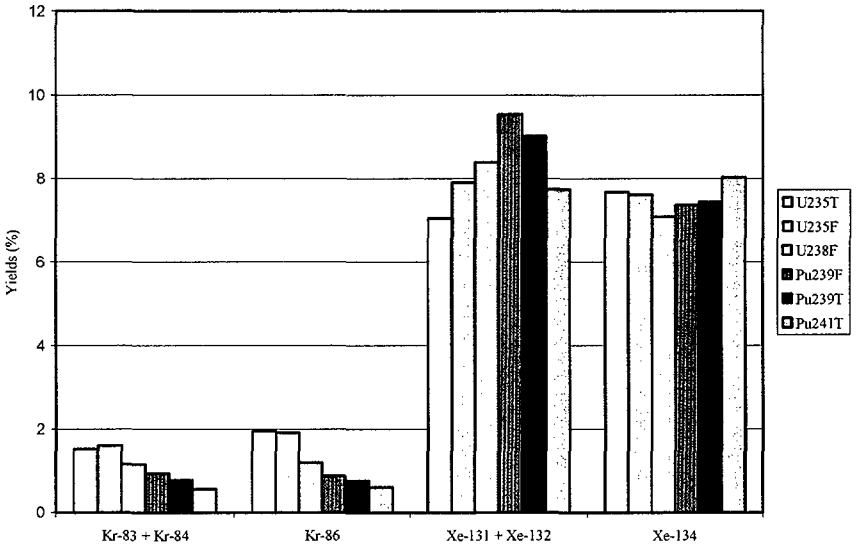

FIG. 1. Fission yields from some heavy nuclides.

3. EXPERIMENTAL DATA

Analysis of the composition of the gases in the plenum exists from many post-irradiation examinations of fuel rods. Results from the majority of the performed determinations in Studsvik of isotopic composition of fission gases with gas mass spectrometry are included in Figure 3[1]. The quotients (Kr-83 + Kr-84)/Kr-86 and (Xe-131 + Xe-132)/Xe-134 are plotted against the quotient Kr-86/Xe-134.

The experimental accuracy is better for the determination of the relative abundance between isotopes of the same element than for comparison between isotopes of different elements.

The two sets of data points in Figure 3 fall mainly along two fairly straight lines in the figure.

[1] Available results from fission gas analysis were only omitted due to apparent errors in the determination of the isotopic composition (insufficient pumping to remove reference gases) or due to uncertainties in the analysis due small amounts of fission gas. (Results are omitted if Kr-84 is less than 0,01 % of the gas content).

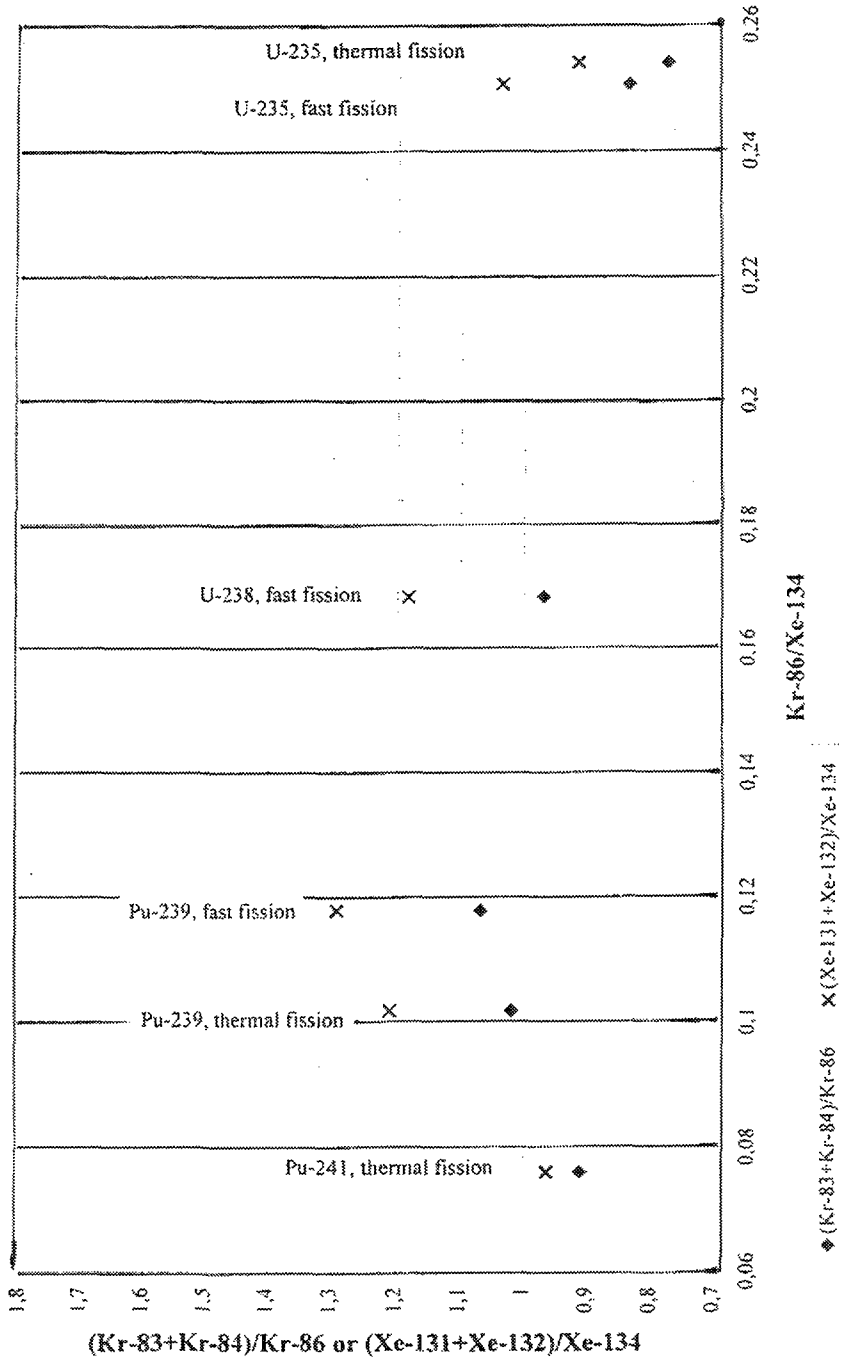

FIG. 2. Quotients between yields.

3.1. Defect fuel

Identification of data points, which deviate most from the general pattern, shows that these points originate from short time tests with fuel irradiated with a high water content in the rods. The explanation for the different composition of the released gas in these cases is presumably changed diffusion in hyperstoichiometric fuel (UO_{2+x}).

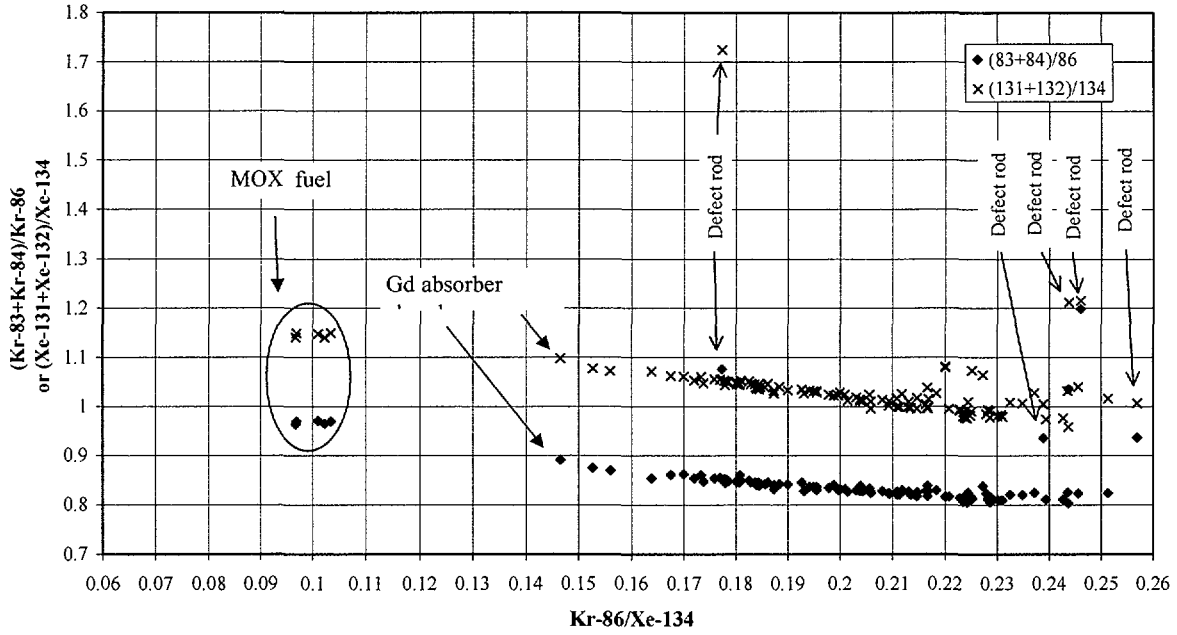

FIG. 3. Relation between quotients, defect and intact rods.

The changed composition of Kr can be explained by a faster diffusion of Br (or Se) in hyperstoichiometric fuel. The krypton isotopes are formed by decay of bromine isotopes. The half-lives of the bromine, selenium and arsenic isotopes are:

Isotope	Half-life	Isotope	Half-life	Isotope	Half-life
As-83	13 seconds	Se-83* Se-83m*	22 minutes 1.2 minutes	Br-83	2.4 hours
As-84	5 seconds	Se-84	3.2 minutes	Br-84	32 minutes
As-86	0,9 seconds	Se-86	15 seconds	Br-86	58 seconds
As decays to Se		Se decays to Br		Br decays to Kr	

* The decay chain for mass number 83 is such that about 1/3 passes the isotope Se-83 and 2/3 passes isotope Se-83m.

The relative amounts of the krypton isotopes are different in fuel rods containing water in such a way that Kr-83 has increased, Kr-84 is about unchanged and the relative content of Kr-86 has decreased.

The changed composition of the released krypton is almost certainly due to increased diffusion velocities of the parent isotopes of krypton, but it is not possible to decide only from these data if the increased diffusion occurs for one or several of the elements bromine, selenium and arsenic.

The changed composition of Xe in fuel with hyperstoichiometric fuel can also be supposed to be caused by faster diffusion of parent nuclides in the decay chain before the formation of Xe. The xenon isotopes are formed by decay of iodine isotopes. The half-lives of the iodine, tellurium and antimony isotopes are:

Isotope	Half-life	Isotope	Half-life	Isotope	Half-life
Sb-131	23 minutes	Te-131* Te-131m*	1.25 days 25 minutes	I-131	8 days
Sb-132	4.2 minutes 2.8 minutes	Te-132	3.3 days	I-132 I-132m	2.3 hours 1.4 hours
Sb-134	10.4 seconds	Te-134	42 minutes	I-134 I-134m	53 minutes 3.5 minutes
Sb decays to Te		Te decays to I		I decays to Xe	

* The decay chain for mass number 131 is such that about 1/8 passes the isotope Te-131 and 7/8 passes isotope Te-131m.

The largest (Xe-131 + Xe-132)/Xe-134 quotients are due to greatly increased content of Xe-132, which can be suspected to be caused by fast diffusion of Te-132 in hyperstoichiometric fuel. A fast diffusion of iodine would instead result in an increased release of Xe-131 due to the longer half-life of I-131. More specifically this indicates that the diffusion of tellurium must be faster than the diffusion of iodine or xenon to explain the experimental results by different diffusion velocities.

In analogy with this it can be suspected that the increase of the quotient (Kr-83 + Kr-84)/Kr-86 in hyperstoichiometric fuel is caused by faster diffusion of the selenium isotopes and not of the bromine isotopes. This analogy is based on the chemical similarity between the two halogens bromine and iodine and between the two group VI elements selenium and tellurium.

3.2. Intact fuel

The 5 points with the smallest Kr-86/Xe-134 quotients in Figure 3 originate from analysis of gas from MOX fuel. Most fissions in MOX fuel occur in plutonium and a smaller Kr-86/Xe-134 quotient can be expected.

The analysis of a fuel rod with Gd also shows a rather small Kr-86/Xe-134 quotient. It is reasonable that the fraction of plutonium fissions can be somewhat larger in a gadolinium rod with a depressed thermal neutron flux.

There is also some other data points in figure 3, which have a large quotient (Xe-131 + Xe-132)/Xe-134. The explanation of these deviations is unclear for the moment and will not be discussed here.

3.3. Ramping of refabricated rods

Figure 4 shows some examples of analysis of released gas in father rods irradiated in power reactors together with analysis of released gas in refabricated rods after test in the R2 reactor in Studsvik.

The measured compositions show that the released gas during ramp in most cases originate to larger extent from U-235 fission than gas released during base irradiation. It is evident that the release during the ramp is dominated by release from the central parts of the fuel with relatively small build-up of plutonium. The release during base irradiation should to a large extent originate from more peripheral parts of the fuel with larger build-up of plutonium.

The release during base irradiation is in all cases below 1 % except for the rod with the highest burnup (70 MWd/kgU). It is likely from the power history that most of the release occurred early during the irradiation of this rod. It is consequently logical that the released gases during the ramp originate to a larger extent from plutonium than the gases released during the base irradiation for this rod.

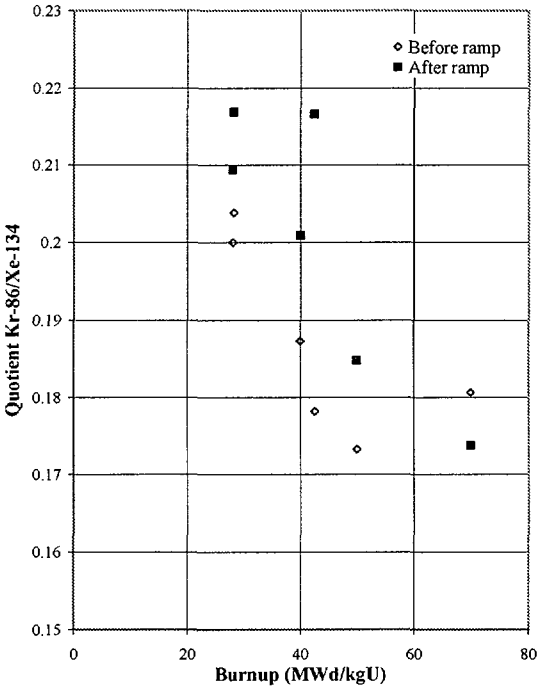

FIG. 4.Comparison between base irradiation and ramp.

4. SUMMARY

The quotient between released krypton and xenon or preferably between the isotopes Kr-86 and Xe-134 gives information about the origin of the released gases. A study of the isotopic compositions of krypton and xenon can in some cases give additional information.

Fuel rods have been irradiated in Studsvik with purposely added water. The isotopic composition of the released gases from the fuel in these rods does not correspond to the composition of the generated gases. It can be ruled out that this is due to separation of the isotopes of the inert gases. It is suggested that separation can occur due to different life times of parent nuclides in the decay chain before formation of krypton and xenon. The suggested explanation is that selenium and tellurium have faster diffusion velocities than inert gases in hyperstoichiometric uranium dioxide fuel.

Several short test rods have been refabricated from power reactor rods for tests in the Studsvik R2 reactor. In some cases gas analysis is available both from the base irradiation and from the R2 test. The fission gas compositions are in such cases different due to release at different times and from different parts of the fuel.