RISØ

RISØ-M-2437

Utilization of the Isotopic Composition of Xe and Kr in Fission Gas Release Research

M. Mogensen

Risø National Laboratory, DK-4000 Roskilde, Denmark
June 1984

10

RISØ-M-2437

UTILIZATION OF THE ISOTOPIC COMPOSITION OF Xe AND Kr IN FISSION GAS RELEASE RESEARCH

M. Mogensen
Metallurgy Department

Abstract. Two examples of how the measured fission gas isotopic composition can be used in the study of fission gas release phenomena are given. In the first example the ratios of Kr85/Kr86 in released and retained gas are used for calculation of the "average time" when the gas was released. This "average time" may be used in code qualification. In the second example the degree of conversion of unstable Xe135 to stable Xe136 is derived from the measured ratio of Xe136 to Xe131+Xe132. This conversion is of importance in the calculation of the total Xe generation during irradiation.

INIS-descriptors: FISSION YIELD; FISSION PRODUCT RELEASE; ISOTOPE COMPOSITION; KRYPTON ISOTOPES; XENON ISOTOPES.

UDC 621.039.548

June 1984
Risø National Laboratory, DK 4000 Roskilde, Denmark

Paper presented at the Enlarged Halden Programme Group Meeting on Fuel Performance Experiments and Analysis Computerized Man-Machine Communication, Hotel Alexandra, Loen, May 23rd-28th, 1983

ISBN 87-550-1018-0
ISSN 0418-6435

Risø Repro 1984

CONTENTS

	Page
INTRODUCTION	5
2. DETERMINATION OF "AVERAGE AGE" OF FISSION GAS	6
2.1. Principle	6
2.2. The case of IFA148	8
2.3. The case of HPO96	9
3. DERIVATION OF THE DEGREE OF CONVERSION OF Xe135 TO Xe136	10
REFERENCES	12
Tables	13
Figures	15

1. INTRODUCTION

Most hot laboratories measure the isotopic composition of released and retained Xe and Kr fission gases on a routine basis. Usually the results are not, however, employed for more than isotope correlations by means of the ratios Xe132/Xe131 and Kr84/Kr83. The objective of this paper is to show two examples of how the measured fission gas isotopic composition can be used directly in the study of fission gas release phenomena.

In the first example, the ratios Kr85/Kr86 in released and retained gas are used to determine the time of gas release in a long term irradiation, assuming that the release happened by one single event. Even though this is not generally the case, this "average age" can be used as a control of code calculations of gas release versus time. The principle in the determination of the "average age" is based on the decay of Kr85 (half life 10.73 y). In the released gas the development of the ratio Kr85/Kr86 follows the usual decay equation whereas the ratio develops differently in the retained gas. It is shown that this can be calculated also in cases where only a very rough irradiation history is known.

The second example is the derivation of the degree of conversion of Xe135 to Xe136 by neutron capture using the Xe isotopic composition. Xe135 is decaying with a half life of 9.17 h. In spite of this, considerable fractions of Xe135 is converted into Xe136 because of the extremely high neutron capture cross section ($2.65 \cdot 10^6$ b). The measured isotope ratios show variations in the conversions of Xe135 to Xe136 from about 40% to about 90% with fuels irradiated in various conditions. The phenomenon is of importance in the calculation of the total Xe generation because up to some twenty per cent of total Xe generation originate in Xe135.

Table 1 gives the fission yields, half lives, and neutron capture cross sections for the Kr and Xe isotopes relevant to fission gas formed in fuel irradiated in a heavy water moderated reactor.

The experimental procedures used at RISØ in obtaining the measured values are described in Refs. 3 and 4.

2. DETERMINATION OF "AVERAGE AGE" OF FISSION GAS

Besides a detailed description of the principle two cases are given to illustrate the utilisation.

2.1. Principle

In short the principle is to determine the two different curves of Kr85/Kr86 as a function of time for released and retained gas. The intersection of these curves gives the "average age" of the gas or the "average time" of the release event. The methods used in establishing these curves are described below:

If the Kr85/86 ratio in a puncture gas is measured at a given time t_m and the measured ratio is denoted Kr85/86(t_m), the ratio Kr85/86(t) at any time t is given by the well known decay equation:

$$Kr85/86(t) = Kr85/86(t_m) \cdot \exp(-\lambda \cdot (t-t_m)) \qquad (1)$$

λ is the Kr85 decay constant.

Regarding the retained gas the situation is more complicated because Kr85 is formed continously by fission. Anyway the concentration of Kr85, C(85), and the concentration of Kr86, C(86), is given by:

$$dC(85)/dt = Y(85)\cdot dF/dt - C(85)\cdot\lambda \quad (2)$$

$$dC(86)/dt = Y(86)\cdot dF/dt \quad (3)$$

Y(85) and Y(86) are the fission yields of Kr85 and Kr86 respectively. dF/dt is the fission rate.

If the power history of the fuel is reduced to periods where dF/dt is constant eq. (2) and eq. (3) have simple solutions.

$$C(85) = (Y(85)/\lambda)\cdot(dF/dt)_n\cdot(1-\exp(-\lambda(t-t_{n-1})) + C(85)_{n-1}\exp(-\lambda(t-t_{n-1})) \quad (4)$$

and

$$C(86) = Y(86)\cdot(dF/dt)\cdot(t-t_{n-1}) + C(86)_{n-1} \quad (5)$$

$(dF/dt)_n$ is the fission rate in the n'th period, t_{n-1} and t_n is the beginning respectively the end of the n'th period, and $C(85)_{n-1}$ and $C(86)_{n-1}$ is the concentration of Kr85 and Kr86 at the end of period number n-1. $t_o = 0$ at the time of charging the fuel into the reactor.

From the time of discharge the decay of Kr85 is calculated according to the simple decay equation i.e. $(dF/dt) = 0$ in eq. (4).

It should be realized that the absolute fission rate is only of secondary importance in determining the ratio of Kr85/86 = C(85)/C(86). This may be recognized by dividing eq. (5) into eq. (4) in the case of $n = 1$, where $C(85)_o = 0$ and $C(86)_o = 0$. The result is:

$$Kr85/86 = C(85)/C(86) = Y(85)\cdot(1-\exp(-\lambda t))/\lambda\cdot Y(86)\cdot t \quad (6)$$

In eq. (6) the fission rate does not appear at all. However, in cases where there is a large difference between two average fis-

sion rates in two long periods i.e. periods of lengths comparable to the Kr85 half life of 10.73 y, the absolute fission rate has to be taken into account. In other cases relative fission rates may be used as a good approximation.

2.2. The case of IFA148

The IFA148 fuel was irradiated in the OECD Halden reactor and later on examined in the RISØ Fission Gas Project as described in Ref. 5, which gives pre-irradiation data as well as base-irradiation and post irradiation data.

Figure 1 shows an extremely simplyfied power history which was used to calculate the development of Kr85/86 by means of eqs. (5) and (6) using the fission yields of Kr85 and Kr86 from U235 fission given in Table 1. The result of this is shown in Fig. 2 together with the calculated curve of Kr85/86 vs. time for the released gas. This is obtained from the measured value of Kr85/86 in the puncture gas. Also Fig. 2 gives the Kr85/86 measured in the retained gas and a good agreement between the measured and the calculated values is observed.

The intersection of the two curves in Fig. 2 gives the "average time" of the release event, i.e. the time of release in case this took place in one event.

The detailed irradiation history of IFA148 was, however, quite complex as shown in Fig. 3. Still it is thought to be useful to know the "average age" of the gas in order to control code calculations. For example, if the code tells that all gas release happened within the first 800 days of irradiation (Fig. 3) the "average age" proves that either is the code input or the code calculation in error.

2.3. The Case of HP096

The Danish fuel experiment HP096 was used in the IAEA sponsored research program for "The Development of Computer Models for Fuel Element Behaviour in Water Reactors" (in short D-COM). The experiment is described in Ref. 6.

Figure 4 shows Kr85/86 vs. time curves for the released and retained gas giving the "average age" of the released gas. The detailed power history of the test is given in Fig. 5 where also the "average time" of release is shown. It is clear from the low power level that the release did not occur at that time. The interesting question here is: How much of the gas was released early in life and how much was released in the bump test? If it is assumed that all gas release happened in two events, the one at maximum linear heat rating at about 2500 h at power (Fig. 5) which is equivalent to about 130 days in Fig. 4, and the other is the bump test the following calculations can be made: The gas released at time 130 days has because of simple decay a Kr85/86 = 0.1146 at the time of discharge. The gas released in the bump test has Kr85/86 = 0.1307, the one of the retained gas. In the final puncture gas Kr 85/86 = 0.1262 immediately after the bump test. From these values which are all derived from Fig. 4 the following equation can be established

$$0.1307x + 0.1146(1-x) = 0.1262$$

where x is the fraction of the released gas which was released during the bump test. The solution to the equation is x = 0.72 i.e. 72% of the puncture gas was released during the bump. The estimated uncertainty on the determination of the average age is ± 50 days.

3. DERIVATION OF THE DEGREE OF CONVERSION OF Xe135 TO Xe136

The importance of the conversion of Xe135 ($T_{1/2}$ = 9.17 h) to Xe136 (stable) by neutron capture is related to the calculation of the amount of fission gas generated during irradiation. In fact the main uncertainty on the fission gas yields generally used is due to the difficulties in the assessment of the degree of conversion of Xe135 to Xe136.

It has been shown recently (Ref. 7) that the fission gas isotope ratios may be used in calculating more accurate fission gas yields. The principle of Ref. 7 can also be used to calculate the degree of conversion of Xe135 to Xe136. In short, the principle is: First the average fission yields of Xe135 and Xe136 are calculated without taking neutron capture or decay into account at all. These yields are denoted $Y_c(135)$ and $Y_c(136)$. Next the actual yield of Xe136 is calculated from the ratio of Xe136 to Xe131 + Xe132 and the yield of Xe131 + Xe132, which is unaffected of neutron capture reactions. The yield of Xe136 obtained in this way includes the Xe136 formed from Xe135 by neutron capture. It is denoted $Y_m(136)$. The degree of Xe135 to Xe136 is then found as $(Y_m(136) - Y_c(136))/Y_c(135)$.

Table 2 gives some examples of conversion degrees together with data necessary for the calculation (burnup distribution and Xe136/Xe131+Xe132) and data characterizing the fuel and the irradiation conditions (initial enrichment, burnup, fission rate). The fuel M2-2B is from the Danish fuel test HP013 (Ref. 8) PA29-4 and M2-2C are from HP022 (Ref. 9). The HP096 and IFA148 fuel is described in Refs. 6 and 5, respectively.

A very large variation in the degree of Xe135 to Xe136 is observed in Table 2 (from 39% to 91%). Initial enrichment, burnup and fission rate are the main parameters determining the Xe135 to Xe136 conversion. This is seen in the fact that the degree of conversion is in the first approximation proportional to the

Xe135 concentration and the neutron flux. The Xe135 concentration is proportional to the actual fission rate, which in turn is proportional to the neutron flux and the actual enrichment which is a function of initial enrichment and burnup. Besides these parameters also the way of operating the reactor is of importance. Because of the relative short half life of Xe135 of 9.17 h, decay will tend to dominate neutron capture in case the reactor has a large number of short time shut downs.

In this way the results of Table 2 may be rationalized qualitatively, whereas a quantitative calculation would need a very detailed power history and a reactor physics code.

REFERENCES

1. MEEK, M.E., and RIDER, B.F., NEDO-12154-1, 1974.
2. Karlsruhe Nuklidkarte, 4. Auflage 1974.
3. LARSEN, E. EGSGAARD, H., and MOGENSEN, M., Int. J. of Mass Spectrometry and Ion Physics, 48 (1983) 385-8.
4. LARSEN, N.R., and LARSEN, E., RISØ-M-2183, 1979.
5. KNUDSEN, P., BAGGER, C., CARLSEN, H., MISFELDLT, I., and MOGENSEN, M., "The RISØ Fission Gas Project", OECD-NEA-CSNI/IAEA Specialists meeting at RISØ, 16-20 May 1983.
6. MISFELDT, I., "The D-COM Blind problem on Fission Gas Release: Experimental Description and Results", OECD-NEA-CSNI/IAEA Specialists Meeting at RISØ, 16-20 May 1983.
7. MOGENSEN, M., Int. J. of Mass spectrometry and Ion Physics, 48 (1983) 389-92.
8. KNUDSEN, P., and BAGGER, C., RISØ-M-2151, 1978.
9. BAGGER, C., CARLSEN, H., and KNUDSEN, P., RISØ-M-2152.

Table 1. Cumulative fission yields (%), half lives $T_{1/2}$, and thermal neutron capture cross sections σ_c (barn) for the Kr and Xe isotopes relevant to fission gas formed in thermal reactor fuel (Refs. 1 and 2).

Isotope	Fission yield from U235	Pu239	Pu241	$T_{1/2}$	σ_c
Kr83	0.531	0.295	0.204	stable	200
Kr84	0.986	0.478	0.352	stable	0.13
Kr85	0.288	0.130	0.086	10.73y	1.66
Kr86	1.951	0.758	0.610	stable	0.06
Xe131	2.835	3.745	3.118	stable	90
Xe132	4.217	5.275	4.625	stable	0.39
Xe133	6.770	6.973	6.673	5.29d	190
Xe134	7.681	7.448	8.031	stable	0.25
Xe135	6.633	7.466	7.084	9.17h	$2.65 \cdot 10^6$
Xe136	6.273	6.627	7.201	stable	0.16

Tabel 2. Some examples of the degree of conversion of Xe135 to Xe136 in case of different initial enrichments, burnups and average fission rates for the two heavy water moderated reactors DR3 at RISØ and HBWR at Halden.

Reactor	Fuel identification code	Initial enrichment % U235	Burnup % FIMA	Burnup contribution (in % FIMA) from U235	Pu239	Pu241	Average fission rate $f/cm^3 \cdot S$	Ratio of Xe136 to Xe131+ Xe132	% Conversion Xe135 to Xe136	PG/RG*
DR3	M2-2B	1.45	2.87	1.19	1.48	0.20	$6.4 \cdot 10^{12}$	1.596	91	PG
	PA29-4	2.28	4.88	1.94	2.53	0.41	$1.3 \cdot 10^{13}$	1.575	88	PG
	M2-2C	2.28	4.39	1.93	2.14	0.32	$1.1 \cdot 10^{13}$	1.560	86	PG
	HP096	2.28	3.7	1.9	1.6	0.2	$1.1 \cdot 10^{13}$	1.592	87	RG
HBWR	IFA148	5.0	2.00	1.80	0.18	0.02	$3.3 \cdot 10^{12}$	1.230	39	
			3.50	2.99	0.48	0.03	$5.7 \cdot 10^{12}$	1.355	53	RG
			4.70	3.71	0.90	0.09	$7.7 \cdot 0^{12}$	1.456	67	

*PG means that the Xe isopotic reatio was measured on puncture gas, and RG means measured on retained gas.

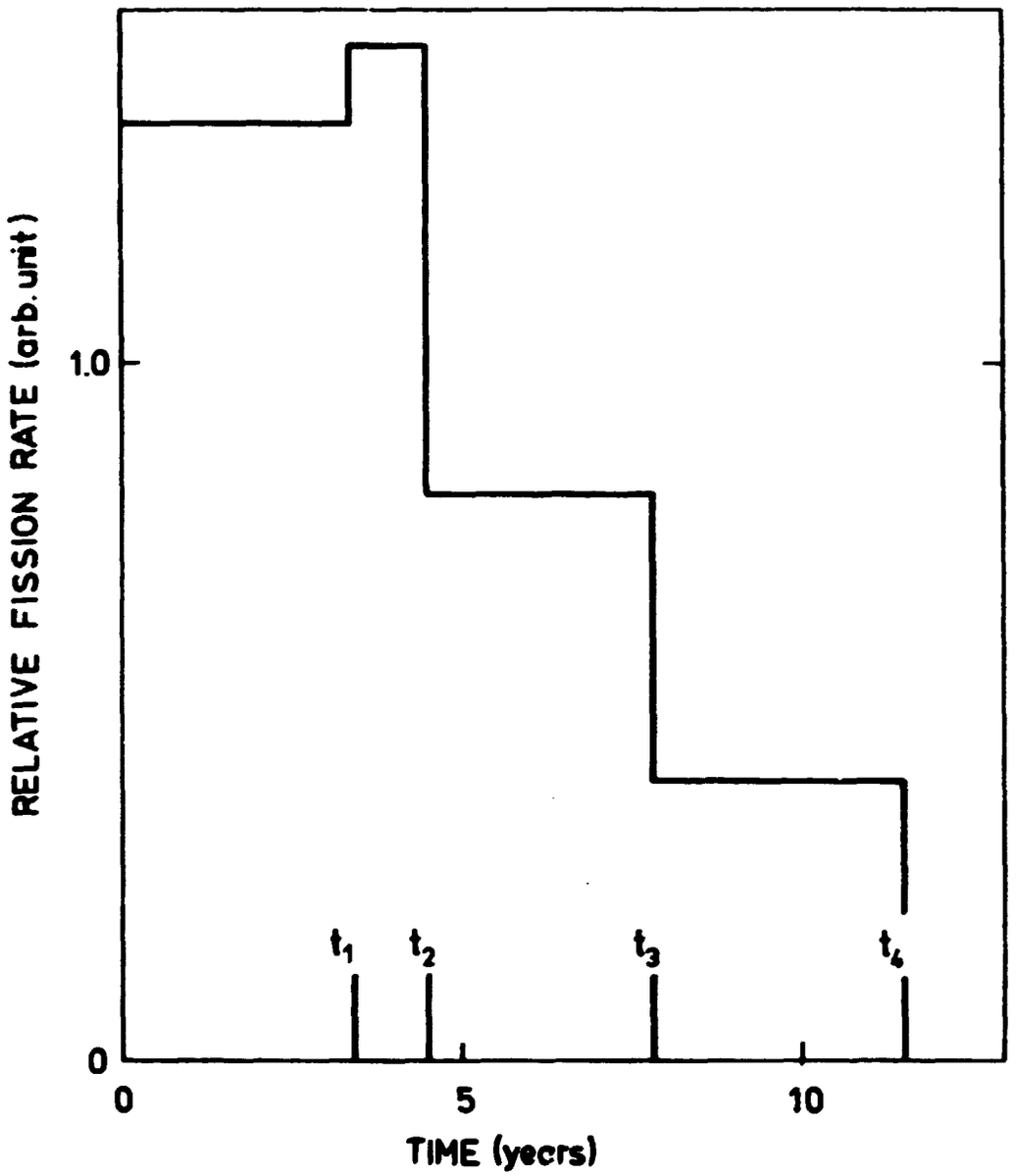

Fig. 1. A very simplified power history for IFA148.

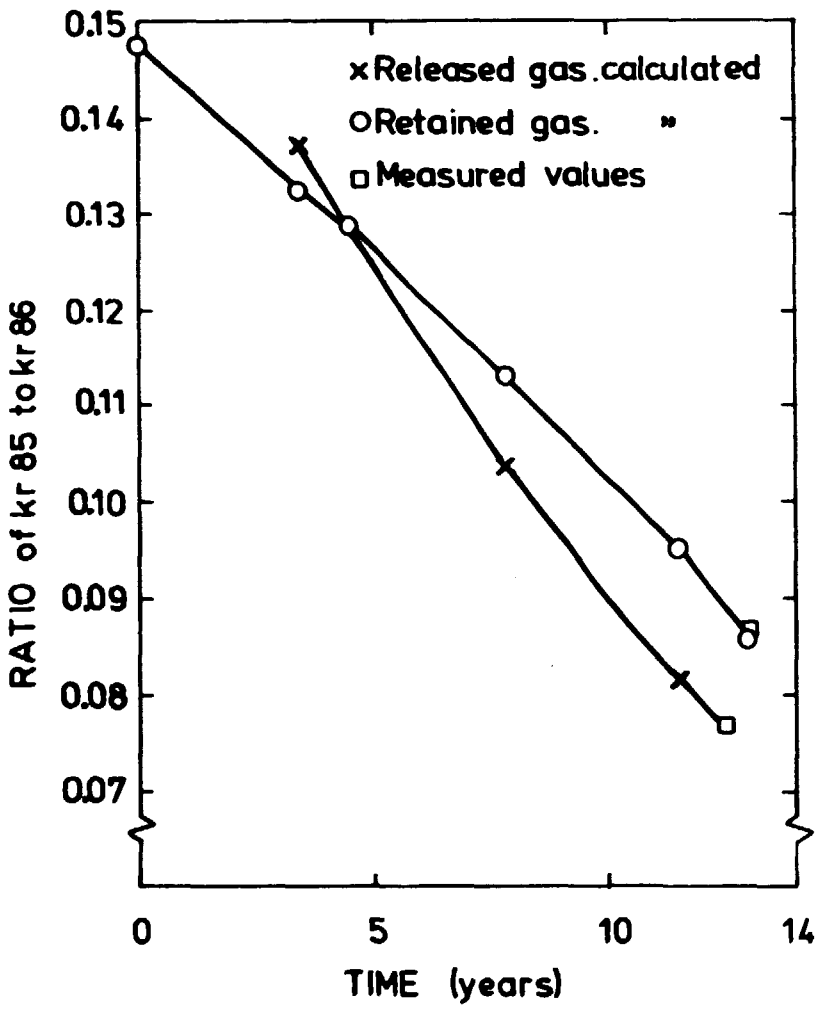

Fig. 2. Calculated Kr85/86 vs. time for released and retained gas during the base irradiation of IFA148. The intersection of the curves gives the "average age" of the released gas.

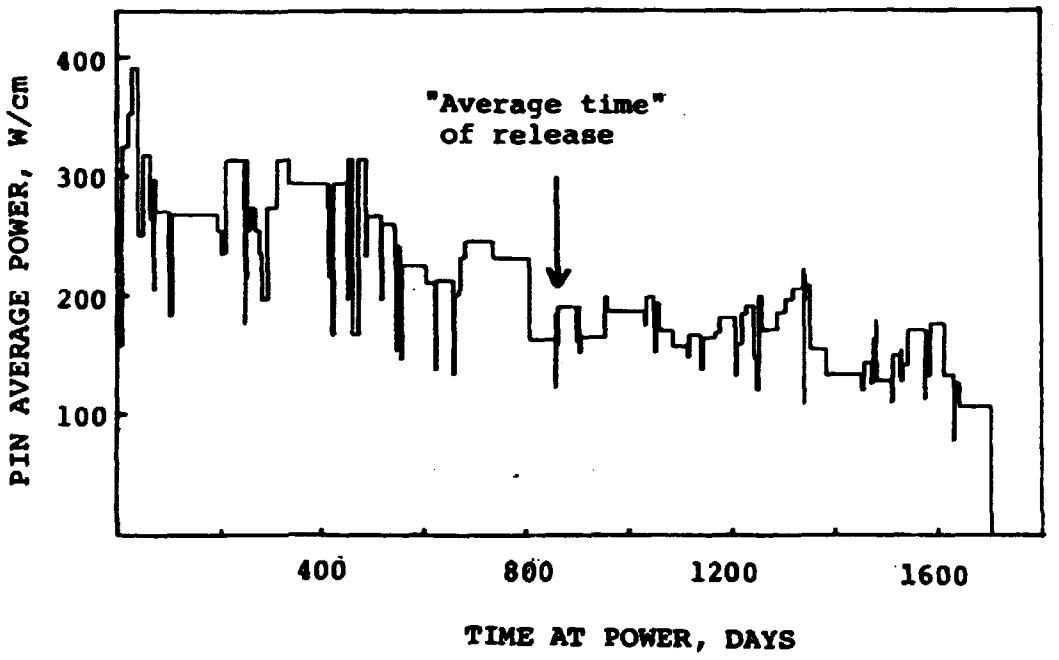

Fig. 3. Assembly power for IFA148.

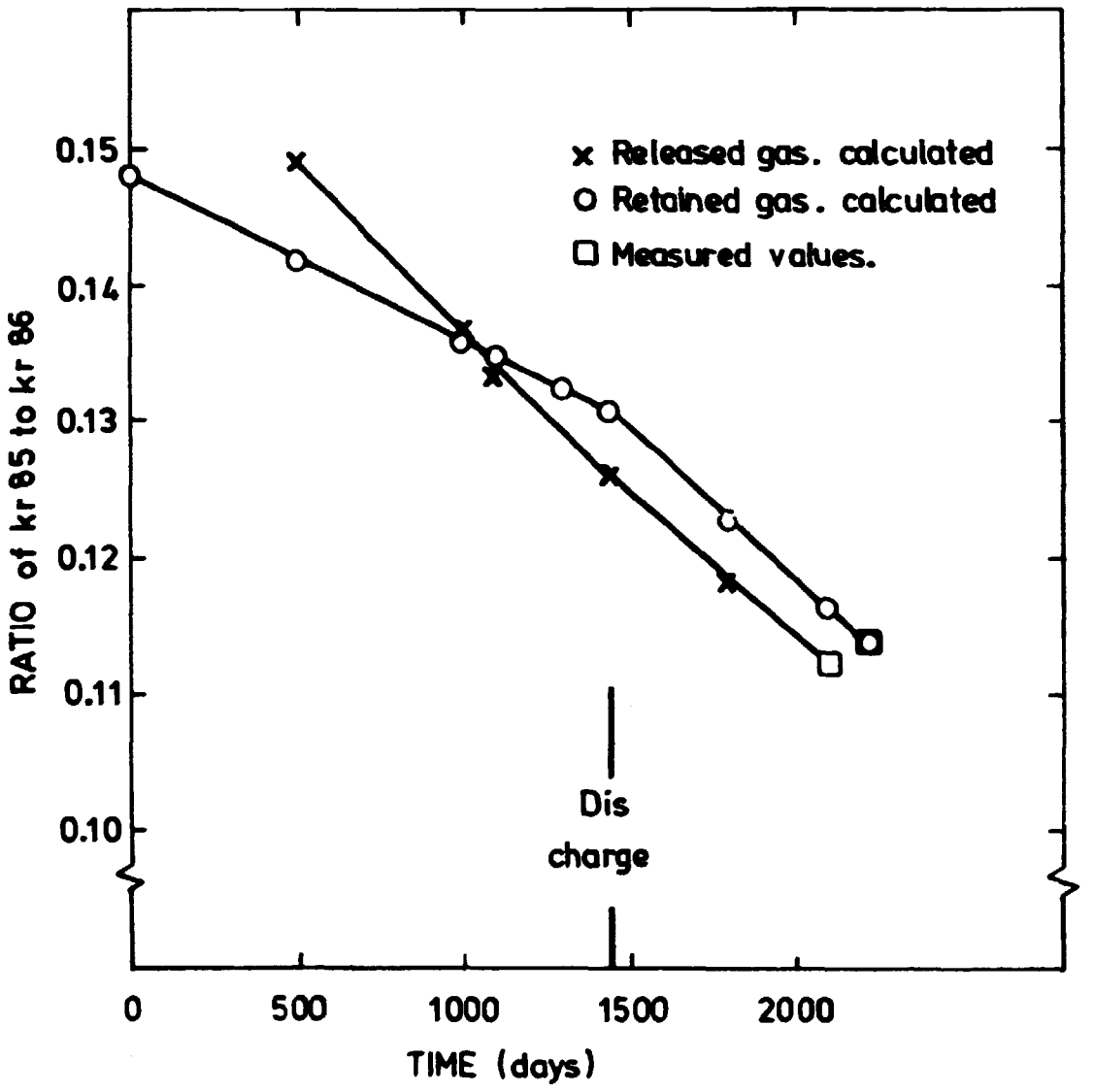

Fig. 4. Calculated Kr85/86 vs. time for released and retained gas during the experiment HP096 and the following bump test. The intersection of the curves gives the "average age" of the released gas.

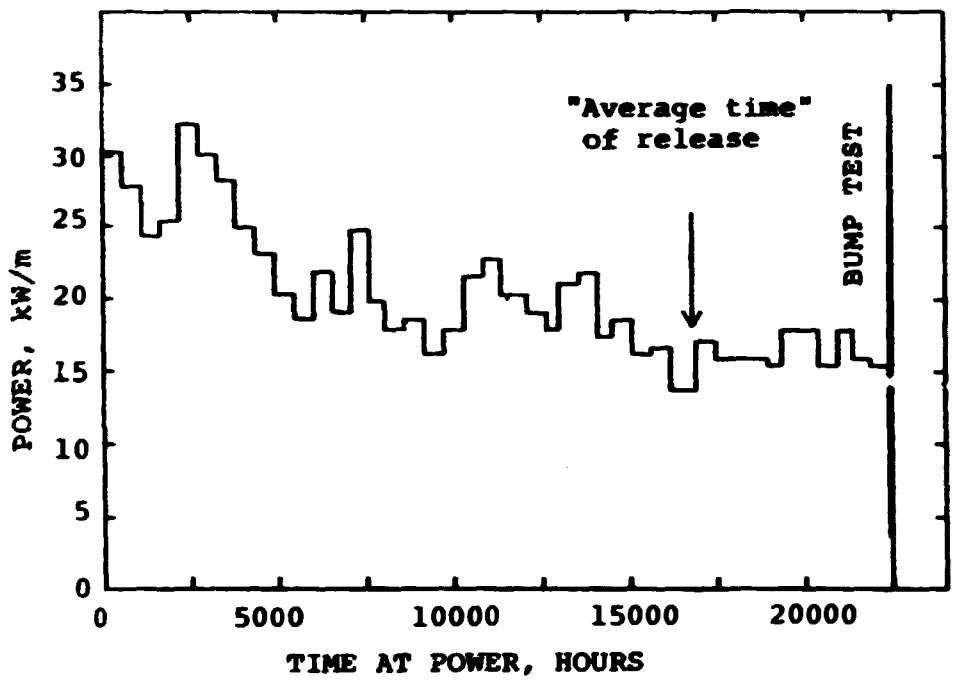

Fig. 5. Base irradiation average assembly power for test HP096.

Risø National Laboratory **Risø-M-** 2437

Risø-M-

UTILIZATION OF THE ISOTOPIC COMPOSITION OF Xe AND Kr IN FISSION GAS RELEASE RESEARCH

M. Mogensen
Metallurgy Department
RISØ National Laboratory
DK-4000 Roskilde, Denmark

Date June 1984

Department or group
Library

Group's own registration number(s)

9 pages + 2 tables + 5 illustrations

Abstract

Two examples of how the measured fission gas isotopic composition can be used in the study of fission gas release phenomena are given. In the first example the ratios of Kr85/ Kr86 in released and retained gas are used for calculation of the "average time" when the gas was released. This "average time" may be used in code qualification. In the second example the degree of conversion of unstable Xe135 to stable Xe136 is derived from the measured ratio of Xe136 to Xe131+Xe132. This conversion is of importance in the calculation of the total Xe generation during irradiation.

Copies to

Available on request from Risø Library, Risø National Laboratory (Risø Bibliotek), Forsøgsanlæg Risø), DK-4000 Roskilde, Denmark
Telephone: (03) 37 12 12, ext. 2262. Telex: 43116

Available on request from
Risø Library
Risø National Laboratory, P.O. Box 49
DK-4000 Roskilde, Denmark
Phone (45) 42 37 12 12 ext. 4004

ISBN 87-550-1018-0
ISSN 0418-6435