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REACTOR COOLANT PRESSURE BOUNDARY LEAKAGE DETECTION SYSTEM

Final report SKI project B28/76

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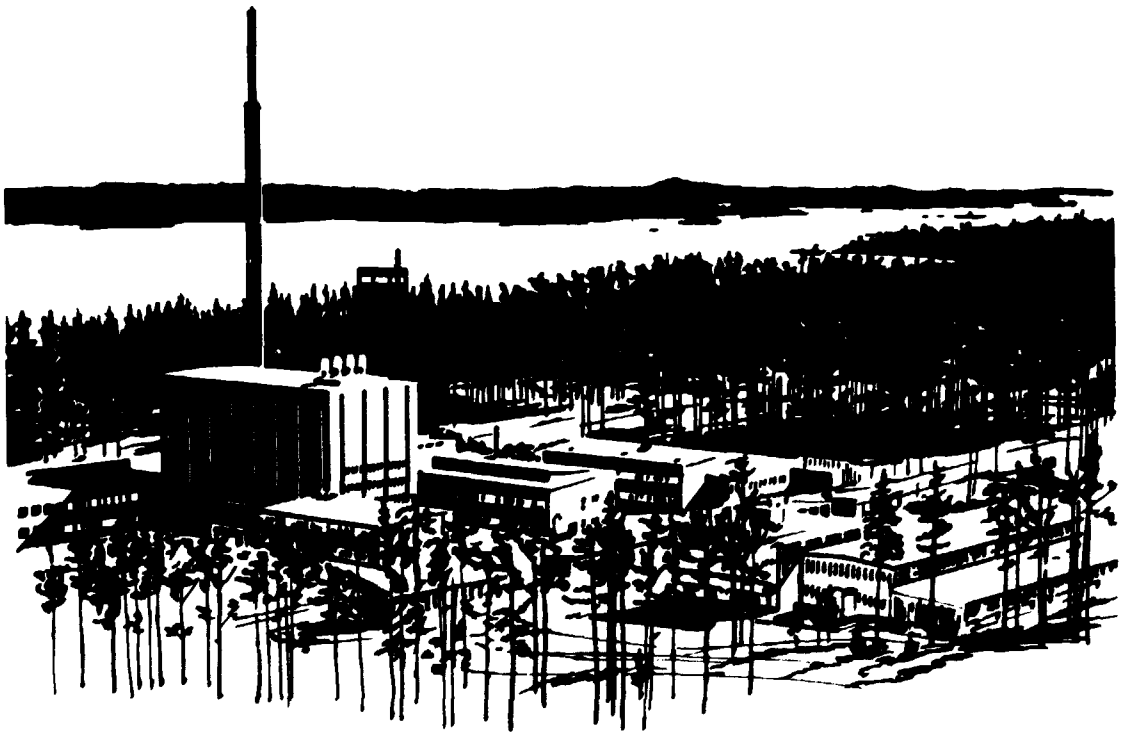

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ABSTRACT

The present paper deals with a system for monitoring the leakage of reactor coolant. This system is based primarily on the detection of the N13 content in the containment atmosphere. N13 is produced from the oxygen of the reactor water via the recoil proton nuclear process H1+O16 → N13+He4. The generation is therefore independent of fuel element leakage and of the corrosion product content in the water. It is solely related to the neutron flux level in the reactor core. Typical figures for the equilibrium N13 concentration in the containment atmosphere following a 4 kg/minute coolant leakage are 5 kBq m^{-3} and 7 kBq m^{-3} for BWR and PWR respectively. These levels are readily measured with a 10 liter Ge(Li) flow detector assembly operated at elevated pressure.

Approved by

LIST OF CONTENTS

1. INTRODUCTION

The USNRC criterion 30-quality of reactor coolant pressure boundary states inter alia that "means shall be provided for detecting and, to the extent practical, identifying the location of the source of reactor coolant leakage". This is generally accomplished with monitoring systems which are based on the measurement of the flow rate from closed systems to collecting tanks.

Such systems are inadquate, however, as regards sensitivity and response time, and also because of excessive levels of identified (legal) and unidentified leakage into the containment atmosphere. As a result, supplementary instrumentation aids are needed.

Suggestions for additional methods for detecting the release of reactor coolant to the containment are listed in the USNRC regulatory guide 1.45. They include the indication and/or monitoring of changes in:

a) airborne particulate radioactivity,
b) airborne gaseous radioactivity,
c) containment atmosphere humidity,
d) containment atmosphere pressure and temperature,
e) condensate flow rate from air coolers.

Provision for the measurement of the content of oxygen and hydrogen will probably also be made in future plants.

This paper deals with leakage monitoring systems based on the measurement of the radioactivity in the containment atmosphere released from the reactor coolant and, particularly, the radioactivity from gaseous isotopes (b).

The information provided by monitoring systems based on the measurement of airborne particulate radioactivity (a) cannot be interpreted into terms of simple coolant leakage rates for several reasons:

The content of activated corrosion products in the coolant varies within the plant production cycle.

The transition to the aerosol phase is characterized by a very low partition factor. Thus, although the concentration in the reactor water, under conditions of normal plant operation, may reach levels comparable to those for noble gases and N13, the overall sensitivity will nevertheless be low.

Finally, the transport of particulate material to the measuring site (outside the containment) is disturbed by deposition in the collecting tubes.

A reactor coolant leakage monitoring system based primarily on the detection of the N13 content in the water is described.

N13 is produced from the oxygen of the reactor water via the recoil proton nuclear process H1+O16 → N13+He4. The generation is therefore independent of fuel element leakage and of the content of corrosion products in the water. It is, instead, solely related to the neutron flux level in the reactor core.

Typical figures for the N13 concentration in the reactor water for a 3000 MWth reactor is 0.4 MBq kg^{-1} (BWR) to 6 MBq kg^{-1} (PWR).

The free volume of the PWR containment is generally much greater than that for the BWR. Therefore the equilibrium concentration in the PWR containment atmosphere which is associated with a 4 kg/minute coolant leakage rate will differ very little from the corresponding concentration in a BWR containment. The values are 5 kBq/m^3 and 7 kBq/m^3 respectively.

These levels are readily measured (511 keV annihilation radiation) with a 10 liter Ge(Li) flow detector assembly operated at elevated pressure (1500 kPa).

Experiments at the R2 material testing reactor in Studsvik, Sweden, indicate that nitrogen is liberated in the containment atmosphere in a chemical form which possesses a very high vapor pressure at room temperature.

Results from experiments that simulate power reactor containment conditions are in good agreement with theoretical models as regards the production rate of N13.

The N13 gas released to the containment atmosphere shows no tendency to redissolve or deposit. The low atomic number involves that N13 exhibits good diffusion properties; this is essential to a representative distribution over the containment volume.

Finally, the decay time (half-life 10 minutes) is ideal from many points of view.

As a supplement to the N13 system the instrumentation can also be used for the detection of the noble gases produced by fission, e.g. Xe135 (half-life 9.1 hour).

With a probable fuel element pin-hole leakage level the long term sensitivity using Xe135 is very high. However, the diffusion of xenon throughout the containment will probably be a rather slow process.

The requirements for an ideal RCPB leakage detection system may be summarized:

1) It has to respond in 1 hour for a 4 kg/minute coolant leakage flow.

2) It has to allow for long sample transmission distances in the reactor containment, making a pattern of semi discrete sampling points possible.

3) It should not depend solely on the fuel element pin-hole level for its operation.

4) It has to be operable even under severe (catastrophic) atmospheric conditions in the reactor containment.

5) The deposition of long-lived radioactive daughters in the measuring system should not to a great extent hazard the system performance at low leakage levels.

The results of the present work seems to indicate that all these requirements could be fulfilled by a pressurized Ge(Li) detector system with air cooler/driers, using N13 as a tracer isotope.

2. SYSTEM DESCRIPTION

The circulating sample flow to and from the reactor containment is accomplished with a two stage piston compressor which at the same time maintains the pressure (1500 kPa) in the measuring chamber (Figure 2.a).

Two coolers before and after the compressor (TD3, TD1) and a refrigeration air drier (Polair 1A2) condition the sample air before it is presented to the measuring chamber.

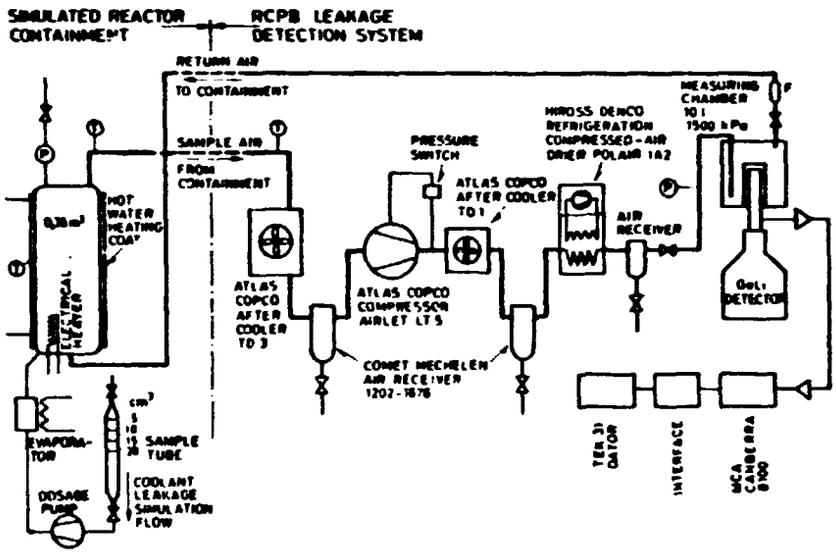

Figure 2.a

RCPB leakage detection system. Experiment at the R2 research reactor, Studsvik, Sweden

The relative humidity of the atmosphere in the measuring chamber which is held at room temperature will be below 30 % which means that the water content in the chamber will not exceed 0.005 g.

The water handling capacity of the system permits measurements to be carried out with a 60° centigrade, 100 % humidity containment atmosphere. Practically all non-volatile fission, corrosion and transuranium element from the containment water are trapped in the air receivers from which they may be collected for laboratory analysis.

The capacity of the LT5 compressor permits a sample air flow rate of 100 g/min, resulting in a collector time constant of about 4 minutes.

2.1 Detector assembly

The measuring chamber (Figure 2.b), approved for 2000 kPa is manufactured from stainless steel (18 % Cr, 11 % Ni, 2.7 % Mo). A wall thickness of 3 mm permits measurement at 250 keV photon energy (Xe135) with only moderate attenuation.

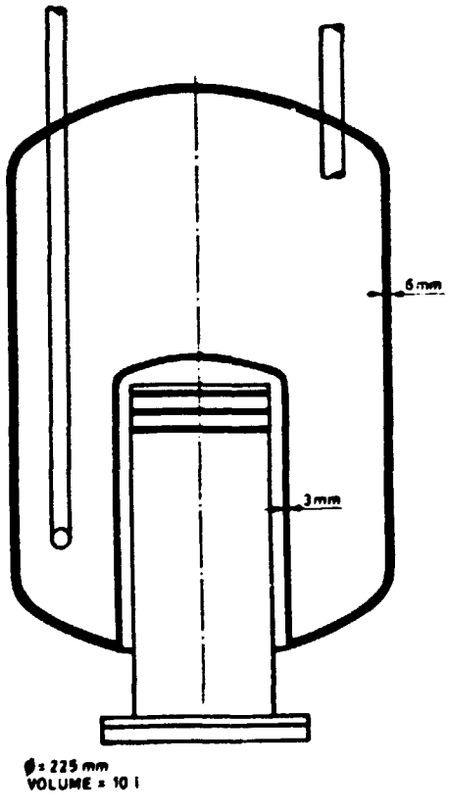

Figure 2.b

Detector assembly 73 cm^3 Ge(Li) detector.
Measuring pressure 1500 kPa.

A Ge(Li) coaxial detector with 73 cm^3 sensitive volume and with a relative efficiency of 15 % is situated in the center of the measuring chamber.

Calibration of the assembly has been carried out with a dummy chamber with Eu152 and Na22 sources using the energies 122, 245, 344 and 511 keV.

The result for 100 kPa sample pressure and with measuring chamber pressure 1500 kPa applied is:

N13, 511 keV, 283 counts per 600 seconds/kBq m^{-3}
F18, 511 keV, 274 "-
Xe135m, 527 keV, 111 "-
Xe135, 250 keV, 255 "-

2.2 Peak integration

The multichannel analyzer is set for 1 keV per channel and simple pedestal integration is used.

Signal as well as background is counted in a 11 keV energy range and the background subtraction is carried out according to the following:

Xe135 (250 keV)

$$n = \sum_{E=245}^{E=255} n_E - \sum_{E=240}^{E=244} n_E - \sum_{E=256}^{E=261} n_E \qquad \text{(Eq 2.a)}$$

N13 (511 keV)

$$n = \sum_{E=506}^{E=516} n_E - \sum_{E=499}^{E=505} n_E - \sum_{E=517}^{E=520} n_E \qquad \text{(Eq 2.b)}$$

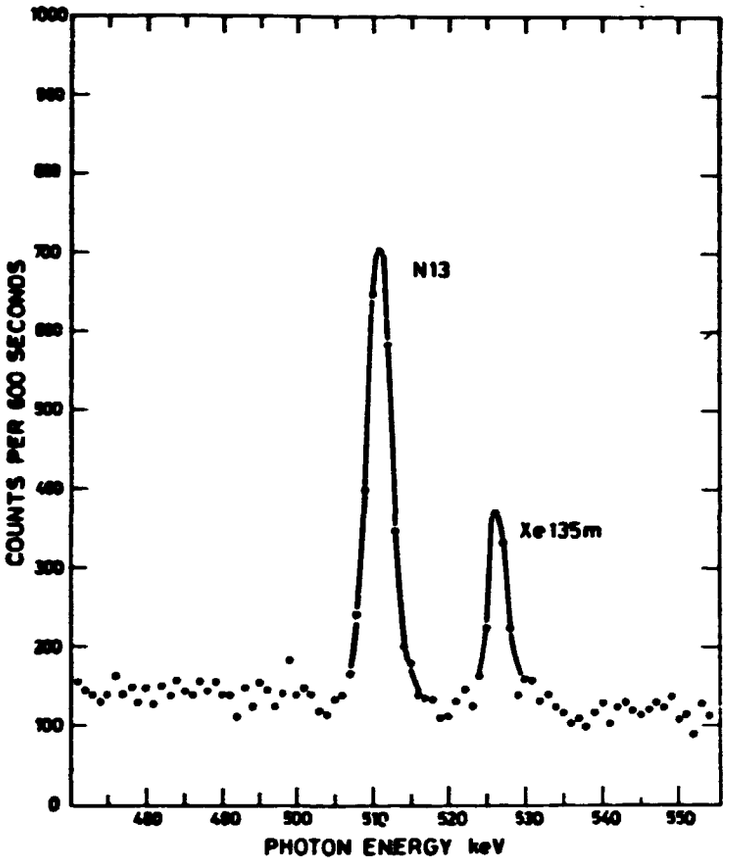

Figure 2.c

Spectrum, containment atmosphere 1978-11-01 at 15.00 hour.

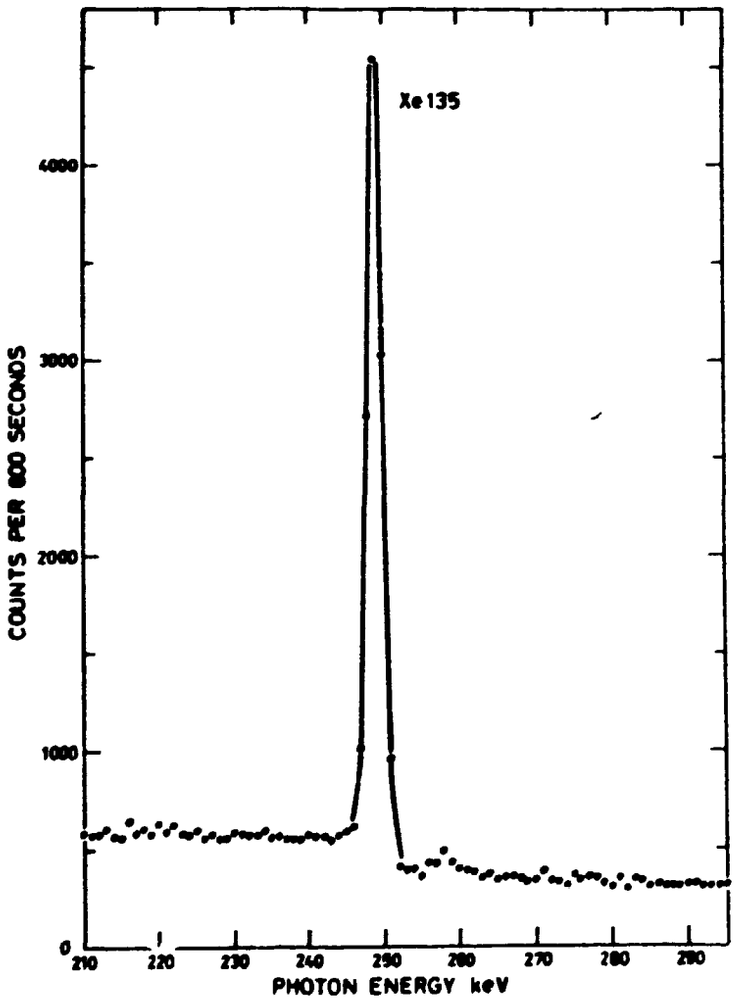

Figure 2.d

Spectrum, containment atmosphere 1978-11-01 at 15.00 hour.

Xe135m (527 keV)

$$n = \sum_{E=522}^{E=532} n_E - \sum_{E=519}^{E=521} n_E - \sum_{E=533}^{E=540} n_E \qquad \text{(Eq 2.c)}$$

The relative great number of signal channels used for the integration allows for a rather extensive peak drift. The allocation chosen of background channels to the high energy and the low energy side of the peak is understood from the plotted spectra, Figures 2.c and 2.c.

2.3 Background count rate

The detector assembly is shielded with 5 cm lead. The background count rate for a given peak is determined by contributions from the radiation level in the measuring room and from the compton level generated in the Ge(Li) detector by the total gamma spectrum from the containment sample air.

2.4 Statistics

For the R2 experiment, the background count rate around the N13 peak may be expressed:

$$n_B \approx n_s \cdot 0.55 + 200 \qquad \text{(Eq 2.d)}$$

in which n_B is the background count obtained in 600 seconds and n_s is the net count for the peak obtained in 600 seconds.

The fractional standard deviation for the determination of the content in the peak will be:

$$\sigma = \frac{\sqrt{2n_B + n_s}}{n_s} = \frac{\sqrt{400 + 2.1 n_s}}{n_s} \qquad \text{(Eq 2.e)}$$

Thus in order to determine the N13 peak content with a standard deviation σ = 10 %, the net count must be $n_s \approx 330$ cp 600 s.

General aspects on statistics and background are included in the discussion of the experimental results.

3. RADIOACTIVE ISOTOPES IN THE COOLANT

It may be assumed that the complete collection of radioactive isotopes present in the reactor water will be released to the containment during a coolant leakage.

According to the volatility and chemical properties of a specific substance and the atmospheric conditions in the containment it will be available for sampling as a gas or as an aerosol or it will deposit in solution or as a solid.

Thus a rough precipitation of non-volatile isotopes will occur at the boundary site for the release while chemical inactive gaseous acitivity (nitrogen and xenon) is transferred to the measuring station outside the containment.

A further precipitation of otherwise interfering isotopes takes place in the three coolers in the pump station (Figure 2a).

A condition for the use of N13 as a tracer isotope is that the level of annihilation radiation from other positron emitters may be neglected.

F18, a positron emitter with a half-life of 110 minutes, is produced in the reactor water in a secondary nuclear reaction ($O18+H1 \rightarrow F18+n$) similar to the one responsible for the N13 production.

Thus F18 would be useful as a coolant tracer element performing high sensitivity in long-time leakage situations. The water/steam distribution factor for fluor is high, however, and therefore the F18 activity is lost in the instrument coolers.

All other positron emitters produced with O15... O18 of the reactor water may be neglected on the basis of low production rate or short half-life.

For obvious reasons, none of the fission products in the reactor water including the noble gases and the halogens will be positron emitters.

Among the radioactive corrosion products typically found in the reactor water are three positron emitters: Co58, Cu64 and Zn65. It appears from experiences with nuclear power stations in operation, that only Cu64 has to be considered.

Information from Swedish BWR indicates that the maximum equilibrium level in the coolant to be expected is about 5 MBq kg^{-1}. This level of concentration is comparable to the N13 level 0.5 - 6 MBq kg^{-1} expected.

Generally the distribution factor water/steam is very high, however, (1000?). Therefore even if some fractional transport to the measuring station may occur, the substance in the form of an aerosol or in solved form in fog will effectively be precipitated in the instrument coolers/ filters.

3.1 N13 production rate

With the knowledge of

$\Sigma_e(E_n)$ the macroscopic neutron elastic scattering cross section (cm^{-1}/steradian)

$\varphi(E_n)$ the normalized neutron flux distribution $\left(\frac{\text{neutrons cm}^{-2}\ \text{s}^{-1}\ \text{MeV}^{-1}}{\text{integral fast neutron flux, neutrons cm}^{-2}\text{s}^{-1}}\right)$

E_{thr} the threshold energy for the O16+H1 → N13+He4 reaction (MeV)

$\Sigma_r(E)$ the macroscopic proton cross-section for the O16+H1 → N13+He4 reaction (cm^{-1}), and

$\frac{dE}{ds}(E)$ the energy loss rate of protons in H_2O, (MeV cm^{-1})

Σ_n the total macroscopic fission neutron cross section for the O16+H1 → N13+He4 reaction

(cm^{-1}, $\frac{\text{reactions } cm^{-3}\ s^{-1}}{\text{neutrons } cm^{-2}\ s^{-1}}$) may be calculated.

Lennart Hammer and Sture Forsén (1).

The figure reported in (1) is:

$$\Sigma_n = 1.35 \cdot 10^{-9}\ cm^{-1} \text{ (measured)}$$

and

$$\Sigma_n = 1.33 \cdot 10^{-9}\ cm^{-1} \text{ (calculated)}$$

The average fast neutron flux from 1 - 5 MeV (≈ 1 - ∞ MeV) has been reported to be (2) $5\ 04 \cdot 10^{13}$ n $cm^{-2}s^{-1}$ at 3250 MW(th), and the volume of coolant over which the average flux is effective has been calculated to be $2.21 \cdot 10^7$ cm^3.

Using these figures determining the total integrated fast neutron flux for a light water moderated reactor we get for the relative N13 production:

$$\left(\frac{A/t}{P_{th}}\right) = \Sigma_n \cdot \rho \cdot \lambda \cdot \frac{5.04 \cdot 10^{13} \cdot 2.21 \cdot 10^7}{3250 \cdot 10^6}\ \text{MBq MW}^{-1}\text{s}^{-1}$$

where

Σ_n = $1.34 \cdot 10^{-9}$ cm^{-1}

ρ = density of coolant ≈ 0.7

λ = decay constant for N13 = $1.16 \cdot 10^{-3}$ s^{-1}

resulting in

$$(\frac{A/t}{P_{th}}) = 0.37 \text{ MBq } MW^{-1}s^{-1}$$

Calculations carried out by Mike Sohan Singh and Lawrence Ruby (2) using a different approach for the determination of the proton flux density versus energy result in a figure

$$(\frac{A/t}{P_{th}}) = 0.85 \text{ MBq } MW^{-1}s^{-1}$$

while according to the same paper the production rate at the 700 MW(th) Dresden-1 reactor has been reported to be 0.36 MBq $MW^{-1}s^{-1}$. The experiment carried out 78-11-01 at the R2-reactor in Studsvik gave as a result for N13:

$$(\frac{A/t}{P_{th}}) = 0.43\text{-}0.46 \text{ MBq } MW^{-1}s^{-1}$$

In the present paper has consistently the figure $(\frac{A/t}{P_{th}})_{N13} = 0.5$ MBq $MW^{-1}s^{-1}$ for the relative N13 production rate been used, bearing in mind, however, that the proper figure may be found anywhere in the range 0.3 - 0.9.

The threshold energy for the O16+H1 → N13+He4 reaction is rather high (E_{thr} = 5.55 MeV). Thus only that part of the fast neutron spectrum in which elastic scattering in the moderator is predominant contributes to the generation of

N13. Consequently the figure 0.5 MBq $MW^{-1}s^{-1}$ should apply approximately for all thermal light water reactors exhibiting a reasonable low fast neutron leakage rate.

3.2 F18 production rate

From the same sources of information the range for the relative F18 production according to the reaction O18+H1 → F18+n (E_{thr} = 2.57 MeV) reported is estimated to be 0.014 - 0.025. In the present paper has consistently the figure $(\frac{A/t}{P_{th}})_{F18}$ = 0.02 MBq $MW^{-1}s^{-1}$ been used.

3.3 Release rates for noble gases

Only a few of the many fission noble gases released to the coolant due to fuel rod pinholes are useful as tracer elements for leakage detection purposes.

Kr87, T(0.5) = 76 minutes, 402 keV, 84 %
Xe135, T(0.5) = 9.1 hours, 250 keV, 91 %
Xe138, T(0.5) = 17 minutes, 258 keV, 40 %
Xe135m, T(0.5) = 16 minutes, 527 keV, 91 %

The release rate has to be high and the half-life time should be in the range 10 minutes-hours.

The measuring chamber is constructed for 1 500 kPa sample gas pressure in order to render it a high efficiency for the important N13 detection. The absorption of photons in the chamber wall and the general efficiency characteristic for the Ge(Li)-detector in practice limits the gamma detector energy range to 200 - 600 keV.

3.3.1 Xe138

In situations where the goal is to accomplish an analysis of the combined effect of fuel-rod and RCPB leakage the short-lived Xe138 may be of interest.

For a "normal" pin-hole situation (release rate 300 MBq s^{-1}) the detection statistics will be acceptable first at a coolant leakage level of about 10 kg/minute.

3.3.2 Kr87

With a lower release rate and a longer half-life time the Kr87 equilibrium concentration of activity in the containment atmosphere will be about the same as that obtained with Xe138. The resulting response time is much longer, however, and the release rate is much lower than that for Xe135.

3.3.3 Xe135m

At the time for the start of this work Xe135m was considered to be the favourite noble gas tracer isotope due to its short half-life time and apparently high release rate.

Later experiment revealed, however, that the effective response time did not correspond to the half-life time, T(0.5) = 16 minutes. Equilibrium was achieved first after several hours.

Obviously although the system is not responding directly to I135, this isotope, when trapped in the instrument coolers, must be taken into consideration as a supplementary source of Xe135m. The half time for the decay of I135 to Xe135m is 6.7 hours.

Thus, with this type of instrumentation, Xe135m may not be considered as a short-lived tracer isotope for RCPB detection work.

3.4 Release rate for Xe135

The calculation of the "normally expected" release of fission products to the coolant due to cladding damages is unfortunately of a rather speculative nature as it involves presumptions considering the pin-hole configuration and level as well as about a proper distribution model.

Knowledge of the production rate and the resulting inventory is of course highly valuable the other way around i.e. to form an idea of the cladding state from release measurements.

Information about the concentration of noble gases in the steam in operating BWR plants and information about the generally used design basis for the release to the coolant of Xe135 from 3000 MW(th) power reactors makes it reasonable to estimate a practical range for the release rate of Xe135:

$$2000 \text{ MBq s}^{-1} > (A/t)_{Xe135} > 20 \text{ MBq s}^{-1}$$

In this report we have consistently used the geometrical mean

$$(A/t)_{Xe135} = 200 \text{ MBq s}^{-1}$$

and consequently

$$\left(\frac{A/t}{P_{th}}\right)_{Xe135} = 0.067 \text{ MBq MW}^{-1} \text{ s}^{-1}$$

For Xe138 the figure:

$$(A/t)_{Xe138} = 300 \text{ MBq s}^{-1}$$

has been used.

4. CALCULATIONS FOR BWR REFERENCE PLANT

Notations

Name	Symbol	Value	Unit
Core thermal power	P_{th}	3 000	MW
Reactor coolant mass	m_c	330 000	kg
Coolant circulation flow	q_c	11 000	kg s^{-1}
Steam flow	q_s	1 600	kg s^{-1}
Coolant circulation time	t_c	30	s
Coolant leakage flow rate	q_l	4	1 min^{-1}
Decay constant of radioactive isotope	λ		s^{-1}
Radioactive isotope production or release rate	(A/t)		MBq s^{-1}
Radioactive isotope content at coolant inlet	$(A/m)_i$		MBq kg^{-1}
Radioactive isotope content in the steam	$(A/m)_s$		MBq kg^{-1}
Content of radioactive isotope in dry-well	$(A/V)_d$		kBq m^{-3}
Dry-well volume	V_d	5 600	m^{-3}
Distribution factor steam/water	(PF)		
Time after a step change of leakage flow rate	t_l		s

The following equations have been used for the calculations:

Radioactive isotope content at coolant inlet,

$$(A/m)_i = \frac{(A/t)}{q_c \left(\frac{q_c}{q_c - q_s\,(PF)} \exp(\lambda t_c) - 1\right)}, \text{ MBq kg}^{-1} \qquad \text{(Eq 4.a)}$$

where (PF) is the distribution factor steam/water.

Radioactive gaseous isotope content in the steam lines,

$$(A/m)_s = \frac{(A/m)_i\ q_c + (A/t)}{q_c}\ (PF),\ MBq\ kg^{-1}$$

(Eq 4.b)

Content of gaseous radioactive isotopes in the dry well,

$$(A/V)_d = \frac{(A/m)_i\ q_\ell\ 1000}{\lambda\ 60\ V_d}\ (1-\exp(-\lambda t_\ell)),\ kBq^{-3},$$

(Eq 4.c)

for coolant leakage and

$$(A/V)_d = \frac{(A/m)_s\ q_\ell\ 1000}{\lambda\ 60\ V_d}\ (1-\exp(-\lambda t_\ell)),\ kBq\ m^{-3},$$

(Eq 4.d)

for a leakage of steam.

q_ℓ is the coolant leakage mass flow or the steam leakage mass flow and t_ℓ is the time after a step increase of the leakage flow.

4.1 Calculations for N13

Decay constant $\lambda = 1.16 \cdot 10^{-3}\ s^{-1}$. With a production rate

$$\left(\frac{A/t}{P_{th}}\right)_{N13} = 0.5\ MBq\ MW^{-1}\ s^{-1}$$

we get for a 3 000 MWth BWR:

$(A/t) = 1500\ MBq\ s^{-1}$ and for the content of N13 in the reactor coolant at the inlet

$$(A/m)_i = \frac{1500}{11000\left(\frac{11000}{11000-(PF)1600}\exp(1.16\cdot10^{-3}\cdot30)-1\right)},$$

MBq kg^{-1}, (Eq 4.e)

With the steam production 1600 kg s^{-1} and the coolant circulation time 30 s the N13 balance for the content in the reactor water is nearby completely determined by the stripping with the steam.

It is assumed, that no nitrogen-13 will pass the condensate clean-up system. Thus the radioactive decay of N13 (T(0.5) = 10 minutes) is without importance, practically, for the balance.

The reactor water clean-up system for BWR has a typical flow rate of less than 10 % of the total reactor coolant mass in 10 minutes and therefore does also not influence the N13 balance.

The steam/water distribution factor (PF) used in (Eq 4.e) determines the transfer of nitrogen to the steam.

Ideally, with (PF) = 1, the detection sensitivity for a ccolant water leakage and a steam leakage will be approximately the same.

However, the effective value for (PF) depends upon the chemical state of the solved nitrogen (3, 4, 5).

The volatility of nitrogen solved in water is much lower for the anionic forms (NO_3^- and NO_2^-) than for the cationic form NH_4^+. When transferred to the steam a great part of the nitrogen is found in the form of neutral ammonia.

With the requirements normally set for BWR coolant water (pH ≈ 7, conductivity < 1 μS cm^{-1}), the value for (PF) is estimated to be in the range 1 - 2.

For the content of N13 in the steam lines we get

$$(A/m)_s = \frac{(A/m)_i \ 11000+1500}{11000} \ (PF), \ MBq \ kg^{-1}, \qquad \text{(Eq 4.f)}$$

while the content of N13 activity in the reactor dry-well is given by:

$$(A/V)_d = \frac{(A/m) \ 4 \cdot 1000}{1.16 \cdot 10^{-3} \cdot 60 \cdot 5600} \ (1-\exp(-1.16 \cdot 10^{-3} \cdot t_\ell)), \ kBq \ m^{-3}, \qquad \text{(Eq 4.g)}$$

where t_ℓ is the time (in seconds) after a step change of the leakage flow rate.

The equilibrium content of N13 (t_ℓ > 30 minutes) in the coolant, in the primary steam and in the dry-well resulting from a 4 kg min^{-1} coolant or steam leakage has been calculated and are given in Table 4.a.

Table 4.a

BWR 3000 MWth
Estimated equilibrium content of N13, F18 and Xe135 in the reactor coolant, in the steam lines and in the containment dry-well with a coolant or steam leakage of 4 kg min^{-1}

Isotope	N13	F18	Xe135
Half-life, T(0.5), minutes	10		
Half-life, T(0.5), hours		1.8	9.1
Isotope release or production rate, (A/t), MBq s^{-1}	1500	60	200
Equilibrium content of activity in the reactor coolant, $(A/m)_i$, MBq kg^{-1}	PF=1	PF=0.5	PF=1
	0.64	0.07	0.11
	PF=2	PF=0.2	PF=2
	0.30	0.16	0.044
Equilibrium content of activity in the steam lines, $(A/m)_s$, MBq kg^{-1}	PF=1	PF=0.5	PF=1
	0.78	0.04	0.13
	PF=2	PF=0.2	PF=2
	0.87	0.03	0.12
Equilibrium content of activity in the dry-well, leakage of coolant, $(A/V)_d$, kBq m^{-3}	PF=1	PF=0.5	PF=1
	6.6	7.9	62
	PF=2	PF=0.2	PF=2
	3.1	18	25
Equilibrium content of activity in the dry-well, leakage of steam, $(A/V)_d$, kBq m^{-3}	PF=1	PF=0.5	PF=1
	8.0	4.5	72
	PF=2	PF=0.2	PF=2
	8.9	3.4	70

4.2 Calculations for F18

Decay constant

$$\lambda = 1.05 \cdot 10^{-4}\ s^{-1}$$

Production rate

$$\left(\frac{A/t}{P_{th}}\right)_{F18} = 0.02 \cdot (A/t)_{F18} = 60\ MBq\ s^{-1}$$

As F18 like N13 is a positron emitter radiating 511 keV annihilation photons and as it has a rather long decay time (T(0.5) = 1.8 hours) it represents a hazard for the use of N13 as a leakage tracer element, permitting measurement with short response time.

Fortunately F18 in aqueous solution has a very low volatility and is therefore trapped very effectively in the instrument coolers.

Exact figures for the distribution factor steam/water for F18 are not available.

Information about the content of F18 in the feed water (before the condensate demineralizer) of operating BWR plants seems to indicate that the value of the effective (PF) is to be found in the range $0.2 < (PF) < 0.5$. Even with these figures for (PF), the effect on the F18 balance from the reactor water clean-up system may be disregarded.

The equilibrium content of F18 in the coolant, in the steam and in the dry-well resulting from a 4 kg min^{-1} RCPB leakage has been calculated and are given in Table 4.a.

Although the production rate for F18 is low (60 MBq s^{-1}) the resulting calculated content in dry-well is rather high, 7.9 - 18 kBq m^{-3}. This is due to the long half-life time for F18 (T(0.5) = 1.8 h). Redeposition on condensing surfaces and in containment coolers which will be very significant for the non-volatile F18 has not been considered. Again it should be emphasized also that the measuring system itself has a very low sensitivity for F18.

4.3 Calculations for Xe135

Decay constant, $\lambda = 2.11 \cdot 10^{-5} s^{-1}$.

The distribution factor steam/water (PF) for Xe to be used for the calculation of the transfer of the solved Xe to the vapour phase is probably rather close to 1.

As Xe is an inert gas, (PF) can reasonably not be smaller than 1.

Some diffusion from the water phase to the vapour phase during the transport of coolant through the reactor core is inevitable.

The high atomic number of Xe renders it a low diffusion coefficient however. Thus a very high value for (PF) is not to be expected.

The equilibrium content of Xe135 in the coolant, in the steam lines and in the dry-well as a result from a 4 kg min^{-1} RCPB leakage has been calculated using (PF) = 1 and (PF) = 2, Table 4.a. The Xe135 release rate to the water was presumed to be

$$(A/t)_{Xe135} = 200 \text{ MBq } s^{-1}.$$

Although the response time for Xe135 is rather long (50 % of the equilibrium value after 9.1 h) the content in the dry-well after only 1 hour will be about 7 % or 2 - 5 kBq m^{-3}, content = equilibrium content $(1-\exp(-2.11\cdot10^{-5}\cdot t_{\ell}))$.

It is noteworthy that although the content in the coolant reaches acceptable values, virtually all the Xe released to the coolant is removed with the steam.

5. CALCULATIONS FOR PWR REFERENCE PLANT

Notations

Name	Symbol	Value	Unit
Core thermal power	P_{th}	3 000	MW
Reactor coolant mass	m_c	220 000	kg
Coolant flow rate	q_c	13 000	kg s^{-1}
Let down clean-up flow rate	$q_{\ell d}$	5	kg s^{-1}
Coolant circulation time	t_c	17	s
Coolant leakage flow rate	q_l	4	kg min^{-1}
Decay constant of radioactive isotope	λ		s^{-1}
Radioactive isotope production or release rate	(A/t)		MBq s^{-1}
Radioactive isotope content at coolant inlet	$(A/m)_i$		MBq kg^{-1}
Content of radioactive isotope in dry-well	$(A/V)_d$		kBq m^{-3}
Dry-well volume	V_d	50 000	m^3
Time after change of leakage flow rate	t_l		s

While stripping with the steam during the boiling process to a great extent is determinant for the isotope balance in the coolant of boiling water reactors, the content in the reactor water of PWR is regulated by the radioactive decay of the elements or the decay due to chemical adsorption in the components of the primary system.

For N13, radioactive decay will probably be dominant while for the long-lived Xe135 and also for F18 trapping in the let down demineralizer and in the volume control tank is of importance.

Conservatively the total demineralizer filter efficiency is set to 100 % i.e. no impurity is expected to be recharged back to the reactor coolant system.

The following equations have been used for the PWR calculations:

For the calculation of the radioactive isotope content at coolant inlet,

$$(A/m)_i = \frac{(A/t)}{q_c \left(\frac{q_c}{q_c - q_{\ell d}} \exp(\lambda tc) - 1\right)} \text{ MBq kg}^{-1}$$

(Eq 5.a)

$$(A/m)_i = \frac{(A/t)}{13000 \left(\frac{13000}{13000-5} \exp(\lambda \cdot 17) - 1\right)} \text{ MBq kg}^{-1},$$

where $q_{\ell d}$ is the let down clean-up flow rate.

For the calculation of the content of gaseous radioactive isotopes in the dry-well,

$$(A/V)_d = \frac{(A/m)_i \, q_\ell \, 1000}{\lambda \, 60 \, V_d} (1 - \exp(-\lambda t_\ell)), \text{ kBq m}^{-3}$$

(Eq 5.b)

$$(A/V)_d = \frac{(A/m)_i \, 4 \cdot 1000}{\lambda \cdot 60 \cdot 50000} (1 - \exp(-\lambda t_\ell)), \text{ kBq m}^{-3}$$

for a 4 kg min^{-1} RCPB coolant leakage.

The results for N13, F18 and Xe135 with the same production or release rates as used in the BWR calculations are given in Table 5.a.

Table 5.a

PWR 3000 MWth
Estimated equilibrium content of N13, F18 and Xe135 in the reactor coolant and in the containment dry-well following a RCPB coolant leakage with a rate of 4 kg min^{-1}

Isotope	N13	F18	Xe135
Half-life, T(0.5), minutes	10		
Half-life, T(0.5), hours		1.8	9.1
Isotope release or production rate, (A/t), MBq s^{-1}	1500	60	200
Equilibrium content of activity in the reactor coolant, $(A/m)_i$, MBq kg^{-1}	5.7	2.1	21
Equilibrium content of activity in the dry-well, $(A/V)_d$, kBq m^{-3}	6.5	27	1300

The result for N13 is remarkably consistent with the result for BWR (6.5 and 3.1 - 6.6 kBq m^{-3} respectively) for coolant leakage.

The suppression of the N13 release to the BWR coolant due to stripping with the steam is comparable with the hold down of the relative content of N13 caused by the great containment volume of PWR.

The equilibrium content in the PWR dry-well for Xe135 is calculated to 1300 kBq m^{-3}. This rather high value may be related to the long half time (9.1 h) for Xe135 and the low let down flow rate (5 kg s^{-1}).

A modest supplementary trapping, i.e. to the pressurizer gas volume, corresponding to a flow of 10 kg s^{-1} will lower the figure to $(A/V)_d \approx$ 600 kBq m^{-3}.

Apparently the Xe135 content in the PWR reactor water will always be high enough to permit a RCPB leakage detection.

With a 4 kg min^{-1} leakage and with 5 kg s^{-1} let down flow rate we get a signal for the dry-well content after 1 hour:

$$(A/V)_d = 1300 \cdot 0.073 = 95 \text{ kBq } m^{-3}.$$

6. RCPB LEAKAGE EXPERIMENT AT THE R2-REACTOR, STUDSVIK

Notations

	Symbol	Value	Unit
R2-reactor			
Core thermal power	P_{th}	40	MW
Reactor coolant mass	m_c	123000	kg
Coolant circulation flow	q_c	1330	kg s^{-1}
Coolant circulation time	t_c	92	s
Ion exchange flow	q_i	60	kg s^{-1}
N13 experiment			
Coolant water transport time reactor core - sample input	t_s	3	min
Dose pump feed interval time	t_d	5	min
Coolant leakage flow rate	q_ℓ	0.004	kg min^{-1}
Total simulated containment volume	V_d	0.6	m^3
Take-up volume (tank + compressor + coolers etc)	V_c	0.45	m^3
Detector assembly volume, effective	[illegible]	0.15	m^3
Return-air flow rate for measuring system	q_m	0.1	kg min^{-1}
Isotope release or production rate	(A/t)		MBq s^{-1}
Radioactive isotope content in coolant (outlet)	$(A/m)_o$		MBq kg^{-1}
Equilibrium content of isotope in simulated drywell	$(A/V)_d$		kBq m^{-3}
Time after change of leakage flow rate	t_ℓ		s

6.1 Experimental set-up

The experimental set-up is shown in Figure 2.a.

The coolant sample tube is filled from a sample tap (PR 404) connected to the R2 primary water system in a point after the reactor core outlet and before the N16 decay tank.

A new sample tube is filled every 5 minutes and transported to the measuring site where it is connected to the dosage pump.

The mean transfer rate of coolant to the simulated reactor containment tank is set to 4 cc min^{-1} or 20 cc in each sample period.

The coolant is injected into the circuit by means of a flow-through evaporator with outlet to the bottom of the containment tank.

The temperature of the sample outlet vapour from the containment tank is regulated with an electrical heater in the tank and a coat in the form of a helical water-hose permits separate heating or cooling of the tank wall.

6.2 Calibration

The following corrections of the results according to the decay time for the isotopes has been used:

Correction for decay due to dose pump feed interval time t_d

$$\text{Correction factor} = \frac{\lambda \cdot t_d \cdot 60}{1-\exp(-\lambda \cdot t_d \cdot 60)}$$

Correction for decay due to sample transport time t_s

Correction factor = $\exp \lambda \cdot t_s \cdot 60$

Correction for decay due to transport delay in the measuring system (transport from containment simulator tank to detector assembly),

$$\text{Correction factor} = 1 + \frac{V_c V_m}{V_c + V_m} \cdot \frac{\lambda \cdot 60}{q_m \cdot 0.09}$$

The total correction factor and resulting calibration for the whole system is given in Table 6.a.

Table 6.a

Calibration of N13 experiment.
Correction for radioactive decay

Isotope	Correction	$(A/V)_d$
N13	1.59	kBq m^{-3} = 0.0056·cp 600 s
F18	1.04	kBq m^{-3} = 0.0038·cp 600 s
Xe135	1.01	kBq m^{-3} = 0.0040·cp 600 s

6.3 Parameters for the experiments

In both experiments reported in this paper (1978-10-25 and 1978-11-01) was the coolant leakage simulation flow 0.004 kg min^{-1} and the flow rate of the return air (practically dry air) to the containment 0.1 kg min^{-1}.

With this return-air flow rate, the maximum flow rate of water to the containment before saturation occurs is given in Table 6.b.

Table 6.b

Saturation coolant flow rate to the simulated containment tank for a 0.1 kg min^{-1} return-air flow rate in the measuring circuit

Containment temperature, ^{o}C	5	10	15	20	25	30	35	40	45	50	60
Saturation flow-rate of water to the containment tank, g min^{-1}	0.55	0.77	1.1	1.5	2.0	2.8	3.7	5.0	6.6	8.8	16

The experiment 1978-10-25 was carried out with a containment temperature 50^{o}C (wall temperature and containment outlet temperature). This corresponds to a relative humidity $\frac{4}{8.8} = 0.45$.

The containment temperature used with the experiment 1978-11-01 was 15^{o}C corresponding to an extensive condensation in the containment tank.

In both experiments condensation occured in the low-pressure after-cooler, simulating cold surfaces and coolers in the reactor dry-well.

6.4 Calculation of the expected equilibrium content of N13 in the R2 coolant and in the simulated containment

The relative low ion exchange flow 60 kg s^{-1} compared with the total reactor coolant mass, m_c = 123000 kg, makes it reasonable to assume that the N13 content in the reactor water is determined only by the production rate and the radioactive decay time.

The sample is taken at the reactor outlet and the following equation is applicable for the content of N13 in the coolant:

$$(A/m)_o = \frac{(A/t)}{q_c(1-\exp(-\lambda t_c))} \quad \text{(Eq 6.a)}$$

With a production rate

$$(A/t) = 40 \cdot 0.5 = 20 \text{ MBq s}^{-1}$$

we get

$$(A/m)_o = \frac{20}{1330(1-\exp(-1.16 \cdot 10^{-3} \cdot 92))} = 0.148 \text{ MBq kg}^{-1}$$

For the content of N13 in the simulated dry-well we get

$$(A/V)_d = \frac{(A/m)_o \, q_l \, 1000}{\lambda \cdot 60 \cdot V_d} = \frac{0.148 \cdot 0.004 \cdot 1000}{1.16 \cdot 10^{-3} \cdot 60 \cdot 0.6} =$$

$$= 14.2 \text{ kBq m}^{-3}$$

6.5 Calculation of the expected equilibrium content of F18 in the R2 coolant and in the simulated containment

If the ion exchanger is 100 % effective for F18 we get at the output from the core:

$$(A/m)_o = \frac{A/t}{q_c(1-\frac{q_c-q_i}{q_c})\exp(-\lambda t_c))}$$

and with $(A/t) = 0.02 \cdot 40 = 0.8$ MBq s^{-1} inserted:

$$(A/m)_o = \frac{0.8}{1330(1-\frac{1330-60}{1330}\exp(-1.05 \cdot 10^{-4} \cdot 92))}$$

$$(A/m)_o = 0.011 \text{ MBq kg}^{-1}$$

For the content in the dry-well we get:

$$(A/V)_d = \frac{(A/m)_o \, q_1 \, 1000}{\lambda \cdot 60 \cdot V_d} = \frac{0.011 \cdot 0.004 \cdot 1000}{1.05 \cdot 10^{-4} \cdot 60 \cdot 0.6} \approx 12 \text{ MBqm}^{-3}$$

(Eq 6.b)

6.6 Calculation of the expected equilibrium content of Xe135 in the R2 coolant and in the simulated containment

The fuel-rod leakage to the R2 reactor water is known to be of a much smaller magnitude than the one accepted with power plants.

The generation of fission products has probably its origin from uranium contamination of surfaces in contact with the water.

Thus the expected level of Xe135 cannot be calculated. However, we will use the measured value for the Xe135 content in the containment to calculate the production rate.

As the R2 demineralizer is operating without any off-gas procedure, the only water/gas barrier in the system is situated in the pressurizer tank. The escape of Xe135 by diffusion to this tank is considered to be of minor importance for the xenon balance. Thus we may write:

$$(A/m)_o = \frac{(A/t)}{q_c(1-\exp(-\lambda t_c))} = 0.39(A/t) \text{ MBq kg}^{-1}$$

(Eq 6.c)

and for the content in the simulated contaiment

$$(A/V)_d = \frac{(A/m)_o \, q_1 \cdot 1000}{\lambda \cdot 60 \cdot V_d} = \frac{(A/t) 0.35 \cdot 0.004 . 1000}{2.11 \cdot 10^{-5} \cdot 60 \cdot 0.6} \text{ kBq m}^{-3}$$

(Eq 6.d)

With the measured value for $(A/V)_d$ inserted in Equation 6.d, the relative production rate for Xe135 $(\frac{A/t}{P_{th}})$ may be calculated.

6.7 Experimental results

Each experiment had a duration of about 7 hours. The leakage flow was turned off after about 3 - 4 hours permitting a complete development of both the rising and falling edge of the time response for N13 (Table 6.d, 6.e and Figure 6.a, 6.b.

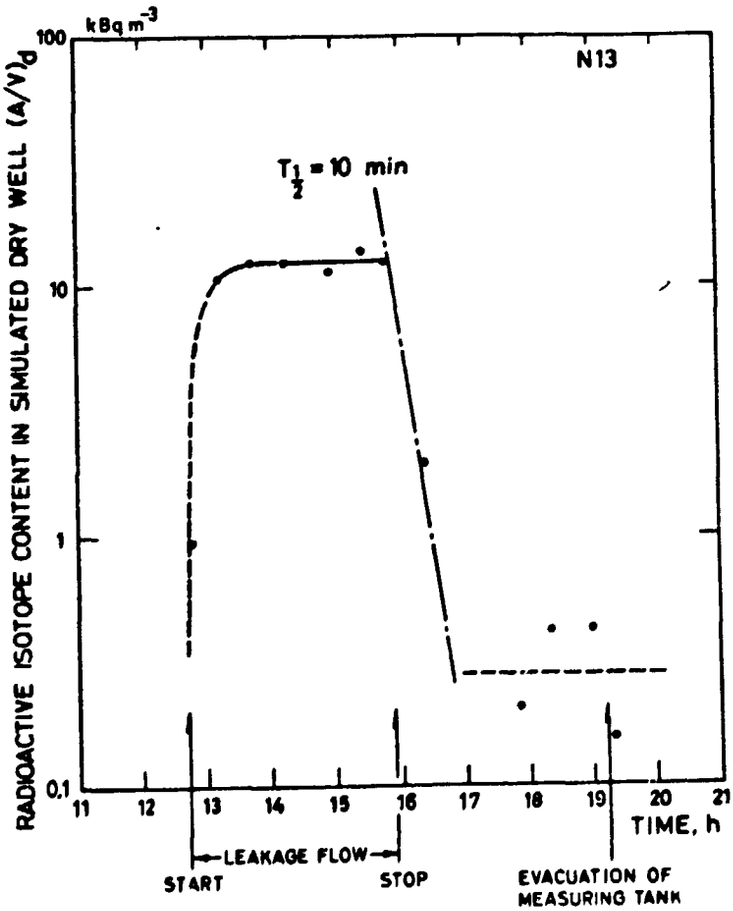

Figure 6.a

RCPB leakage detection experiment 1978-10-25
Isotope N13
Coolant flow to containment
Simulation tank, 0.004 kg min^{-1}
Simulated containment volume, 0.6 m^3
Simulated containment temperature, 50°C
R2-reactor core thermal power, 40 MW

A spectrum for N13 and Xe135 was taken every 30 minutes and stored in the computer memory for calculation of the content in the dry-well.

Several spectra covering the total energy range 100 keV to 750 keV were printed out for isotope and background control. One of them (15.00 h, 1978-11-01) is shown in Table 6.c. The spectra for N13, Xe135m and Xe135 are plotted in Figure 2.c and Figure 2.d.

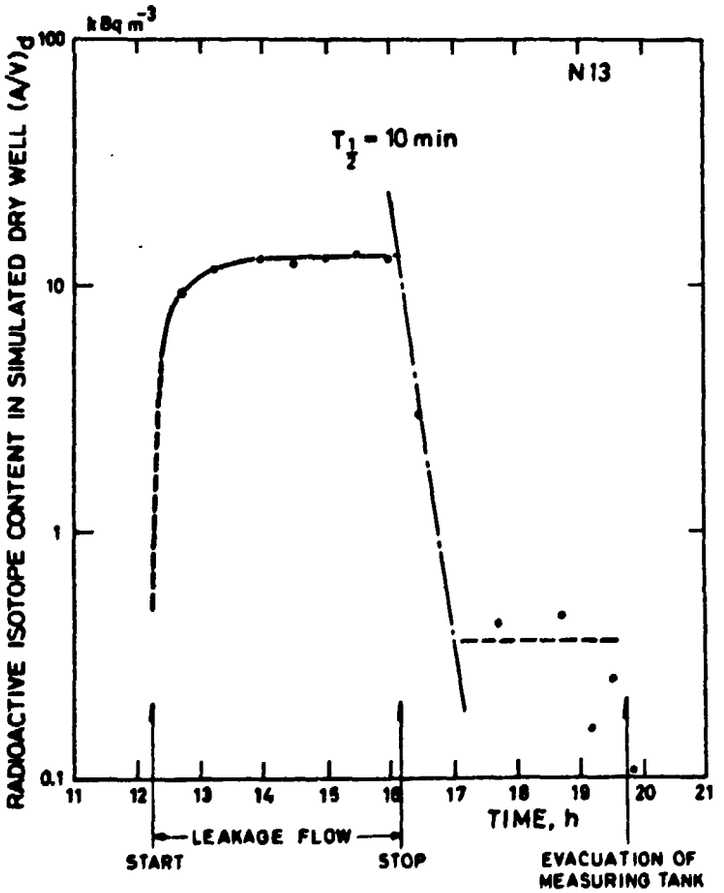

Figure 6.b

ACPB leakage detection experiment 1978-11-01
Isotope N13
Coolant flow to containment
Simulation tank, 0.004 kg min^{-1}
Simulated containment volume, 0.6 m^3
Simulated containment temperature, 15°C
R2-reactor core thermal power, 40 MW

The time response for N13 does apparently not differ much from the shape predictable from the decay time constant.

The N13 level after turn off of the leakage flow (measurement at 17.45 h and 17.50 h respecti.ely) is seen to be satisfactory low. Thus the trapping of F18 in the instrument coolers may be considered effective.

Kr87 with 402 keV and Xe138 with 258 keV are clearly identified on Table 6.c although the count-rates have been very low.

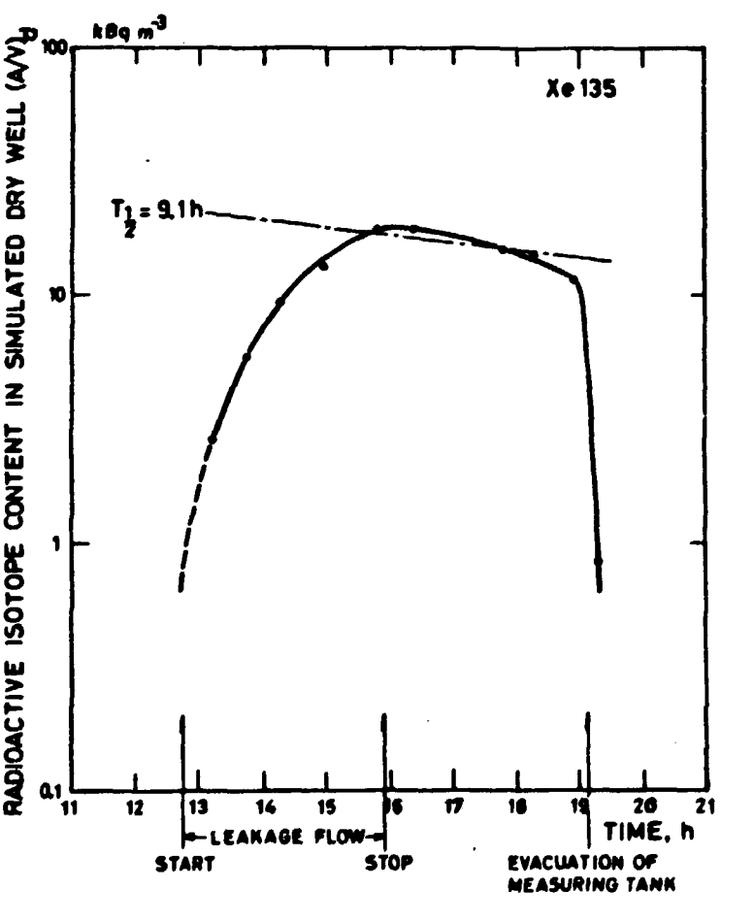

Figure 6.c

RCPB leakage detection experiment 1978-10-25
Isotope Xe135
Coolant flow to containment
Simulation tank, 0.004 kg min^{-1}
Simulated containment temperature, 50°C
R2-reactor core thermal power, 40 MW

The effective half-life time for Xe135 (Table 6.f, 6.g and Figure 6.c, 6.d) is somewhat shorter than the nuclear decay time (T(0.5) = 9.1 h) indicating probably some escape from the instrument system to the surroundings.

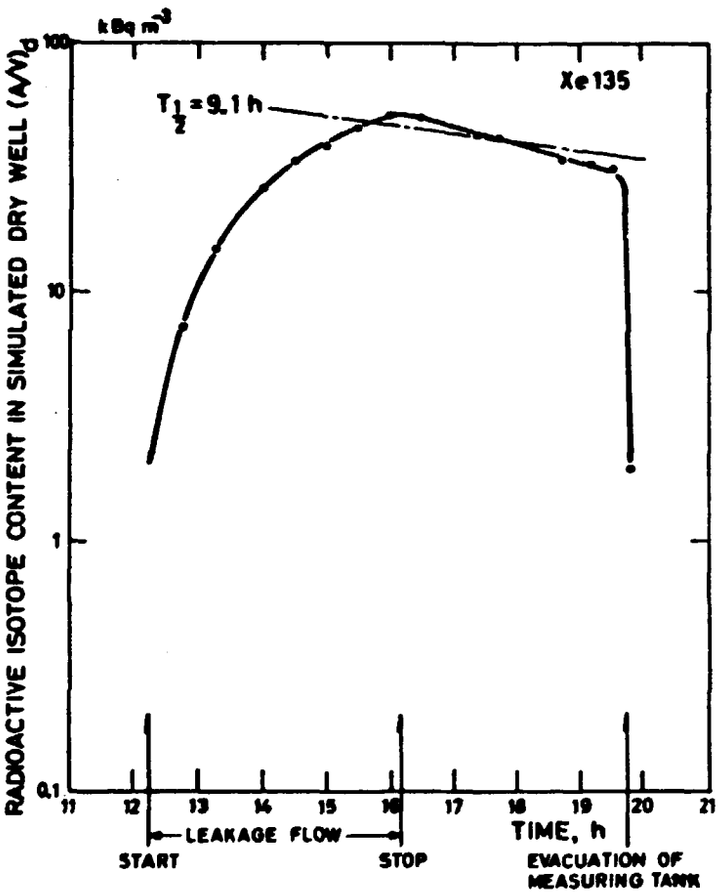

Figure 6.d

RCPB leakage detection experiment 1978-11-01
Isotope Xe135
Coolant flow to containment
Simulation tank, 0.004 kg min^{-1}
Simulated containment volume, 0.6 m^3
Simulated containment temperature, 15 °C
R2-reactor core thermal power, 40 MW

The experimental results are summarized in Table 6.h from which it appears:

- The N13 relative production rate has been measured to 0.43 and 0.46 MBq MW^{-1} s^{-1} respectively.
- Although the relative humidity in the containment tank was only 0.45 during the experiment 1978-10-25, while the experiment 1978-11-01 was carried out under conditions resulting in extensive condensation, the results do not differ much, $(A/V)_d$ = 12 and $(A/V)_d$ = 13 kBq m^{-3}.
- The sensitivity for F18 is sufficiently low.
- The pin-hole level for R2 is much lower than the one to be expected with a power reactor.

Table 6.c

RCPB leakage detection experiment 1978-11-01.
Gamma spectrum at 15.00 hour. Counts per 600 seconds.

E,keV	100	150	200	250	300	350	400	450	500	550	600	650	700
00	656	993	560	3027	269	217	194	176	137	106	119	117	98
01	663	911	587	968	270	215	256	146	147	115	113	98	100
02	689	738	609	402	313	219	303	143	138	89	109	102	93
03	693	745	585	380	297	217	303	160	117	127	123	108	95
04	681	715	568	395	305	205	214	138	114	113	123	103	87
05	738	690	566	354	284	205	174	162	134	120	125	109	106
06	724	701	574	433	295	193	173	162	138	115	112	106	103
07	697	676	542	431	270	239	191	138	166	101	129	111	85
08	698	659	547	482	283	180	175	138	237	120	164	92	110
09	683	713	576	433	272	226	179	143	396	110	156	103	113
10	718	703	578	397	291	206	150	148	646	122	110	114	95
11	706	703	576	382	298	211	179	167	701	92	110	119	106
12	757	657	574	376	287	218	141	187	582	129	114	90	97
13	712	682	602	353	261	211	175	164	347	128	104	122	100
14	671	644	566	368	269	216	182	157	200	93	107	81	92
15	739	663	567	337	266	218	159	169	180	106	121	127	115
16	737	666	644	343	265	186	157	144	137	107	105	117	100
17	744	614	586	354	257	206	180	137	133	102	101	122	118
18	714	674	595	343	259	228	169	140	132	122	101	117	97
19	807	634	568	324	238	188	139	142	110	125	115	113	97
20	783	682	624	338	263	188	149	135	111	101	111	100	118
21	772	697	589	380	267	198	155	154	132	122	119	123	110
22	782	639	621	332	260	212	171	144	145	113	134	105	102
23	786	651	583	322	252	186	156	136	125	130	97	106	121
24	734	664	577	307	217	179	171	128	163	94	103	88	96
25	745	658	595	350	264	192	124	137	224	147	113	118	114
26	773	675	557	332	250	187	151	161	370	120	122	102	107
27	761	647	577	357	224	178	170	139	333	124	117	103	118
28	809	602	552	349	261	207	180	147	223	101	103	114	106
29	812	660	556	316	241	186	169	128	136	102	100	109	103
30	800	639	583	308	259	182	142	148	159	126	111	109	106
31	822	646	578	353	241	206	164	126	157	119	111	99	96
32	786	642	572	289	234	175	160	149	130	116	101	112	116
33	850	597	566	344	251	199	198	136	136	119	111	109	96
34	782	621	590	322	262	179	204	156	123	118	122	114	102
35	818	621	556	305	233	201	200	142	117	115	110	105	92
36	764	598	559	312	253	189	179	138	102	101	103	113	103
37	750	568	550	304	275	162	147	155	108	129	94	97	102
38	803	601	547	304	254	176	154	141	99	121	115	104	106
39	811	567	542	302	232	201	168	154	116	121	111	116	113
40	721	629	575	314	205	210	146	137	129	90	100	103	112
41	791	605	571	309	251	192	145	137	101	129	114	125	93
42	746	604	562	294	212	198	162	110	123	110	96	113	89
43	794	631	541	299	198	180	152	148	129	123	101	97	93
44	715	640	578	305	205	205	134	123	118	100	102	122	106
45	787	797	590	301	248	191	158	155	112	114	99	98	103
46	764	789	615	282	230	215	153	144	119	112	100	106	98
47	787	693	1018	293	193	184	141	122	128	104	106	106	107
48	780	583	2716	294	213	170	139	139	124	114	89	118	93
49	829	613	4532	291	220	168	150	182	135	119	94	130	90

Table 6.d

RCPB leakage detection experiment 1978-10-25
Isotope, N 13

Calibration, kBq m^{-3} = 0.0056·cp 600 s
Temperature of simulated containment, 50°C
Coolant leakage flow, 0.004 kg min^{-1}

$$\text{integration, } n = \sum_{E=506}^{E=516} n_E - \sum_{E=499}^{E=505} n_E - \sum_{E=517}^{E=520} n_E$$

E, keV

496	50	92	122	126	136	151	95	59	56	34	21
497	56	83	95	130	134	140	117	60	44	42	17
498	51	83	103	134	146	135	116	67	42	36	15
499	62	95	108	149	140	132	107	60	44	43	18
500	61	55	92	137	141	139	105	70	50	32	15
501	50	85	121	108	122	123	117	63	58	30	19
502	54	89	122	134	118	129	122	58	53	38	17
503	47	115	122	124	137	152	117	46	51	34	16
504	49	85	120	126	134	155	124	58	41	33	18
505	67	98	99	128	147	143	128	51	55	39	14
506	76	105	106	149	166	143	119	53	38	46	14
507	88	140	146	179	64	176	126	58	48	38	18
508	188	187	230	218	280	258	128	57	58	41	16
509	254	349	372	363	394	384	148	46	52	39	12
510	378	502	543	545	553	512	181	60	67	49	16
511	449	587	606	611	631	648	182	80	58	57	22
512	435	525	519	517	562	577	176	73	62	44	28
513	281	369	371	356	472	398	128	70	64	35	22
514	162	206	193	212	279	265	155	46	55	41	20
515	89	119	134	131	182	159	104	64	63	33	22
516	51	83	126	116	146	146	101	57	37	44	18
517	28	79	81	118	122	132	84	57	51	41	19
518	50	73	91	110	109	129	101	58	44	33	15
519	47	81	97	137	128	132	98	60	43	38	19
520	39	80	93	109	98	134	105	48	40	33	11
521	43	69	105	105	105	125	89	53	42	39	19
522	51	77	103	98	133	133	107	43	45	33	13
523	39	80	105	104	166	152	98	48	53	35	18
524	51	84	123	141	171	160	129	59	35	44	19
525	74	97	141	175	241	215	152	68	68	47	16
526	78	135	171	228	266	319	188	88	90	47	20

n, cp600 s	1897	2197	2200	2017	2433	2166	340	35	72	73	27	
$(A/V)_d$, kBqm^{-3}	10.6	12.3	12.3	11.3	13.6	12.1	1.9	0.2	0.4	0.4	0.2	
time	12.45	13.16	13.47	14.18	15.00	15.30	15.50	16.25	17.50	18.20	19.00	19.20

start ◄——— leakage flow ———► stop ↑ 0.1 MPa

Table 6.e

RCPB leakage detection experiment 1978-11-01
Isotope, N 13

Calibration, kBq m^{-3} = 0.0056·cp 600 s
Temperature of simulated containment, 15°C
Coolant leakage flow, 0.004 kg min^{-1}

$$\text{Integration, } n = \sum_{E=506}^{E=516} n_E - \sum_{E=499}^{E=505} n_E - \sum_{E=517}^{E=520} n_E$$

E, keV

496	45	72	86	134	144	157	166	123	72	55	46	40	33	19
497	48	84	90	124	122	168	157	116	80	41	49	41	31	13
498	60	83	119	144	139	155	167	127	67	56	44	32	39	13
499	43	89	123	157	182	138	168	121	72	67	48	56	43	15
500	49	80	101	140	137	158	146	135	65	82	54	46	40	16
501	59	73	96	114	147	140	158	108	69	64	56	49	32	21
502	58	74	108	119	138	148	162	121	61	57	43	56	56	14
503	45	67	109	139	117	151	149	109	68	48	57	37	38	17
504	44	74	104	132	114	140	190	112	53	56	50	44	32	20
505	50	78	119	120	134	149	143	128	77	50	49	46	38	13
506	54	80	119	147	138	146	174	119	59	59	58	54	33	18
507	82	117	149	170	166	189	193	140	60	54	54	52	36	13
508	115	169	208	254	237	261	259	154	67	35	54	48	33	16
509	248	312	343	376	396	402	431	171	82	77	51	44	48	18
510	382	561	586	598	646	645	689	247	99	71	66	58	61	22
511	518	621	711	685	701	788	781	264	89	65	75	56	60	35
512	373	482	585	568	582	594	611	221	77	81	60	49	46	12
513	192	276	303	329	347	330	297	156	57	61	52	41	43	24
514	92	124	170	179	200	231	216	118	67	52	62	44	30	19
515	53	76	125	147	180	155	156	147	71	58	45	28	38	14
516	40	55	105	142	137	141	144	119	62	50	54	37	53	10
517	39	63	105	130	133	124	146	120	58	54	52	39	51	20
518	48	84	100	106	132	149	136	130	64	69	45	33	40	16
519	46	78	106	130	110	131	157	111	71	55	44	47	29	15
520	36	66	94	115	111	122	133	122	59	61	54	32	53	19
521	59	90	94	121	132	131	142	119	67	49	42	39	46	16
522	51	61	129	124	145	134	119	125	80	51	57	42	36	19
523	36	88	106	122	125	151	155	122	67	53	36	41	36	13
524	53	71	109	139	163	162	172	142	79	56	60	44	43	18
525	59	114	160	194	224	241	258	227	102	79	46	57	41	19
526	85	163	238	307	370	347	432	373	123	109	91	84	67	12

n, cp600 s	1632	2067	2238	2193	2275	2332	2263	519	73	0	79	26	42	15
$(A/V)_d$, kBqm^{-3}	9.1	11.6	12.5	12.3	12.7	13.1	12.7	2.9	0.4	0	0.4	0.1	0.2	0.1
time	12.15	12.45	13.15	14.00	14.30	15.00	15.30	16.00	16.30	17.45	18.15	18.45	19.12	19.32	19.50

start ◄——— leakage flow ———► stop ↑ 0.1 MPa

Table 6.f

RCPB leakage detection experiment 1979-10-25
Isotope, Xe135

Calibration, kBq m^{-3} = 0.040·cp 600 s
Temperature of simulated containment, 50°C
Coolant leakage flow, 0.004 kg min^{-1}

Integration, n = $\sum_{E=245}^{E=255} n_E - \sum_{E=240}^{E=244} n_E - \sum_{E=256}^{E=261} n_E$

E,keV

235	186	304	343	440	493	533	384	212	190	142	55
236	176	333	392	454	474	477	435	217	184	134	55
237	186	306	346	445	464	520	364	226	201	166	73
238	212	294	385	452	456	491	394	212	185	154	59
239	187	283	357	426	462	484	385	216	211	146	50
240	179	315	375	422	446	491	364	214	204	170	64
241	192	310	379	364	486	496	334	200	152	141	59
242	185	337	359	396	484	497	395	203	185	125	55
243	203	295	377	433	485	530	387	220	179	140	57
244	184	331	336	402	468	474	377	214	196	127	55
245	188	291	356	421	469	485	375	226	189	146	57
246	206	324	367	470	492	546	391	219	187	161	49
247	230	387	477	592	732	792	525	393	333	194	63
248	287	568	782	971	1207	1302	1167	862	766	537	80
249	372	750	1059	1268	1476	1733	1685	1344	1287	1123	109
250	372	693	939	1311	1483	1639	1567	1250	1145	1056	119
251	307	556	715	897	1151	1187	942	697	645	421	83
252	189	354	444	522	610	675	407	243	265	151	57
253	163	242	336	361	395	454	321	199	140	113	50
254	149	255	294	359	375	365	330	154	120	95	47
255	173	244	317	336	390	454	294	146	144	97	45
256	194	309	319	385	402	451	268	158	136	110	50
257	194	288	334	408	379	459	269	153	122	119	41
258	189	292	352	401	466	442	290	161	131	107	60
259	172	295	346	374	475	449	288	162	128	102	50
260	158	254	305	366	412	442	284	149	124	98	36
261	147	255	307	309	373	370	260	140	131	88	29
262	148	247	305	334	353	361	278	154	126	92	41
263	146	226	276	331	358	362	245	123	129	85	54
264	142	233	265	314	368	352	275	147	124	86	46
265	175	251	273	300	368	351	283	169	123	91	60
n,cp 600 s	639	1383	2297	3248	3904	4531	4488	3739	3533	2767	203
$(A/V)_d$,kBqm^{-3}	2.6	5.5	9.2	13.0	15.6	18.1	18.0	15.0	14.1	11.1	0.8
time 12.45	13.16	13.47	14.18	15.00	15.30	15.50	16.25	17.50	18.20	19.00	19.20

start ← leakage flow → stop 0.1 MPa

Table 6.g

RCPB leakage detection experiment 1978-11-01
Isotope, Xe 135

Calibration, kBq m^{-3} = 0.040·cp 600 s
Temperature of simulated containment, 15°C
Coolant leakage flow, 0.004 kg min^{-1}

Integration, n = $\sum_{E=245}^{E=255} n_E - \sum_{E=240}^{E=244} n_E - \sum_{E=256}^{E=261} n_E$

E, keV

235	208	301	433	539	556	542	658	544	380	265	246	189	69
236	201	298	425	482	559	629	651	511	327	218	228	198	70
237	200	348	434	534	550	602	636	562	319	229	251	212	52
238	190	301	423	506	547	580	621	539	322	231	227	200	46
239	179	312	416	481	542	607	622	485	322	250	239	205	68
240	189	300	422	521	575	564	657	506	313	233	240	219	47
241	200	301	410	464	571	612	620	516	309	266	202	212	55
242	195	296	418	527	562	579	607	498	317	242	218	213	59
243	188	274	427	484	541	597	635	503	316	244	214	190	58
244	171	311	409	495	578	588	634	534	332	266	221	212	62
245	194	271	422	533	590	583	602	544	335	263	241	211	53
246	199	321	451	540	615	648	697	610	393	326	255	265	36
247	275	475	753	905	1018	1145	1219	1191	859	551	570	503	79
248	575	1097	1890	2315	2716	3156	3465	3274	2600	1971	1970	1717	124
249	967	1887	3178	3847	4532	5250	5808	5445	4336	3547	3466	3281	226
250	691	1285	1988	2608	3027	3444	3872	3517	2821	2525	2268	2361	231
251	285	476	766	923	968	1167	1188	1165	794	687	600	662	132
252	151	251	389	409	402	476	476	389	234	181	164	152	59
253	167	231	331	354	380	451	449	349	206	122	127	106	57
254	164	218	275	384	395	401	421	326	203	141	111	115	31
255	164	252	327	362	354	403	456	347	189	142	115	105	47
256	167	237	328	405	433	460	464	321	212	122	108	94	40
257	220	293	433	450	431	472	499	374	192	144	116	101	55
258	222	300	445	461	482	520	523	363	182	122	124	113	45
259	174	265	326	373	433	466	449	334	176	123	110	94	54
260	157	241	300	355	397	379	445	335	167	119	119	94	41
261	148	228	310	339	382	388	438	330	186	135	123	119	62
262	154	225	299	348	376	407	436	338	162	157	104	124	53
263	138	217	294	340	353	351	370	339	219	147	134	90	48
264	148	203	303	354	368	365	371	341	166	126	125	94	40
265	136	209	294	302	337	372	436	319	166	126	120	94	48
n, cp600 s	1801	3738	6522	8304	9612	11499	12682	12543	10268	8440	8092	7816	489
$(A/V)_d$,kBqm^{-3}	7.2	15.0	26.1	33.2	38.4	46.0	50.7	50.2	41.1	33.8	32.4	31.3	1.96
time 12.15	12.45	13.15	14.00	14.30	15.00	15.30	16.00	16.30	17.45	18.45	19.12	19.32	19.50

start ← leakage flow → stop 0.1 MPa

Table 6.h

Results from RCPB leakage experiment at the R2 Research Reactor, Studsvik, Sweden.
Calculation of expected equilibrium content of N13 and F18 in simulated containment

Isotope	N13	F18	Xe135
Half-life, T(0.5), minutes	10		
Half-life, T(0.5), hours		1.8	9.1
Production rate, calculated, (A/t), MBq s^{-1}	20	0.8	?
Equilibrium content of activity in the reactor coolant, calculated, $(A/m)_0$, MBq kg^{-1}	0.15	0.011	
Equilibrium content of activity in simulated dry-well, calculated, $(A/V)_d$, kBq m^{-3}	14	12	
Equilibrium content of activity in simulated dry-well, $(A/V)_d$, kBq m^{-3}			
measurement, R2, 1978-10-25	12	<0.3	85
measurement, R2, 1978-11-01	13	<0.3	200
Relative production or release rate of isotope in R2 coolant calculated on the basis of the measurements, $(\frac{A/t}{P_{th}})$, MBq $MW^{-1}s^{-1}$			
measurement, 1978-10-25	0.43		0.001
measurement, 1978-11-01	0.46		0.002

7. DISCUSSION

N13

The leakage flow rate used at the experiments (0.004 kg min^{-1}, Table 4.a and 5.a) corresponds to a coolant or steam RCPB leakage flow rate

BWR	8 - 18 ≈	10 kg min^{-1} coolant
	6 - 7 ≈	6.5 kg min^{-1} steam
PWR	≈	8.5 kg min^{-1} coolant

With a coolant leakage rate of 4 kg min^{-1} the net count, n, obtained in 600 seconds and the fractional standard deviation for the determination of the N13 activity content in the containment atmosphere, σ, will be (Table 6.d and 6.e)

BWR	$n \approx 1000$, $\sigma \approx 5$ %
PWR	$n \approx 1200$, $\sigma \approx 4.5$ %

while the figures to be expected for a 10 kg min^{-1} leakage will be

BWR	$n \approx 2500$, $\sigma \approx 3$ %
PWR	$n \approx 3000$, $\sigma \approx 2.5$ %

The ultimate low-level limit for the use of the system ($\sigma \approx 10$ %) will be at a leakage flow rate about 1 kg min^{-1}.

Xe135

With the estimated release rate (A/t) = 200 MBq s^{-1} for Xe135 used in the calculations for 3 000 MWth power plants (Table 4.a, 5.a) the time response obtained in the experiments (Figure 6.c, 6.d may be considered as representative for a BWR with a

10 kg min^{-1} coolant leakage rate. The equilibrium content in the BWR containment for a 4 kg min^{-1} leakage was calculated to 25 - 62 kBq m^{-3} (Table 4.a), while the measured content of Xe135 in the experiments was 85 - 200 kBq m^{-3} (Table 6.h).

However, release rates of less than 1 MBq s^{-1} have been reported for BWR with low pin-hole levels.

For a BWR reactor in this condition the equilibrium net count for the Xe135 peak will only be about n = 100, a figure which is to low to achieve a reasonable statistics.

For a PWR plant the estimated content of Xe135 in the containment was calculated to 1300 kBq m^{-3} (Table 5.a).

Thus Xe135 may be used for RCPB leakage detection systems in PWR even if the fuel leakage level is very low (i.e. < 1 MBq s^{-1}).

However, also in a BWR plant with a "normal" fuel leakage level or generally in situations with high RCPB leakage rates, Xe135 may prove to be useful as a tracer isotope. It should be emphasized, that Xe135 has the advantage of a long half-life time (9.1 h). This could be of importance for the follow-up of a leakage event after reactor shut-down.

7.1 Sampling strategy

The course of the distribution of N13 over the dry-well volume as a result of a RCPB leakage has not been investigated in this work. All calculations for the isotope content in the

dry-well has been carried out under the assumption of an instantaneous complete mixture of the gas in the containment.

However, with the application of a selective sampling procedure from a pattern of sampling points, the detection efficiency and probably also the time response may be improved.

Specific areas in dry-well may be assigned a higher priority than others in a sequental sampling system.

7.2 Background shielding

Only gaseous activity is presented to the Ge(Li) detector assembly. This allows a great freedom in the choice of equipment disposal outside the containment, thus facilitating background shielding.

7.3 Contamination

Deposition of long-lived activity in sample tubes is not representing a threat to the system. Improper release of particulates from the tube walls has no chance of passing the instrument filters and reach the measuring chamber.

The effect of deposition of long-lived noble gas daughters inside the measuring chamber has been studied extensively. There seems to be no risk that a prohibitory increase of the general background will occur in a reasonable time.

8. CONCLUSIONS

The use of N13 and Xe135 as tracer isotopes for a RCPB leakage detection system has been demonstrated with experiments at the R2 research reactor at Studsvik, Sweden.

The results obtained have been used for the calculation of the performance to be expected with the system installed in a typical BWR or PWR plant.

A coolant leakage to the containment with a rate of 4 kg min^{-1} (as stated in the USNRC Regulatory Guide 1.45) will according to these calculations result in a detector signal with a 5 % fractional standard deviation.

Apprehensions considering the risk for interference from positron emitters other than N13 may probably be dismissed.

The general principle of the system makes it tolerable to the atmospheric conditions in the containment and resistant against interference from depositing particulates.

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REACTOR COOLANT PRESSURE BOUNDARY LEAKAGE DETECTION SYSTEM

Final report SKI project B28/76

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