nagra

National Cooperative for
the Disposal of Radioactive Waste

TECHNICAL REPORT 91-07

DETERMINATION OF THE ^{36}CL CONTENT IN REACTOR COOLING WATER AND ACTIVE RESINS FROM SWISS NUCLEAR POWER PLANTS

APRIL 1991

LIU XINQI*, H. W. GÄGGELER, D. LASKE [1]
H. SYNAL, W. WÖLFLI [2]
F. C. J. BRANDT [3]
J. C. ALDER, K. KURTZ [4]

[1] PSI, CH-5232 Villigen-PSI

[2] Institut für Mittelenergiephysik,
ETH, CH-8093 Zürich

[3] KKW Gösgen-Däniken AG
CH-4658 Däniken

[4] NAGRA, CH-5430 Wettingen

* On leave from Peking University,
Beijing, P.R. China

nagra

TECHNICAL

REPORT 91-07

DETERMINATION OF THE ^{36}CL CONTENT IN REACTOR COOLING WATER AND ACTIVE RESINS FROM SWISS NUCLEAR POWER PLANTS

APRIL 1991

LIU XINQI*, H. W. GÄGGELER, D. LASKE [1]
H. SYNAL, W. WÖLFLI [2]
F. C. J. BRANDT [3]
J. C. ALDER, K. KURTZ [4]

[1] PSI, CH-5232 Villigen-PSI

[2] Institut für Mittelenergiephysik,
ETH, CH-8093 Zürich

[3] KKW Gösgen-Däniken AG
CH-4658 Däniken

[4] NAGRA, CH-5430 Wettingen

* On leave from Peking University,
Beijing, P. R. China

ABSTRACT

^{36}Cl concentration was determined in reactor cooling water and in Reactor Coolant Cleanup System (RCCS) resins from a Swiss pressurized water reactor (PWR). For detection of ^{36}Cl, both accelerator mass spectroscopy (AMS) and liquid scintillation counting (LSC) techniques were used. In addition, the ^{36}Cl content in resins from a second PWR and also from a boiling water reactor were determined. In all samples the concentration of the key nuclide ^{137}Cs was also measured, which allowed the scaling factors for ^{36}Cl relative to ^{137}Cs to be determined. The resulting scaling factors for the resin samples are respectively $[1.4, 2.2$ and $17]x10^{-6}$, in reasonable agreement with the single predicted value of $6.0x10^{-6}$ used so far by Nagra for estimating the ^{36}Cl content of the resins from different reactors.
Measured and previously estimated ^{36}Cl concentrations lie within a factor 2.4.
For the water sample from the PWR, the measured scaling factor was $5.8x10^{-5}$.

ZUSAMMENFASSUNG

Im Kühlwasser und in Ionenaustauscherharzen eines schweizerischen Druckwasserreaktors wurde ^{36}Cl bestimmt. Für den Nachweis von ^{36}Cl wurde sowohl die Flüssigszintillationstechnik wie auch Beschleunigermassenspektrospie verwendet. Zudem wurde auch der ^{36}Cl-Gehalt in Harzen eines zweiten Druckwasserreaktors wie auch eines Siedewasserreaktors bestimmt. In allen Proben wurde auch die Aktivität des Schlüsselnuklids ^{137}Cs bestimmt. Dadurch konnten die Korrelationsfaktoren für ^{36}Cl relativ zu ^{137}Cs erhalten werden. Die resultierenden Korrelationsfaktoren für die Harze waren [1.4, 2.2 und 17]x10^{-6}, in recht guter Uebereinstimmung mit dem einzigen vorhergesagten Wert von 6x10^{-6}, der bisher von der Nagra für die Abschätzung von ^{36}Cl-Gehalten in Harzen verwendet wurde.
Der gemessene und der abgeschätzte Wert liegen somit innerhalb eines Faktors 2.4.
Für die Wasserprobe des Druckwasserreaktors betrug der experimentelle Korrelationsfaktor 5.8x10^{-5}.

RESUME

La concentration en ^{36}Cl dans l'eau de refroidissement et dans les résines du système de purification de cette eau a été mesurée dans un réacteur suisse à eau pressurisée. La détection du ^{36}Cl a été effectuée par spectroscopie de masse par accélérateur et par scintillateur liquide. Les concentrations en ^{36}Cl dans les mêmes résines d'un second réacteur à eau pressurisée et d'un réacteur à eau bouillante ont été encore mesurées. Dans tous les échantillons, la mesure de la concentration en ^{137}Cs a aussi été faite, permettant la détermination du facteur de corrélation entre activités du ^{36}Cl et du ^{137}Cs. Les facteurs de corrélation pour les résines sont respectivement $[1.4, 2.2$ et $17]x10^{-6}$, en relativement bon accord avec la seule valeur prédite de $6.0x1C^{-6}$ utilisée jusqu'ici par la Cédra pour estimer le ^{36}Cl dans les résines des différents réacteurs.
Les concentrations en ^{36}Cl mesurées et celles estimées auparavant ne diffèrent que d'un facteur 2.4 au plus.
Quant au facteur de corrélation mesuré pour l'eau de refroidissement du premier réacteur, il est de $5.8x10^{-5}$.

TABLE OF CONTENTS

4 tables

5 figures

1   INTRODUCTION

^{36}Cl is a long-lived cosmogenic nuclide (half-life $= 3.01 \times 10^5$ years). Natural ^{36}Cl is produced mainly by spallation of ^{40}Ar by cosmic rays in the atmosphere (LAL & PETERS 1967). In nuclear power plants ^{36}Cl is formed via neutron activation of ^{35}Cl in the cooling water system. Although the ^{35}Cl concentration in the water system can be as low as several ppb (1ppb $= 1\mu g$ Cl/kg H_2O), the amount of ^{36}Cl produced can be considerable due to the high nuclear reaction cross-section of ^{35}Cl (43 barns) and the high neutron flux. ^{36}Cl is a soft β-emitter ($E_{max} = 709$ keV) and, hence, a low-toxicity nuclide. However, it may play a prominent role in the inventory of a radioactive waste repository due to its long half-life and conservative geochemical behaviour (i.e. no retardation or retention due to interaction with the rock). The absolute significance of the radionuclide is dependent on its inventory which, currently, is based primarily on rough estimates. In particular, the few measurements of ^{36}Cl concentrations in primary reactor water and in ion-exchange resins made so far have not been published in the open literature. ^{36}Cl activities in the conditioned and packaged resins (and other waste streams) of the Swiss nuclear power plants have been estimated by NAGRA (NAGRA 1984) using an estimated scaling factor for the $^{36}Cl/^{137}Cs$ activity ratio of 6×10^{-6}. The purpose of this work was therefore to determine the ^{36}Cl concentrations in the resins (the most important operational waste stream) of several Swiss nuclear power plants as well as in the primary reactor water, along with the ^{137}Cs concentrations, in order to verify the estimates made previously.

Accelerator mass spectroscopy (AMS) is the most important tool for the detection of long-lived radionuclides such as ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca and ^{129}I at low concentrations (WÖLFLI 1987, ELMORE & PHILLIPS 1987). Since ELMORE et al. (1979) measured ^{36}Cl in environmental water samples with AMS in 1978, the applications of this nuclide have grown extensively and include groundwater dating (BENTLEY et al. 1986), use as a geothermal (PHILLIPS et al. 1984) or infiltration tracer (NORRIS et al. 1987), palaeoclimatic research through measuring ^{36}Cl in ice cores (ELMORE et al. 1987) or continental saline sediments (PHILLIPS et al. 1983), the study of meteorites (NISHIIZUMI et al. 1983), etc.. The sensitivity of ^{36}Cl measurement with AMS has been increased to 1×10^{-14} - 2×10^{-15} for the $^{36}Cl/Cl$ ratio (ELMORE & PHILLIPS 1987, SYNAL et al. 1987, KUBIK et al. 1984).

Liquid scintillation counting (LSC) can also be used for the detection of ^{36}Cl. Bagge and Kiel (BAGGE & KIEL 1966) determined the level of ^{36}Cl in a Turkish salt lake using a liquid scintillation technique based on ethylene dichloride as the sample. TAMERS et al. (RONZANI & TAMERS 1966, TAMERS et al. 1969, ROMAN & AIREY 1981) investigated ^{36}Cl in hydrological applications using LSC.

In this work, the ^{36}Cl concentration was measured in reactor cooling water and ion-exchange resins of the reactor coolant cleanup system (RCCS) of a Swiss PWR (denoted hereafter PWR-A) using LSC and AMS. The total chlorine content in the water and in the resins was determined by ion chromatography (IC). In addition, the ^{36}Cl concentrations in the active resins of a second PWR (PWR-B) and a BWR (BWR-A) were also determined using both the AMS and LC methods.

2 EXPERIMENTAL

A schematic overview of a PWR is shown in Fig. 1. The samples analyzed in this work were taken from the positions indicated by W (water) and R (resins).
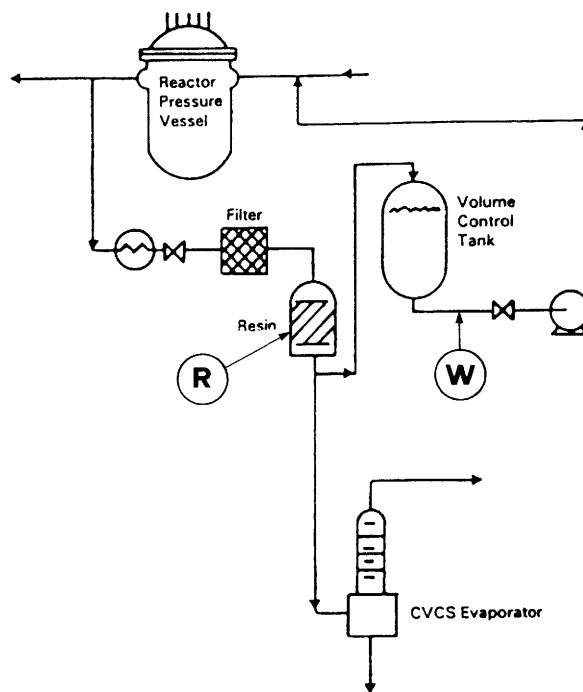
Fig. 1: Simplified overview of a PWR chemical and volume control system (DEGUELDRE & HUWYLER 1986).

2.1 Determination of ^{36}Cl in water samples

2.1.1 AMS measurement

Two types of reactor water from PWR-A were used. One was "fresh" water which was collected directly from the water cooling system. The samples were analyzed with a Dionex 2020i ion chromatography apparatus. A typical result is shown in Fig. 2.A, which demonstrates that, besides BO_3^{3-}, only small amounts of Cl^- were found. The concentration of Cl^- was determined as 10 ppb.

In addition, "old" water samples which had been stored in a container for about one year were also analyzed. An IC analysis of such a sample is shown in Fig. 2.B. Besides BO_3^{3-} and Cl^- (94 ppb), NO_3^- (26 ppb) and SO_4^{2-} (490 ppb) were also detected. We attribute the rather large amount of Cl^- and the significant peaks of NO_3^- and SO_4^{2-} mainly to contamination during storage.
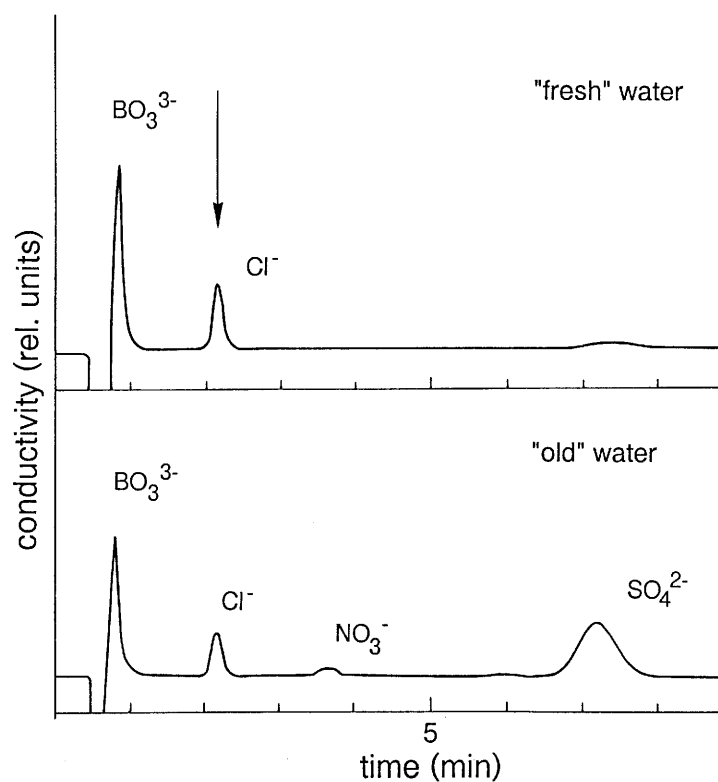
Fig. 2: Ion chromatography spectrum of a "fresh" and an "old" water sample.

The "fresh" water sample was also analyzed for gamma-active radionuclide concentrations. The results for some activities are summarized in Table 1.

Table 1: Activities of some radionuclides in reactor water and RCCS resins from different nuclear power plants.

Nuclide	Half-life	PWR-A "fresh"water (Bq/ml)	PWR-A resin (Bq/g)	PWR-B resin (Bq/g)	BWR-A resin (Bq/g)
^{54}Mn	312.2 D	1.44	8.2×10^4	-	-
^{60}Co	5.27 Y	7.03	1.4×10^7	6.1×10^5	9.0×10^3
^{134}Cs	2.06 Y	74.0	3.0×10^6	5.6×10^5	1.3×10^3
^{137}Cs	30.17 Y	77.7	5.8×10^6	2.2×10^6	6.4×10^4

The chemical procedure for the preparation of the samples for an AMS measurement was as follows (CONARD et al. 1986): 3.0 ml of Cl- carrier (^{36}Cl-free, 3.8 mg of Cl-/ml) was added to 1 ml (2 samples), 5 ml (2 samples) and 10 ml (1 sample) of reactor cooling water. The solution was acidified with 1 ml of concentrated nitric acid and diluted to 30 ml with deionized water. 2 ml of saturated $Ba(NO_3)_2$ was added and the solution was left for several hours. The solution was then filtered through a 0.45 μm filter-paper. 1 ml of 10% $AgNO_3$ was added to the filtrate in the centrifuge tube to precipitate AgCl. The centrifuge tube was wrapped with aluminium foil to protect it from light and left for several hours. After centrifuging, the supernatant was decanted and the AgCl precipitate was dissolved in 1 ml of NH_4OH plus 9 ml of deionized water. The solution was filtered through a 0.45 μm filter-paper and the filtrate acidified with concentrated HNO_3 to reprecipitate AgCl. After storage in the dark for several hours, the solution was centrifuged, decanted and the precipitate was suspended twice in deionized water. After centrifuging and decanting the supernatant, the AgCl precipitate (still in the centrifuge tube) was wrapped in aluminium foil and dried in an oven at 60-70 °C for about 20 hours.

The ^{36}Cl concentration measurements were carried out at the PSI/ETH AMS facility. Details of the procedure are given in Ref. (SYNAL et al. 1987).

2.1.2   LSC measurement

Although LSC is a good tool for measuring β-emitters, the concentration of ^{36}Cl in the reactor cooling water is too low to be detected directly. However, IC can be used to enrich the trace amounts of chloride by using special large-capacity columns. After preconcentration, ^{36}Cl from the eluted Cl- peak of the chromatogram can then be detected with LSC.

In order to eliminate disturbance of the ^{36}Cl measurement by the short-lived nuclide ^{131}I, "old" reactor cooling water was used. Preconcentrations and separations of Cl- were performed with the commercially available column TAC-1 and an AS 2 A column from DIONEX respectively. A concentration pump rate of 12 ml/min and an eluent pump rate of 3 ml/min were used. The eluent composition was 3 mmol of Na_2CO_3 / 2 mmol of NaOH. Firstly, 14 ml of the "old" water was rinsed through to condition the IC system, then about 50 ml of "old" water was injected into the preconcentration column and the Cl- peak was separated (about 3.6 ml). This enrichment process was repeated about 60 times with new "old" water. All the Cl- samples (about 220 ml) were combined and enriched a second

time, yielding a final Cl⁻ fraction of about 10 ml. From the Cl⁻ measurement in the original and the final sample, an enrichment factor of about 100 was achieved. The above-mentioned procedure was repeated once more, leading finally to two samples with 3.31 ppm and 3.69 ppm of Cl⁻. No other anions or gamma-active nuclides were found in the two samples. Each of the samples was mixed with the scintillator INSTA-GEL, which is well suited for applications with aqueous solutions (HANDLER & ROMBERGER 1973). The samples were measured with a TRI-CARB 1500 Liquid Scintillation Analyzer. The energy window chosen was optimized with a ³⁶Cl standard solution measured under the same quenching conditions as the real samples. This was important due to large quenching of the sample. A typical value for the quench factor tSIE was about 250 (tSIE is a parameter used to quantify quenching; no quenching: tSIE = 1000; total quenching: tSIE = 0). The measuring energy range was found to be optimum between 50 and 250 keV, even though ³⁶Cl is a β-emitter with E_{max} = 709 keV (Fig. 3).
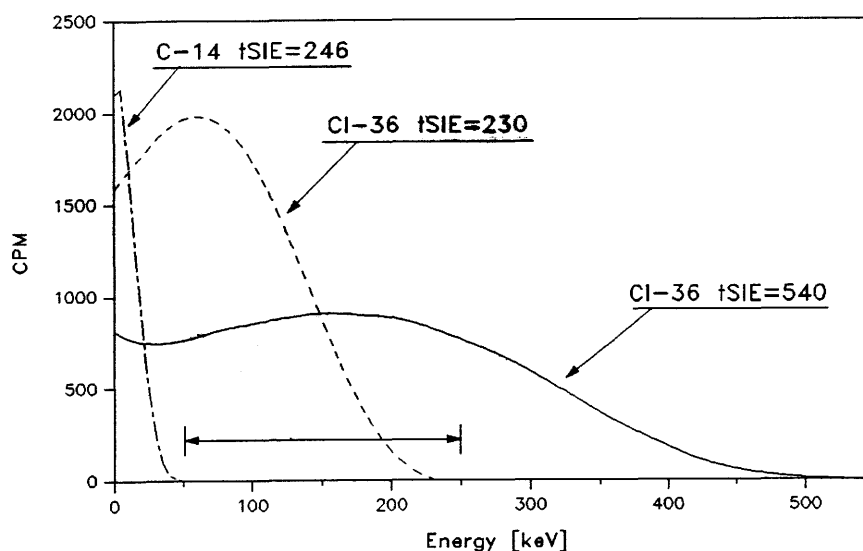

Fig. 3: Energy spectra of ³⁶Cl standard solution with large (tSIE = 230) and small (tSIE = 540) quenching and of ¹⁴C with large quenching (tSIE = 246). The counting window used in the work is indicated by the arrow (50 to 250 keV).

2.2   Determination of ^{36}Cl from active resins

2.2.1   Separation of chloride

The active RCCS ion-exchange resins which have been analyzed originated from three nuclear power plants. The resin from PWR-A was left for about one year and those from PWR-B and BWR-A for about 10 years before being sampled. The resin from BWR-A was in powder form. All resins (50% resin, 50% water) were cation-anion exchange mixtures. These samples were first analyzed for gamma-active nuclides. The results are summarized in Table 1. For the resins from PWR-A, two measurements, one at PSI and the other at the nuclear power plant, agreed well.

In the following the desorption of Cl⁻ from the resins is described. Bearing in mind the long storage times, we considered it difficult to elute the ions using conventional elution methods. On the other hand, the ^{36}Cl concentrations in the resins, if compared to the nuclides listed in Table 1, were very low. This means that minor contaminations in the eluent by trace amounts of cations prevent ^{36}Cl from being measured directly by LSC.
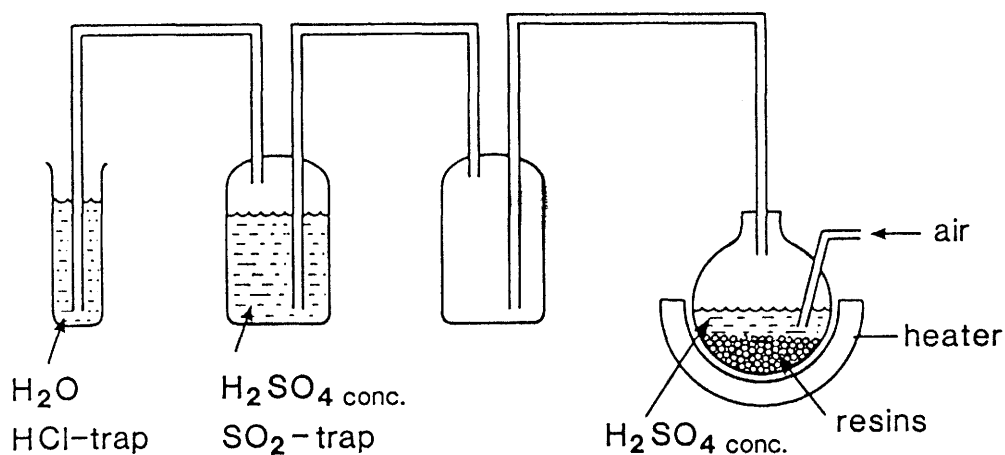

Fig. 4: Schematic illustration of the distillation system.

We therefore used a distillation method to separate Cl- in the form of HCl. The schematic diagram of the distillation system is shown in Fig. 4. The separation was performed in the following way: 2.0 g of active resins were transferred to a hermetically-sealed flask and 15 ml of concentrated sulphuric acid was added to the flask to carbonate the resins and to release the chloride as HCl. Air with an appropriate flow-rate was bubbled through the H_2SO_4 solution to carry the HCl to an absorption trap containing 20 ml of deionized water. Prior to this HCl trap, the gas was bubbled through an H_2SO_4 trap to absorb trace amounts of SO_2 and reduce the amount of sulphuric species in the final chloride samples. Without this trap, the concentration of SO_4^{2-} in the Cl- fraction was too high, preventing determination of the total Cl- concentration by IC. The distillation temperature was regulated to about 200 °C. Even with this SO_2 trap, a quantitative determination of Cl- failed for some samples due to high contamination with SO_4^{2-}.

2.2.2   Determination of ^{36}Cl from the chloride fraction

LSC was used to detect ^{36}Cl from the HCl trap (absorbent solution) and to monitor the distillation process in order to determine the end of the chlorine release. 10 ml of the absorbent solution was measured for 100 min as described above. In pre-experiments it was found that the sulphate concentration and the pH had no effect on ^{36}Cl detection.

AMS was also used for measuring ^{36}Cl in the absorbent samples, in order to provide an independent determination. Since the absorbent samples were contaminated by sulphur, it was necessary to remove ^{36}S as an isobaric interference. Saturated $Ba(NO_3)_2$ solution was therefore added to precipitate $BaSO_4$. Because the ^{36}Cl concentration in the absorbent solution was too high, it had first to be diluted with deionized water to meet the appropriate $^{36}Cl/Cl$ range of 10^{-10} to 10^{-13} for an AMS measurement. The diluted absorbent solution was prepared for AMS measurement following the procedure described above.

The total chloride concentration in the absorbent solution was determined with IC.

3   RESULTS AND DISCUSSION

3.1   Reactor water

All results from the ^{36}Cl and total chlorine determinations from the reactor cooling water of PWR-A are shown in Table 2. The symbols F denote "fresh" and O "old" water. F-1 and F-2 are duplicate determinations from 1 ml samples, F-3 and F-4 from 5 ml, O-1 and O-2 from 0.1 ml, O-3 and O-4 from 1 ml and F-5 from a 10 ml sample, respectively. O-5 and O-6 are about 100-fold enriched 10 ml samples (see Experimental). As a result, $[6.1 \pm 0.4]\text{x}10^{10}$ atoms of ^{36}Cl per milliliter of "fresh" water (= $4.5\text{x}10^{-3}$ Bq/ml) and $[1.62 \pm 0.05]\text{x}10^{10}$ atoms of ^{36}Cl per milliliter of "old" water (= $1.2\text{x}10^{-3}$ Bq/ml) were detected in the AMS measurements. The corresponding result from the LSC determination is $[1.6 \pm 0.2]\text{x}10^{10}$ atoms of ^{36}Cl per milliliter of "old" water, in good agreement with the AMS result. The data show that the ^{36}Cl concentration in "old" water is roughly a factor of four lower compared to the "fresh" water sample. It remains open whether this reflects a different irradiation history for both sets of water samples or whether some of the ^{36}Cl in the "old" sample was lost due to isotope exchange reactions with chloride atoms adsorbed onto the walls of the storage container.

Also shown in Table 2 are the total chlorine concentrations measured with IC. They are also listed in the form of atoms per milliliter. One determination was performed for each of the "fresh" and "old" water samples for the AMS measurements and for the "old" sample for the LSC measurements. Agreement between the three different determinations is rather poor, ranging from $1.7\text{x}10^{14}$ Cl- atoms per milliliter of "fresh" water (= 10 ppb) to $5.8\text{x}10^{14}$ and $1.6\text{x}10^{15}$ Cl- atoms per milliliter of "old" water (= 34 and 94 ppb) respectively. This may reflect contamination problems encountered when storing and processing water samples over long time periods. This is also illustrated in Fig. 2 where "old" water samples show contamination with NO_3^- and SO_4^{-2} not appearing in the "fresh" water samples. We therefore consider only the chloride concentrations of 10 ppb measured in the "fresh" water samples to be of relevance. The relative ^{36}Cl/Cl concentration in "fresh" water samples then results as $[3.6 \pm 0.2]\text{x}10^{-4}$. The corresponding numbers for the "old" samples given in Table 2 are of a qualitative nature only, due to the arguments presented above.

Table 2: Results from the Cl- and ^{36}Cl measurements of PWR-A reactor water using IC, AMS and LSC. The symbols are explained in the text (F: "fresh" water, O: "old" water).

Sample	by IC	by AMS	
(No.)	N(Cl-)/ml	N(^{36}Cl)/ml	^{36}Cl/Cl
F-1		6.23×10^{10}	3.66×10^{-4}
F-2		6.38×10^{10}	3.76×10^{-4}
F-3	1.70×10^{14}	6.07×10^{10}	3.58×10^{-4}
F-4		6.46×10^{10}	3.81×10^{-4}
F-5		5.74×10^{10}	3.39×10^{-4}
O-1		1.60×10^{10}	1.01×10^{-5}
O-2	1.59×10^{15}	1.66×10^{10}	1.04×10^{-5}
O-3		1.57×10^{10}	0.99×10^{-5}
O-4		1.64×10^{10}	1.03×10^{-5}

Sample	by IC	by LCS	
(No.)	N(Cl-/ml)	N(^{36}Cl)/ml	^{36}Cl/Cl
O-5	5.83×10^{14}	1.36×10^{10}	2.33×10^{-5}
O-6		1.86×10^{10}	3.19×10^{-5}

It is interesting to compare the ^{36}Cl activity from reactor water with that of the key nuclide ^{137}Cs. Taking the average ^{36}Cl concentration in "fresh" water from Table 2 (6.1×10^{10} atoms/ml or 4.5×10^{-3} Bq/ml) and the ^{137}Cs activity from Table 1, we determine an experimental scaling factor for ^{36}Cl relative to ^{137}Cs of [4.5×10^{-3} Bq/ml]/[77.7 Bq/ml] = 5.8×10^{-5} for the "fresh" reactor water of PWR-A.

3.2   Resins

Firstly the kinetics of chlorine release from the resins was investigated. In Fig. 5 the results for the 4 PWR-A samples are shown. After each differential distillation step, 50 % of the Cl- absorbent (i.e. 10 ml) was forwarded to LSC counting, yielding the data shown in Fig. 5. For the next distillation step, fresh H_2O was put into the HCl trap and the process repeated. The data shown in Fig. 5 then depict the cumulative count-rate of ^{36}Cl for increasing distillation steps. All residual 50% samples of the Cl- absorbent solutions for each distillation step were finally combined for an AMS measurement of ^{36}Cl and an IC determination of the total chlorine content. Some of these cumulative absorbent samples were also counted for gamma-active nuclides. No indications of nuclides such as ^{60}Co, ^{137}Cs or ^{54}Mn were found.
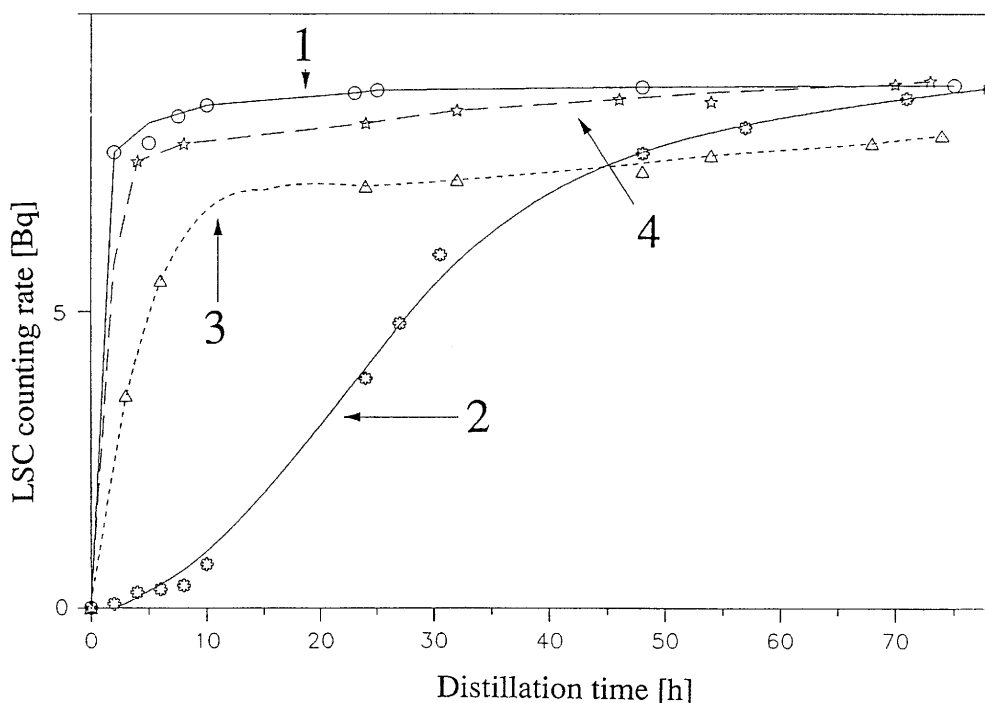

Fig. 5: Release of ^{36}Cl (measured with LSC) from 4 PWR-A resin samples as a function of the distillation time.

As Fig. 5 shows, for three samples the total release of chlorine occurred within about 10 hours. In one case (sample 2) total release was achieved only after some 70 hours of distillation. This may be caused by incorrect heating i.e. contact problems between heater and flask (see Fig. 4).

Table 3 summarizes all results from the resin experiments. The ^{36}Cl contents for the identical samples PWR-A 1 to 4, but also for the independent determinations with LSC and AMS, agree exceptionally well. The average result for PWR-A is $[1.1 \pm 0.2] \times 10^{14}$ atoms of ^{36}Cl per gram of resin containing 50% of water (= 8 Bq/g). For the PWR-B and BWR-A, the results are 6.7×10^{13} ^{36}Cl/g (= 4.9 Bq/g) and 1.5×10^{13} ^{36}Cl/g (= 1.1 Bq/g) respectively.

If we again compare the ^{36}Cl activities in the resins with the key nuclide ^{137}Cs (see Table 1), then the following experimental scaling factors are obtained: 1.4×10^{-6}, 2.2×10^{-6} and 17×10^{-6} respectively for PWR-A, PWR-B and BWR-A. These values should be compared to the theoretical estimate of 6×10^{-6} given for the PWR and BWR resins (DEGUELDRE & HUWYLER 1986, NAGRA 1984). We therefore conclude that the experimental results are in reasonable agreement with the predictions.

Table 3: Determination of Cl$^-$ and ^{36}Cl from active resins of the nuclear power plants PWR-A, PWR-B and BWR-A.

Sample	by IC	by LSC			by AMS		
(No.)	N(Cl$^-$)/g	Bq/g	N(^{36}Cl)/g	^{36}Cl/Cl	Bq/g	N(^{36}Cl)/g	^{36}Cl/Cl
PWR-A:1	1.52×10^{18}	8.8	1.21×10^{14}	0.80×10^{-4}	6.9	0.94×10^{14}	0.62×10^{-4}
PWR-A:2	-	8.8	1.20×10^{14}	-	8.2	1.12×10^{14}	-
PWR-A:3	1.04×10^{18}	8.0	1.09×10^{14}	1.05×10^{-4}	9.1	1.24×10^{14}	1.19×10^{-4}
PWR-A:4	0.68×10^{18}	8.9	1.22×10^{14}	1.79×10^{-4}	8.5	1.16×10^{14}	1.71×10^{-4}
PWR-B	-	4.9	6.66×10^{13}	-	5.3	7.16×10^{13}	-
BWR-A	-	1.1	1.52×10^{13}	-	1.1	1.55×10^{13}	-

3.3   Comparison with previous estimates

The results for the resins are compared in Table 4 with values derived from the ^{36}Cl activities and the resin masses (50% resin, 50% water) per package as assumed in the Nagra "1984 Swiss Radioactive Waste Inventory" (NAGRA 1984). In this reference, the ^{36}Cl activities were estimated on the basis of a scaling factor of 6×10^{-6} to measured ^{137}Cs activities.

Table 4: Comparison of measured (this work) and scaled ^{36}Cl specific activities and of measured (this work) and estimated scaling factors for resins of the reactor coolant cleanup system.

^{36}Cl specific activities [Bq/g]		
Reactor	This work	(NAGRA 1984)
PWR-A	8.0	6.2
PWR-B	4.9	6.9
BWR-A	1.1	2.6

^{36}Cl/^{137}Cs scaling factors		
Reactor	This work	(DEGUELDRE and HUWYLWER 1986) (NAGRA 1984)
PWR-A	1.4×10^{-6}	6.0×10^{-6}
PWR-B	2.2×10^{-6}	6.0×10^{-6}
BWR-A	17×10^{-6}	6.0×10^{-6}

Despite the inherent uncertainties in estimating scaling factors, which is particularly the case for ^{36}Cl (^{36}Cl is produced via ^{35}Cl from Cl^- impurities in the water and ^{137}Cs comes from fuel leakage), the agreements between the present measured activities and the activities previously estimated from ^{137}Cs measurements and a single scaling factor are surprisingly good. Hence, the results of this work confirm experimentally the estimates for the ^{36}Cl concentrations of the RCCS resins for the disposal of this waste (NAGRA 1984).

The measured scaling factor $^{36}Cl/^{137}Cs$ for the water sample from the reactor PWR-A was 5.8×10^{-5}. This value implies a scaling factor modifier of 1/40 between water and resin samples in order to explain the measured scaling factor of 1.4×10^{-6} for the resin. A theoretical scaling factor modifier of 1 has been proposed for the primary clean-up resins (LIEBERMANN et al. 1983). The difference found in our work illustrates the need for further experimental investigations if the ^{36}Cl activity in the different waste streams is to be estimated from ^{137}Cs through coolant scaling factors and scaling factor modifiers.

4   REFERENCES

BAGGE, E. & KIEL, H.W. 1966: Atomkernenergie 11 176.

BENTLEY, H.W., PHILLIPS, F. M., DAVIS, S. N., HABERMEHL, M. A.,
      AIREY, P. L., CALF, G. E., ELMORE, D., GOVE, H.E. & TORGERSEN,
      T. 1991: Water Resources Research 22 (1986).

CONARD, N.J., ELMORE, D., KUBIK, P.W., GOVE, H.E., TUBBS, L.E.,
      CHRUNGK, B.A. & WAHLEN, M. 1986: Radiocarbon 28 556.

DEGUELDRE, C. & HUWYLER, S. 1986: NAGRA Technical Report, 86-28.

ELMORE, D., CONARD, N.J., KUBIK, P.W., GOVE, H.E., WAHLEN, M., BEER,
      J. & SUTER, M. 1987: Nucl. Instr. & Meth. B29 207.

ELMORE, D., FULTON, B.R., CLOVER, M.R., MARSDEN, J.R. & LITHERLAND,
      A.E. 1979: Nature 277 22.

ELMORE, D. & PHILLIPS, F. 1987: Science 236 543.

HANDLER, L.H. & ROMBERGER, J.A. 1973: Int. J. Appl. Radiat. and
      Isotopes 24 129.

KUBIK, P.W., KORSCHINEK, G., NOLTE, E., RATZINGER, U., ERNST, H.,
      TEICHMANN, S., MORINAGA, H., WILD, E. & HILLE, P. 1984: Nucl.
      Instr. & Meth. B5 326.

LAL, D. & PETERS, B. 1967: Handbuch der Physik, 46/2, ed. by
      S. Flugge, Springer-Verlag, Berlin, pp. 551-612.

LIEBERMANN, J.A., MCILVAINE, J.B., MILLER, A.D. & RODGER, W.A. Dec.
      1983: Methodology for the classification of low-level
      radioactive wastes from nuclear power plants. AIF/NESP-027.

NAGRA 1984: NAGRA Technical Report, 84-47.

NISHIIZUMI, K., ARNOLD, J.R., ELMORE, D., MA, X., NEWMAN, D. & GOVE,
      H.E. 1983: Earth Planet. Sci. Lett. 62 407.

NORRIS, A.E., WOLFSBERG, K., GIFFORD, S.K., BENTLEY, H.W. & ELMORE, D.
      1987: Nucl. Instr. & Meth. B29 376.

PHILLIPS, F.M., GOFF, F., VUATAZ, F., BENTLEY, H.W., ELMORE, D. &
      GOVE, H.E. 1984: Geophysical Research Letters, 11 1227.

PHILLIPS, F.M., SMITH, G.I., BENTLEY, H.W., ELMORE, D. & GOVE, H.E.
      1983: Science 222 925.

ROMAN, D. & AIREY, P.L. 1981: Int. J. Apl. Radiat. & Isotopes 32 287.

RONZANI, C. & TAMERS, M.A. 1966: Radiochim. Acta 6 206.

SYNAL, H., BEER, J., BONANI, G., HOFMANN, H.J., SUTER, M. & WÖLFLI, W. 1987: Nucl. Instr. & Meth. B29 146.

TAMERS, M.A., RONZANI, G. & SCHARPENSEEL, H.W. 1969: Atompraxis 15 433.

WÖLFLI, W. 1987: Nucl. Instr. & Meth. B29 1.