

RETENTION OF ^{14}C IN NUCLEAR POWER PLANTS AND REPROCESSING PLANTS

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INTRODUCTION

Up to about 1972 it was unknown that the emissions of ^{14}C from nuclear power plants and reprocessing plants represent significant contributions to radiation exposure. Calculations, however, have shown that ^{14}C may give the highest contributions to the individual dose (1), (2). Following measurements confirmed the emission rate (3), (4). The collective dose commitment, however, is of greatest importance for ^{14}C . Because of the long half life of 5730 a it is very high, too (5).

PRODUCTION RATES OF ^{14}C

In reactors, ^{14}C is mainly produced due to neutron reactions from $^{13}\text{C}(n, \gamma)^{14}\text{C}$, $^{14}\text{N}(n, p)$ and $^{17}\text{O}(n, \gamma)$. The ^{14}C produced in the fuel is not emitted until dissolution, while ^{14}C produced in the coolant enters the air-circulating plant via leakages and is emitted from there or is emitted via the off-gas-system. Tab. 1 shows the production rates in fuel and coolant of all common reactor types. Details of the calculation are represented in (6).

^{14}C RETENTION FACILITY

^{14}C is emitted as gaseous compound, mainly as $^{14}\text{CO}_2$, but also as ^{14}CO and $^{14}\text{C}_n\text{H}_m$ together with a considerably higher part of ^{12}C compounds. If significant amounts of other radio carbon compounds exist beside $^{14}\text{CO}_2$, it is sensible, to oxidize these to $^{14}\text{CO}_2$ before the retention. Then the "direct" or the "double" alkali process can be used to retain ^{14}C .

Both processes have two considerable advantages in comparison with other processes:

1. The absorption of the CO_2 is supported by a chemical reaction, whereby high decontamination factors are reached.
2. ^{14}C becomes available as calciumcarbonate, a solid stable compound that can be concreted.

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Up to now, mainly the direct alkali process has been examined, because it seems technically more simple to realize and more economical (7),(8), although the chemical reactions of the double alkali process and its control action concerning changes in the mass flows are more favorable. This is important, because the ^{14}C release takes place instantaneously from nearly all nuclear facilities; both the volume flow and the concentration of the carbon compounds often vary.

Therefore, a ^{14}C retention facility with the double alkali process has been developed at the professorial chair for reactor technology at Aachen Technical University. Concerning the process scheme, the retention facility has been constructed as simple as the direct alkali process, but it has all the advantages of the double alkali process. Fig. 1 shows the flow sheet.

From a reservoir, sodium hydroxide is pumped through a packed column where inert gas containing CO_2 flows in inverse current. When the alkaline solution is converted to 60 %, the total alkaline solution inventory is pumped into the agitator for regeneration with $\text{Ca}(\text{OH})_2$. At the bottom of the agitator CaCO_3 is bottled as a viscous slurry with a high percentage of liquid. The high percentage of liquid, approx. 50 % wt, is used for concreting. Fig. 2 shows the facility, the agitator has not yet been finished. The facility is used to obtain operation experiences with the new process and to optimize the process parameters. The most important construction data are:

max. gas mass flow : 40 kg/h

max. liquid flow : 500 kg/h

operation pressure : 1.5 ... 3 bar.

The actual operation experiences are well. A ^{14}C retention facility with this process scheme could be installed in LWR's and reprocessing plants for LWR fuel elements.

COST-BENEFIT ANALYSIS OF THE ^{14}C RETENTION FACILITY

If the cost-benefit analysis recommended in ICRP publication 26 is used, see Fig. 3, it is obvious that a ^{14}C retention facility with oxidation-process should be installed in reprocessing plants for LWR fuel elements (5). An installation in LWRs doesn't seem sensible to us because of the low ^{14}C emission rates. For reprocessing plants for HTR fuel elements with burn-leach-head-end the proposed method cannot be used because the waste volumes become too large. In this case the head-end should be changed in order to solve the ^{14}C problems. In big reprocessing plants for nitridic fuel elements of the fast breeder such a high amount of ^{14}C is set free (10^6 Ci/a) that in spite of high decontamination factors so much ^{14}C would be released that this fuel cycle should not be continued any further.

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Tab.1: Production of ^{14}C in different reactor types in Ci/GW(e)a

Specification		¹⁴ C-Production in Ci/GW(e)a							
		BWR	PWR	HWR	MAGNOX	AGR	HTR	FBR	
Outer surface of pressure vessel	¹⁴ N	5 · 10 ⁻⁴	0,005	0,005	5 · 10 ⁻⁴	0,001	0,005	1	
Coolant	¹³ C	7 · 10 ⁻⁷	8 · 10 ⁻⁷	3 · 10 ⁻⁵	0,06	0,06	4 · 10 ⁻⁷	5 · 10 ⁻⁷	
	¹⁴ N	0,6	0,8	25	7,3	7,1	0,02	0,02	
	¹⁷ O	5,1	6,2	175	1,1	1	5 · 10 ⁶	2 · 10 ⁶	
Fuel element	Fuel	Fission	0,6	0,6	0,6	0,6	0,5	0,5	0,5
		¹³ C	2 · 10 ⁻⁴	2 · 10 ⁻⁴	7 · 10 ⁻⁴	8 · 10 ⁻⁴	2 · 10 ⁻⁴	9 · 10 ⁻⁵	1 · 10 ⁻⁵
		¹⁴ N	7,6	7,8	26	130	13	3,1	2
		¹⁷ O	4,4	4,5	13	0,01	3,3	1,6	3
	Canning	¹³ C	3 · 10 ⁻⁴	5 · 10 ⁻⁴	7 · 10 ⁻⁴	4 · 10 ⁻⁴	6 · 10 ⁻⁴	—	1 · 10 ⁻⁵
		¹⁴ N	17	20	34	35	32	—	8
		¹⁷ O	0,015	0,02	0,03	0,003	0,003	—	2 · 10 ⁻⁴
	Graphite moderator	¹³ C	—	—	—	110	35	32	—
		¹⁴ N	—	—	—	180	59	54	—
		¹⁷ O	—	—	—	0,02	7 · 10 ⁻⁴	7 · 10 ⁻⁴	—
Total production rate		34	40	274	500	85	91	15	

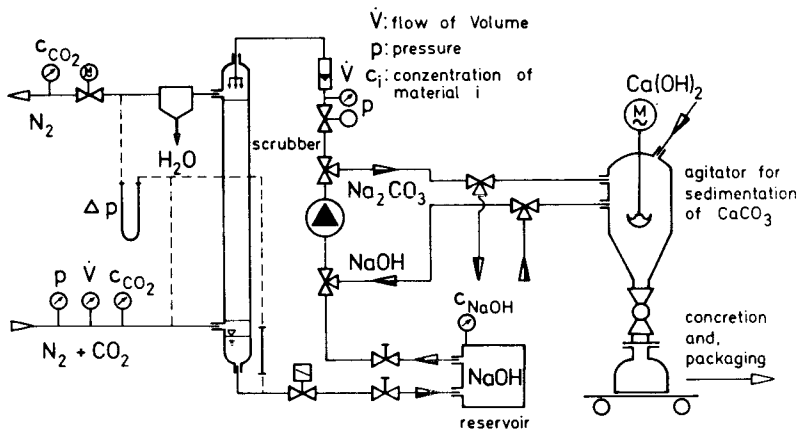


Fig.1: Flow sheet of the double alkali process



Fig.2: Experimental facility for the retention of ^{14}C

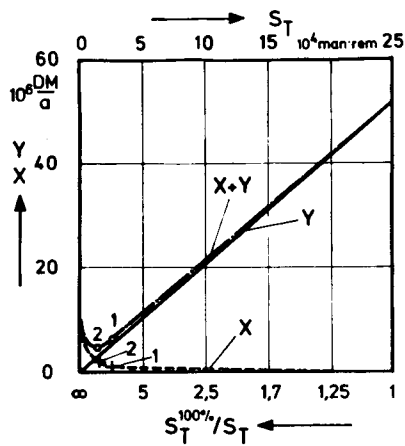


Fig.3: Cost-benefit-analysis of ^{14}C -retention in processing plants with a capacity of 1500 t/a

1: $^{14}\text{CO}_2$ only
 2: $^{14}\text{CO}_2 + ^{14}\text{CO} + ^{14}\text{C}_n\text{H}_m$