PRODUCTION AND EMISSION OF CARBON-14 FROM NUCLEAR POWER STATIONS AND REPROCESSING PLANTS AND ITS RADIOECOLOGICAL SIGNIFICANCE

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1. INTRODUCTION

Up to 4 years ago C 14 was not recognized as a radionuclide with consequences within the nuclear fuel cycle (1), (2), (3), (4). New investigations have shown that the radiation exposure from this isotope is not negligible when compared to the other emissions from nuclear facilities.

2. PRODUCTION OF C 14 IN NUCLEAR POWER REACTORS AND EMISSION FROM NUCLEAR FACILITIES

In nuclear power reactors C 14 is mainly produced due to (n,γ)-reactions with C 13, (n,p)-reactions with N 14 and (n,α)-reactions with O 17. Other possible reactions with their threshold energies are illustrated in Fig. 1 (1). The most important nuclides out of which C 14 is produced are present in the reactor-materials and in the atmosphere around the reactor pressure vessel.

The C 14 production, due to the C-, N- and O-impurities listed in Tab. 1, the oxygen content of the coolant and of the air around the reactor pressure vessel in Light Water Reactors (LWR) and the carbon of the graphite moderator in High Temperature Reactors (HTR), is estimated in the affected parts of the facilities. The results for plants with 1000 MW_e full load capacity are shown in Tab. 2. In contrast to reference (2) these new estimations for Liquid Metal Fast Breeder Reactors (LMFBR) are based on the assumption that the sodium layer between reactor core and reactor tank is greater in this reactor type. Furthermore a C 14-production by ternary fission seems to be possible (5), although an estimation of this content is nowadays very difficult because of the uncertain fission yields. A rough estimation indicates the values listed in Tab. 2. As can be seen from this table the main production of C 14 is caused by the N-impurities in the fuel and the graphite, and by the oxygen in the water and the fuel-elements.

The emissions from nuclear facilities are estimated assuming that the C 14 in the coolant of the reactor, and the C 14 entering the reprocessing plant (RP) during the leaching of the fuel elements is totally emitted from the respective facility especially as the chemical form of CO_2 via the stack of a HTR-reprocessing plant with assumed grind-burn-leach head-end. The emission data of LWR and RP for LWR-fuel elements are verified in the meantime by several measurements (6), (7), (8), (9). It is interesting that C 14 of BWR (6) and RP (9) is found as CO_2 in the waste air, whereas hydrocarbons such as CH_4 and C_2H_6 are mainly found in PWR (7), (8). The waste water emission from LWR seems to be about 1% of the total C 14 release (10), (11).

3. RADIATION EXPOSURE DUE TO C 14 FROM NUCLEAR FACILITIES

The radiological significance of C 14 can be seen from Tabs. 3 and 4, where the radiation exposure of this nuclide is compared to that of other isotopes. These values are estimated with the emission data of Tabs. 5 and 6 and are valid for the maximum concentration in air, Fig. 2. To assess the exposure in a conservative manner, the contaminated foodstuffs are stipulated to be produced at the specific site, C 14 being in the chemical form of CO_2. By means of a specific activity model the dose conversion factor for the ingestion of this isotope is assessed at 42 rem m^3/Ci s for the total body, the dose of the natural content of C 14 in the total body being 1.41 mrem/a with a corresponding CO_2 air concentration of 325 ppm. The results indicate that C 14 is the most important radionuclide for the total body radiation exposure in the vicinity of LWR. It is also significant for RP, especially those for HTR-fuel-elements with grind-burn-leach head-end. Therefore it seems to be doubtful if it is possible to allow a release of the total C 14 O_2 from a large reprocessing plant for HTR-fuel elements in the future. Many problems will also arise in connection with the reprocessing of nitride fuel elements (12) for LMFBR, in which the production rate will be about 10^4 Ci/a for 1000 MW_e installed power capacity.

The collective dose is important as a further criterion for the radiological significance of C 14, the dose for the first pass exposure being about 1000 man-rem with a stipulated emission of 500 Ci and present population density. The contribution of the same amount of globally distributed C 14 to the collective dose depends on the integration time - see Fig. 3 - and will be about $2 \cdot 10^5$ man-rem for a stipulated population of $1 \cdot 10^{10}$ human beings. Regarding the total body the collective dose by C 14-emissions as shown in Tab. 6 will be of more significance than those of all other nuclides (13).

The meaning of globally distributed C 14 for the collective dose is also illustrated by the future radiation exposure, see Fig. 4, which is estimated with a prognosis of the installed nuclear power capacity according to reference (14). About two thirds of this future dose is caused by emissions from HTR-RP, a total release of C 14 being stipulated. If it is possible to retain about 80% of this isotope in RP and to finally store this amount, the dose due to globally distributed C 14 will be in acceptable limits.

4. SUMMARY

In nuclear power reactors with 1000 MW_e full load capacity C 14 will be produced at rates of 10 to 100 Ci/a according to the reactor type. The yearly emission rate for LWR is assessed at 10 Ci, for HTR at 0.1 Ci and for LMFBR at 1 Ci. The corresponding RP for 40.000 MW_e installed nuclear reactor capacity may release about 500, 3000 and 100 Ci/a according to the nitrogen impurities in the fuel elements and the coolant of the reactor type. This C 14 will significantly influence the radiation exposure of the population.

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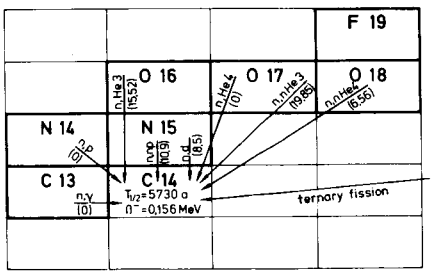

Fig 1: Neutron reactions leading to C 14 with the threshhold energies in MeV

Reactor type	Impurities [ppm] or [vpm]								
	Coolant			Fuel elements					
				Fuel			Canning		
	C	N	O	C	N	O	C	N	O
BWR	1	5	-	50	6	-	270	80	1500
PWR	1	5	-	50	6	-	270	80	1500
HTR (Spherical)	CO 3 vpm, CO_2 1 -, CH_4 01 -, 1 vpm		CO 3 vpm, H_2O 12 -, CO_2 1 -	50	6	-	1)	5 1)	2)
LMFBR	20	1	1	50	6	-	1000	1000	30

Tab 1 C, N and O-impurities used to calculate the rate of C-14 production in the different nuclear reactor types
1) Graphite
2) In the volume of voids similar impurities as in the coolant

Specification			Reactor type			
			BWR	PWR	HTR (gaher coil)	LMFBR
Outer surface of pressure vessel		N 14	5 E-4	5 E-3	4 E-3	1
Coolant		C 13	4.6E-7	4.5E-7	5.5E-7	1.2E-6
		N 14	1.3	1.3	4.8E-2	2.6E-3
		O 17	9.9	9.8	7.3 E-6	4.3E-7
Fuel element	Fuel	Fission	0.6	0.6	0.5	0.5
		C 13	4.9 E-4	4.1 E-4		3.7E-4
		N 14	9.9	8.4	2.7	1.6
		O 17	8.4	7.1	2.3	0.5
	Canning	C 13	6 E-5	8 E-5	40	
		N 14	3	3.8	3.1	14.5
		O 17	2 E-3	3 E-3		
Release from reactor			11.2	11.1	0.1	1
Release from reprocessing plant			18.9	16.1	76.5	2.6
Stored with canning			3	3.8	–	14.5

Tab. 2: Yearly production of C14 in different reactpr types (1000 $MW_{(e)}$ full load) and emission rates of nuclear power plants and reprocessing plants.

Organ	Radiation exposure [mrem/a]						
	Nuclear power plant				Reprocessing plant		
	BWR	PWR	HTR	LMFBR	LWR	HTR	LMFBR
Total body	0.14	0.15	0.02	0.04	3.2	13.6	1.3
Skin	0.17	0.18	0.025	0.12	7.8	21.1	3.7
Thyroid (Infant)	6.8	3.5	0.04	0.1	50.2	63.8	41.3
Lung	0.14	0.15	0.02	0.04	4.1	14.1	4.1
Bone	0.16	0.17	0.02	0.04	5.2	15.6	4.6

Tab. 4: Maximum possible radiation exposure due to gaseous releases from nuclear power plants [1000 MW(e)] and reprocessing plants [40.000 MW(e) full load] in the maximum of the concentration

Emission height
Nuclear power plant 100 m , $\chi = 3E-7$ s/m³
Reprocessing plant 200 m , $\chi = 1E-7$ s/m³

Nuclide	Release rates [Ci/a]			
	BWR	PWR	HTR	LMFBR
H 3	30	20	10	
C 14	10	10	0.1	1
Kr 85	700	700	120	1000
Kr 88	30	50	80	50
Sr 89	1E-3	1E-3		
Sr 90	1E-4	1E-4		
I 131	0.1	0.05	3E-4	9E-3
Xe 133	2500	2500	40	7000
Cs 134	1E-4	1E-4		
Cs 137	2E-4	2E-4		
α-rays	1E-5	1E-5		1E-5

Tab. 5: Expected radioactive effluents from a nuclear power plant (1000 MW(e)) to the atmosphere

Nuclide	Organ	Exposure pathway	Radiation exposure [mrem/a]					
			Nuclear power plant				Reprocessing plant	
			BWR	PWR	HTR	LMFBR	LWR	HTR	LMFBR
H3	T-Body	Inh.	5 E-4	3 E-4	2 E-4		0,42	0,36	0,36
	T-Body	Ing.	8 E-4	5 E-4	3 E-4		0,63	0,54	0,54
	Skin	β-Subm.	1 E-5	9 E-6	5 E-6		0,02	0,01	0,01
C14	T-Body	Ing.	0,13	0,13	1 E-3	0,01	2,1	12,6	0,42
Kr85	T-Body	γ-Subm.	7 E-5	7 E-5	1 E-5	1 E-4	0,04	0,07	0,02
	Skin	β-Subm.	8 E-3	8 E-3	1 E-3	0,01	4,6	7,6	2,3
Kr88 +Rb88	T-Body	γ-Subm.	4 E-3	7 E-3	0,01	7 E-3			
	Skin	β-Subm.	7 E-4	1 E-3	2 E-3	1 E-3			
Sr89	Bone	Inh.	3 E-6	3 E-6			9 E-5	9 E-5	9 E-5
		Ing.	7 E-4	7 E-4			0,02	0,02	0,02
Sr90	Bone	Inh.	9 E-5	9 E-5			0,03	0,03	0,03
		Ing.	5 E-3	5 E-3			1,5	1,5	1,5
Xe133	Skin	β-Subm.	0,02	0,02	3 E-4	0,06			
	T-Body	γ-Subm.	9 E-3	9 E-3	1 E-4	0,03			
I 129	Thyroid (Infant)	Inh.					0,02	0,02	0,01
		Ing.					40,0	44,0	32,0
I 131	Thyroid (Infant)	Inh.	0,03	0,01	8 E-5	3 E-3	0,02	0,01	0,02
		Ing.	6,6	3,3	0,02	0,6	7,0	6,2	7,9
Cs134	T-Body	Inh.	6 E-7	6 E-7			2 E-4	2 E-4	2 E-4
		Ing.	1 E-4	1 E-4			0,03	0,03	0,03
Cs137	T-Body	Inh.	7 E-7	7 E-7			1 E-4	1 E-4	1 E-4
		Ing.	1 E-4	1 E-4			0,02	0,02	0,02
Pu238	Lung	Inh.	4 E-5	4 E-5		6 E-5	0,23	0,50	0,85
	Bone		4 E-5	4 E-5		5 E-5	0,2	0,42	0,8
Pu239	Lung	Inh.	5 E-6	5 E-6		2 E-5	0,03	4 E-4	0,3
	Bone		5 E-6	5 E-6		2 E-5	0,03	4 E-4	0,34
Pu240	Lung	Inh.	7 E-6	7 E-6		2 E-5	0,04	9 E-4	0,35
	Bone		8 E-6	8 E-6		3 E-5	0,04	1 E-3	0,4
Am241	Lung	Inh.	2 E-6	2 E-6		8 E-6	0,02	7 E-4	0,17
	Bone		1 E-6	1 E-6		5 E-6	0,01	4 E-4	0,10
Cm242	Lung	Inh.	1 E-4	1 E-4		1 E-4	0,35	8 E-3	0,9
	Bone		5 E-6	5 E-6		5 E-6	0,01	3 E-4	0,04
Cm244	Lung	Inh.	4 E-5	4 E-5		7 E-6	0,22	0,05	0,11
	Bone		3 E-5	3 E-5		6 E-6	0,18	0,04	0,09

Tab. 3: Maximum possible radiation exposure due to gaseous releases from nuclear power plants [1000 MW(e)] and reprocessing plants [40000 MW (e) full load] in the maximum of the concentration.

Emission height :
Nuclear power plant - 100 m , $\bar{\chi} = 3E-7$ s/m³
Reprocessing plant - 200 m , $\bar{\chi} = 1E-7$ s/m³

Nuclide	Release rates (Ci/a)		
	LWR	HTR	LMFBR
H 3	7 E4	6 E4	6 E4
C14	5 E2	3 E3	1 E2
Kr85	1,2E6	2 E6	6 E5
Sr89	0,1	0,1	0,1
Sr90	0,1	0,1	0,1
I 129	0,2	0,22	0,16
I 131	0,16	0,14	0,18
Cs134	0,1	0,1	0,1
Cs137	0,1	0,1	0,1
Pu238	0,013	0,028	0,053
Pu239	0,0015	2 E-5	0,017
Pu240	0,0022	5 E-5	0,02
Am241	0,001	4 E-5	0,01
Cm242	0,07	0,0015	0,18
Cm244	0,012	0,0025	0,006

Tab. 6: Expected radioactive effluents from reprocessing plants [40000 MW(e) full load]
H3 and Kr 85 retention 90 %
Iodine retention 99,5%
Decay time 200d

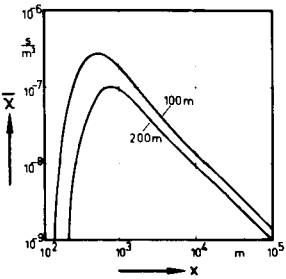

Fig 2 Average long-time diffusion factor $\bar{\chi}$ near ground level in the main wind direction near Frankfurt for a release height of 100 m and 200 m

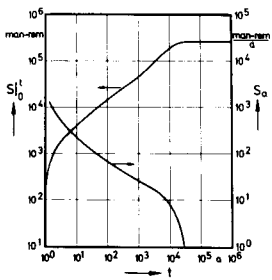

Fig 3 : Collective dose in dependance of integration time $S|_0^t$ and yearly collective dose S_a by a release of 500 Ci C14 (World population 10^{10})

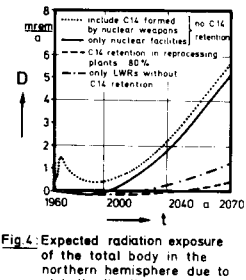

Fig 4 : Expected radiation exposure of the total body in the northern hemisphere due to globally distributed C14 from nuclear power plants, reprocessing plants and nuclear explosions (including future increase of CO_2 concentration)