Investigations on the Aging of Activated Carbons in the Exhaust Air of Pressurized Water Reactors

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Introduction

Nuclear power plants are equipped with iodine filters (adsorbers) containing impregnated activated carbon to keep the release of airborne radioiodine as low as reasonably achievable.[1,2]

Impregnated activated carbons are known to age, i.e. to deteriorate in performance with respect to the retention of radioiodine, particularly in iodine filters that are continuously operated (e.g. containment exhaust air filters of pressurized water reactors). The aging of carbons in iodine filters is essentially due to the adsorption of organic compounds (e.g. solvents) and of inorganic compounds (e.g. O_x, SO_2, NO_x). In the first case the effective surface of the carbon is lowered. In the second case reactions with the carbon (and the impurities such as sulphur) as well as with the impregnant may occur.

In the iodine filters of German nuclear power stations the carbon 207B (KI) (see below) is usually employed. Investigations are being conducted on the retention of methyl iodide ($CH_3^{131}I$) by different carbons as a function of the aging time under simulated normal operating conditions to find out whether longer stay times can be achieved with other carbons, in particular with those developed in recent years.

This paper covers investigations on the aging of five impregnated activated carbons in the containment exhaust air of two German pressurized water reactors (PWR3 and PWR4, respectively) over a period of three months. More details of the measurements in PWR4 can be found elsewhere.[3]

Experimental

The five commercial activated carbons investigated are contained in the table. The impregnants are indicated in parentheses. Three groups of impregnants can be distinguished: KI_x (carbons 1 and 2), KI plus tertiary amine (carbons 3 and 4) and tertiary amine (carbon 5). The base material of the carbons 1,3 and 5 consists of coal (mesh size: 8 - 12), that of the other carbons of coconut shell (mesh size: 8 - 16).

It is mentioned that there are some doubts on the suitability of TEDA impregnated carbons for use in iodine filters of nuclear power plants because of the low inflammation temperature and high volatility of TEDA. However, in recent comparison tests with 207B (KI) and 207B (TEDA) at 180 °C, no difference with respect to the retention of $CH_3^{131}I$ was observed.[4]

The carbons were challenged with the containment exhaust air during power operation of the PWRs. The operating conditions of the carbon beds corresponded essentially to the operating conditions of the iodine filters of German nuclear power stations in normal situations and were largely identical with those in the subsequent laboratory tests with $CH_3^{131}I$. For comparison, also laboratory tests with fresh carbons were performed.

The parameters of the laboratory tests are given in the table. The preconditioning was ≥ 16h for fresh carbons and, to minimize the desorption of pollutants, 1h for aged carbons.

The total bed depth was 50 cm equivalent to a total residence time of 1s. The beds were sectioned to establish the retention or penetration as a function of the bed depth or residence time. The original sequence of the sections was maintained, i.e. the aged carbon was not mixed prior to the laboratory test.

Details on the performance on the laboratory tests are to be found in the literature.[1]

Results

The table contains the penetration of the five impregnated activated carbons by $CH_3^{131}I$ at different aging times, i.e. no aging or aging over 3 months in PWR3 and PWR4, respectively. Values for bed depths of 12.5 and 25.0 cm are given.

The results may be summarized as follows: For the fresh carbons differences in penetration of up to about three orders of magnitude were found (at a bed depth of 12.5 cm). The carbons impregnated with KI_x only exhibited a higher penetration than those impregnated additionally or exclusively with an amine. The lowest penetration was found for 207B (TEDA).

As for the aged carbons, the differences in penetration were up to around one order of magnitude at a bed depth of 12.5 cm and up to about five orders of magnitude at a bed depth of 25 cm. Except

Penetration of Various Impregnated Activated Carbons by $CH_3{}^{131}I$ at Different Aging Times.

Activated carbon		Bed depth (cm)	Residence time (s)	Penetration (%) [a]		
Number	Type			0 months	3 months (PWR3)	3 months (PWR4)
1	207B (KI)	12.5 25.0	0.25 0.50	2.77 (−3) −	2.17 (+1) 1.08 (0)	4.65 (0) 1.21 (−3)
2	727 (KI₃)	12.5 25.0	0.25 0.50	1.52 (−3) −	1.09 (+1) 1.03 (−1)	3.67 (+1) 5.35 (−1)
3	207B (KI, TEDA)	12.5 25.0	0.25 0.50	2.60 (−6)[b] −	2.45 (0) 1.01 (−3)	5.20 (0) 1.77 (−3)
4	Kiteg II (KI, amine)	12.5 25.0	0.25 0.50	3.61 (−5) −	5.84 (0) 2.67 (−2)	1.21 (+1) 2.23 (−2)
5	207B (TEDA)	12.5 25.0	0.25 0.50	1.10 (−6)[b] −	9.85 (−1) < 1.00 (−5)	1.12 (0) 5.80 (−5)

Temperature   : 30 °C
Relative humidity: 40 %
Face velocity   : 50 cm/s

Preconditioning time: ≥ 16 or 1 h
Injection time   : 1 h
Purging time   : 2 h

[a] 2.77 (−3) = 2.77 · 10^{-3} etc.; −: penetration lower than minimum detectable penetration (10^{-5} − 10^{-4} %)

[b] extrapolated from lower bed depths

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for 207B (KI) aged in PWR4, the carbons impregnated with KI_x only showed again a higher penetration than those impregnated additionally or exclusively with an amine. The lowest penetration was again found for 207B (TEDA). There were no significant differences in the results obtained in the two PWRs, apart from 207B (KI). The relatively large difference found for this carbon is not understood.

It is pointed out that also other investigations found a relatively low penetration for carbons impregnated with TEDA or another amine when aged under various conditions.[5,6,7]

It is also mentioned that in the present investigations high loadings with low-volatile organic compounds (mainly decane and dodecane) were detected at bed depths of < 12.5 cm, that is at bed depths with the highest aging. This is in agreement with former former measurements in German PWRs.[8] Changes in the alkalinity of the carbon were not observed.

Summary and Conclusions

In the investigations with aging times of up to three months the penetration by $CH_3{}^{131}I$ was mostly higher for activated carbons impregnated with KI_x only than for those impregnated additionally or exclusively with a tertiary amine (e.g. TEDA). These results seem to suggest that the longest stay times can be obtained with carbons whose impregnant contains a tertiary amine. Results for longer aging times must be awaited for a final judgment.

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