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BEHAVIOUR OF RADIOIODINE IN GASEOUS EFFLUENTS

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Abstract

BEHAVIOUR OF RADIOIODINE IN GASEOUS EFFLUENTS. Because of the different chemical forms in which radioiodine occurs in the gaseous state, it is important when designing efficient filters to know the chemical forms which may be present in the effluent gases when various operations are being carried out and to know the effect of different gaseous environments on the filtration efficiency. To obtain this information it is necessary to have available reliable means of characterizing different chemical forms and to sample gaseous effluents when these operations are being carried out. This paper describes the use for identifying molecular iodine of metallic screens in a multi-component sampling pack in different gaseous environments.

Using multi-component sampling packs, the fractionation of iodine nuclides between different chemical forms was measured in the effluent gases escaping from an in-pile test loop in which the fuel was deliberately ruptured by restricting the flow of coolant. Sequential samples were taken for six hours after the rupture and it was possible to follow during this period the individual behaviours of ^{131}I, ^{133}I and ^{135}I. Simultaneous samples were also obtained of the noble gases in the effluent gas stream and of the iodine nuclides in the loop coolant.

Similar experiments have been carried out with a view to characterizing the different chemical behaviour of radioiodine as it is released from a variety of operations in the nuclear industry including the cutting of fuel sections in metallurgical examination caves and an incinerator.

Introduction

Airborne radioiodine is a mixture of chemical forms, so that filtering contaminated air efficiently depends on knowing both the behaviour of the various forms with filter media under different operating conditions and the relative amounts of each form in the mixture. Laboratory experiments have been useful for finding out how the various forms behave chemically, but they do not give any information about the composition of the mixture generated in nuclear reactor accidents. In this paper I will give the results obtained using a "May Pack" to separate the mixture of iodine forms following a deliberate rupturing of the sheathing of a single fuel element in a reactor. The accident simulated was reduction of coolant flow caused, for example, by either pump failure or partial block in the entrance to a fuel channel.

Description of Experiment

Water coolant flow to a single closed circuit experimental fuel channel in the NRX reactor at Chalk River was progressively reduced until the fuel sheathing ruptured. The reactor was

then immediately shut down and full cooling restored. A large fraction of the UO_2 fuel from the centre three elements of the seven element string was dispersed throughout the coolant.

Fission products are released dissolved in water escaping from small leaks and subsequently become free when the water evaporates. The piping, valves and pumps etc. for the single fuel channel were confined within a $50m^3$ room ventilated at a rate of $20m^3$/min. Air in the ventilating duct was sampled for one hour every hour 6 m from the enclosure using a modified "May Pack".

This consisted of the following filter elements in order:

i. 3 copper screens (100 mesh, 25 mm diameter) etched with hydrogen iodide solution (sp. gr. 1.70) and dried immediately before use.

ii. 1 glass fibre filter (Gelman Type E, 25 mm diameter)

iii. 3 charcoal impregnated filter papers (ACG/B, 25 mm diameter)

iv. 1 tube (16 mm diameter, 40 mm length) containing activated charcoal granules (18/35 mesh)

Coolant water was sampled 15 min. after rupture, separated from suspended solids and analysed 14 days later. The original coolant and water subsequently used to clean out the piping were drained to a catch tank but a leaking pump resulted in flooding of the floor of the small catch tank room. Both the water and air in the room were sampled and analysed four days later.

Each filter in the pack was counted individually using a 100 channel pulse height analyser. Only isotopes of iodine could be detected. All counts were corrected for decay to the time of the rupture.

Results and Discussion

In Table 1 the radioiodine found on each part of the pack is expressed as a percentage of the total in the sample.

During the first two or three hours more than half the iodine appeared on the copper screens, but this decreased slowly thereafter. On the other hand, the fraction found on the charcoal impregnated paper filters increased rapidly and more than doubled during the 6 hr. sampling period.

This trend may have continued as seen from the last column in which is given the composition of the mixture four days later in air above the flooded floor of the catch-tank

Table 1

Distribution of ^{133}I in components of the May Packs

(The other iodine nuclides ^{131}I and ^{135}I behaved identically).

	% of total					
Time	14:25 15:25	15:30 16:30	17:10 18:10	18:25 19:25	19:47 20:47	4 days later
Copper screen	57	59	53	52	46	7
Glass Fibre	15	3	1	1	1	6
Charcoal paper	20	28	28	36	45	64
Charcoal column granules	8	11	17	11	7	22

room. By this time, less than 1/10th of the iodine was found on the copper screens and nearly 2/3rds on the charcoal papers. However, because of the change in conditions the two sets of results are probably not directly comparable.

Throughout the first 6 hours the fraction found on the charcoal column averaged about 10%. This is important since it is this fraction which may contain methyl iodide and removing this particular form from air streams by conventional charcoal filters has proved troublesome in some operating conditions.

In fact, the fraction found on the copper screens is most probably elemental iodine (1) which is readily absorbed by surfaces so that by the time the air stream reached the sampling point, the mixture would have been prefereentially depleted in elemental iodine. Thus the values in Table 1 are minimal for the copper screen fraction and maximal for the other fractions and the maximum contribution from methyl iodide would have been less than 10%.

Despite the large proportion of UO_2 dispersed, less than 0.2% of the iodine fission products it contained were dissolved in the water coolant. Their subsequent escape to the atmosphere would thereafter depend on the rate of coolant leakage from the system.

Reference

(1) Barry, P. J., Health Physics (1968), in the press.

DISCUSSION

F. J. VILES: Did you use a metal or metal oxide fuel? If a metal fuel is used, significant HI may be produced and this raises the question as to where the HI would be collected in the "May Pack".

P. J. BARRY: It was UO_2 fuel. I believe that HI would behave more or less like elemental iodine; i. e. that it would be highly reactive and would be readily absorbed by the copper.

F. J. VILES: We carried out experiments which showed that freshly cleaned copper is not too efficient in collecting HI, but collection would be efficient if oxide were present on the copper.

J. L. KOVACH: Did you calibrate the May Packs used, to see if the copper screen collects all the elemental iodine?

P. J. BARRY: Yes, experiments which we did showed that the three copper screens used would collect at least 90% of the elemental iodine. The remainder could probably be removed by using more screens.