The Characteristics of ^{14}C Chemical Composition in Reactor Coolant Water from PWR

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1. Introduction

Operation of nuclear power reactors produces a wide range of radioactive nuclides by products. The characteristics of ^{14}C that distinguish it from many other radio nuclides produced by nuclear power operation, are long half-life of 5730 years and ease of assimilation into living organisms. Further more, the release of gaseous ^{14}C compounds also result in a global dispersion of the nuclide. it is necessary to control the production in, as well as the release from nuclear facilities. ^{14}C is a weak beta emitter(maximum energy 156 KeV) with both natural and anthropogenic source terms. ^{14}C is produced mainly through neutron-induced reactions with ^{17}O and ^{14}N. A variety of gaseous, solid and liquid wastes containing ^{14}C are generated during reactor operation. The production of ^{14}C in the reactor coolant is almost entirely responsible for the release from PWR at the power plant site. Gaseous releases constitute the main release from this production source; the rest being accumulated in ion exchange resins or released as liquid discharges. This paper presents the results of an extensive investigation initiated by the domestic PWR through reactor coolant, with the general aim of obtaining new experimental data on ^{14}C in terms of concentration and distribution(inorganic and organic).

2. Methods and Result

2.1 sampling

Coolant samples from a wide range of process systems were collected at PWR in Mar.-Nov. 2007. Most of the systems were sampled once a week using pre-evacuated sampling vessels (20mL glass vials with a septum) filled with process water. Sampling was preformed in a closed system without atmospheric contact. To minimize the laboratory work and cost, five samples were combined prior to ^{14}C extraction to give a composite samples(~100mL), representing a certain period of time(generally five weeks during the operation phase).

2.2 Method and procedure for ^{14}C extraction

To separately determine the fractions of inorganic and organic ^{14}C compounds attached to the chemical treatment optimized for 100ml of process water, a method developed by Dr. Manusson and Stenstrom 2005 was used(see Figure 1 for relevant system parameter). Pretreatment was optimized using ^{14}C-labelled Na_2CO_3 and sodium acetate with recovery of >95% for both compounds. The loss of sodium acetate during acid stripping (resulting in overestimation of the inorganic ^{14}C content) was determined to be 1.2%. The ^{14}C extraction from process water generally follows a three-step procedure : gas phase extraction, acid(H_2SO_4) stripping and wet oxidation($K_2S_2O_8$+$AgNO_3$). The procedure allows quantification of organic and inorganic ^{14}C compounds in the gaseous phase, as well as in liquid phase, resulting in four fractions.

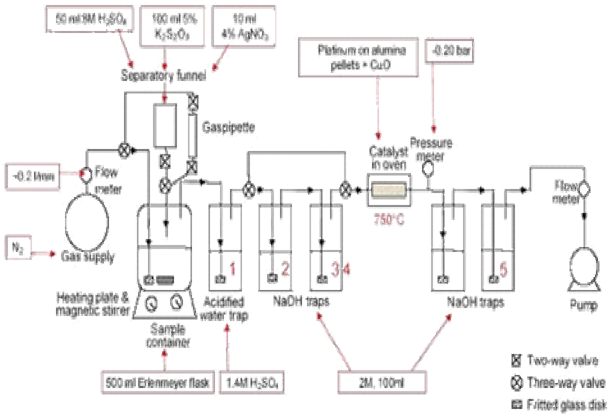

Fig. 1 Basic system set-up used for the extraction of organic and inorganic ^{14}C from reactor coolant.

2.3 Analysis

The sample processing and measurements were conducted in laboratory of KEPRI . Samples collected from the alkaline gas washing bottle after pretreated samples were measured by Liquid Scintillation Counter(LSC) in duplicate to obtain the activities of the inorganic and organic ^{14}C fraction associated with the water sample. The test samples were prepared by adding 3ml of sample solution(2M NaOH) to 17ml of scintillation cocktail(Optiphase Hisafe 3) and then counted for 15 minutes 5 times on a Wallac Guardian 1220 LSC.

2.4 Results and discussion

The variation and distribution of ^{14}C in the reactor coolant of the domestic PWR are shown in Fig. 2. The different lines represent the four fractions (organic and inorganic ^{14}C in gas phase; and organic and inorganic ^{14}C in liquid phase) analyzed separately for each composite sample. According to measuring results during operation period, the chemical composition of

process water samples shows : ~98% organic ^{14}C and ~2% inorganic ^{14}C. A high fraction of organic carbon compounds is expected due to the prevailing reducing condition.
The cover gas in the VCT of PWR systems is predominantly H_2. In a H_2 atmosphere under intense radiation, any free carbon or carbon compounds tend to be reduce to the organic fractions.
Although the results of the measurement indication a strong evidence of ^{14}C specification in reactor coolant, additional data should be evaluated to confirm the quantity of the ^{14}C specification in the PWR environment.

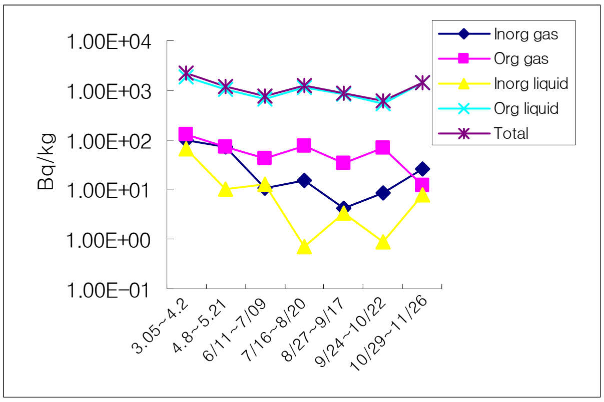

Fig. 2 The variation and distribution of ^{14}C in the reactor coolant from the PWR(2007)

3. Conclusions

The survey provided an overview of the concentration and potential inventory of ^{14}C in process water systems, and of its behavior and variation(inorganic and organic of ^{14}C) during the fuel cycle(Mar.-Nov. 2007)
As expected from the reducing chemical conditions in the coolant, organic ^{14}C compounds generally constituted 92~98% of the total ^{14}C during full power operation. 94% of C_nH_m in liquid and 6% of CH_4-form in gas of the total organic of ^{14}C is showed.

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