Analysis of the Chemical forms on Radioactive carbon in Reactor Coolant during normal operation from Domestic PWRs

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1. Introduction

^{14}C has a half-life of 5,730 years and emits a pure beta ray having a maximum energy of 0.156MeV, and the average energy is 0.045MeV. So far, according to the published papers, $^{14}CO_2$ form prevails at more than 90% in CANDU type reactors, but, in PWR type reactors, the organic compound (mainly CH_4 form) prevails at 70~80%. The organic compound is light, so it is easily dispersed with wind so that a human's internal dose effect is minor. When $^{14}CO_2$ is purged, the effect is severe because $^{14}CO_2$ remains in a human's body as well as in plants via the nutrition acquisition after the photosynthesis reaction in plants. When radioactive ^{14}C is nurtured, the biological effects occur during the process of metabolism, so ^{14}C chemical compound shall be controlled. In this sense, the chemical form of ^{14}C which is purged to the environment should be controlled and monitored. ^{14}C consists of organic and inorganic ^{14}C in the purged air, which is purged to the environment through the plant stack. To evaluate the chemical equipment is required which can separate the organic form and inorganic $^{14}CO_2$ form. This study used the Bubbling method, and we took samples from the reactor coolant system(RCS). By the evaluation of ^{14}C samples that are produced in various types according to plant operation conditions. we got the chemical forms of ^{14}C. This study describes the results of analysis for ^{14}C form.

2. Methods and Results

2.1 the liquid sample's pretreatment

We have performed the analysis of liquid type and gaseous type samples at PWR type reactors. Three reactor types among all the nuclear power plants in Korea were selected considering each set of reactor type conditions.

The configuration drawing of the experimental equipment for liquid sample pretreatment is shown in Fig. 1. The configuration of equipment is as follows: at the bottom area, the temperature controlling hot plate is installed, and the magnetic stirrer stirs the sample for homogeneous mixing. The three connecting lines to the flask are used. First line is for the injection of N_2 gas, and the second line is for the injection of necessary chemicals. The sample is injected through this second line. The third line is for the production gas evacuation line to the next stage. Impurities in this gas are removed by 1.4mol H_2SO_4 solution stored in the acidified water trap vessel. The reason why we use the trap is to remove other impurities except ^{14}C. In the next stage, we use the first $^{14}CO_2$ collection bottle, which contains 2mol NaOH to trap $^{14}CO_2$. Two bottles are also used for the same reason to trap the $^{14}CO_2$ perfectly. The fourth bottle is used for the trap of organic ^{14}C, and the converted $^{14}CO_2$, which passed through catalyst column, is absorbed. The fifth bottle collects $^{14}CO_2$ that is not absorbed during the absorption process in the fourth bottle. Organic CH compounds that passed through second and third stage collection bottles are converted into $^{14}CO_2$ form in the high temperature catalyst reaction furnace (750°C). At the final stage, a vacuum pump is installed for maintaining the negative pressure in the pipeline to protect against the leakage of radioactive gases and the back stream of samples. The water cooling system is operated to maintain low temperature to protect against the evaporation of the moisture, which is composed of production gas and is sprayed with it during the exothermic reaction in the reaction flask.

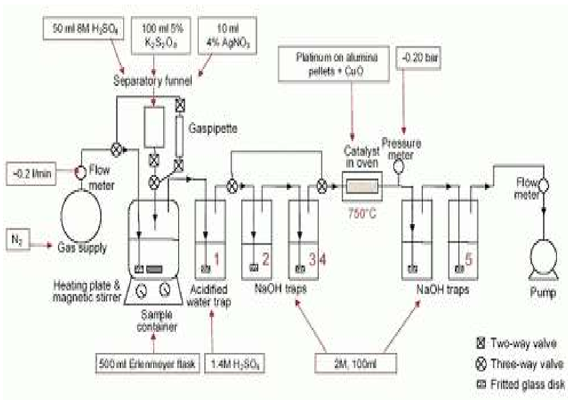

Fig.1 Schematic drawing of ^{14}C process sampler

2.2 Measurements and results

^{14}C behavior at PWR is quite different from CANDU's because the coolant is in deoxidized state due to the injection of hydrogen (25 ~ 50cc/kg) to control dissolved oxygen in RCS. At PWR, the source terms of ^{14}C are the nitrogen remaining in nuclear fuel, the nitrogen which is used for cover gas in the system, and ^{17}O & dissolved nitrogen(NO_2^-) dissolved in condensing water that is recycled by the reactor's make-up storage tank and evaporator. According to the operation condition of RCS, even the ^{14}C inventory shows some differences, but it shows a flat value, as shown in Figs. 2, 3, and 4, which show the monthly basis C concentration in RCS.

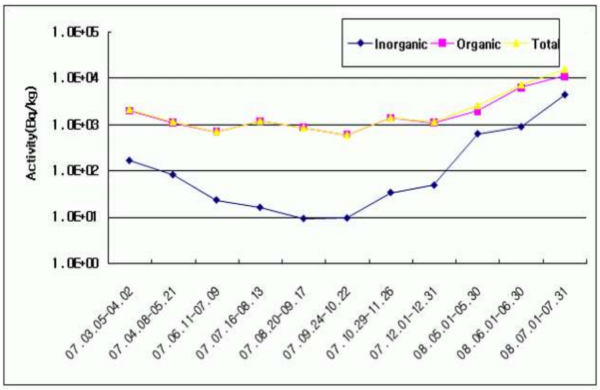

Fig. 2. Chemical form ^{14}C in RCS (W Type)

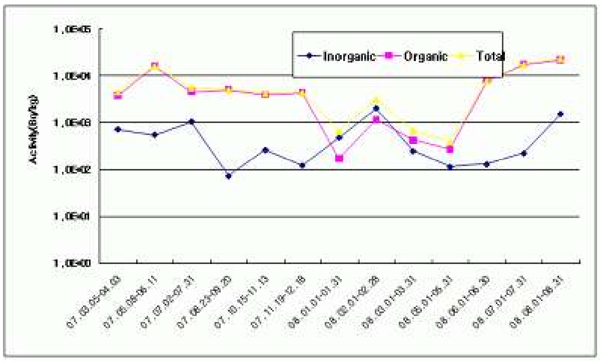

Fig.3. Chemical form of ^{14}C in RCS (CE Type)

Fig. 4. Chemical form of ^{14}C in RCS(Framatome Type)

These investigations were performed for three reactor types during three years.

The organic form of ^{14}C prevailed in RCS at over 84.8%.

During normal operation, organic form and inorganic form were possessed at about 74.6% and 25.4%, respectively, in RCS at W type, 89.9% and 10.1% at CE type, 89.8% and 10.2% at Framatome type. The total average of three reactor types remains inorganic form 84.8% and organic form 15.2%. These values show that organic form of ^{14}C is dominant at PWR type. The W type shows a very low value of ^{14}C concentration in the organic and inorganic states because of the condensed water, which comes from evaporator during condensation operation for the recovery of boron in boric acid's dilution water, is discharged without being reused in the system water. The CE type and W type exhibits a high concentration value in the liquid state organic compound in RCS because the compound remains even though the gaseous phase organic compound in RCS is discharged through the radwaste treatment system during the continuous operation of gas stripper. In CE type, the concentration of ^{14}C is higher than that of the W type because the condensated water in the evaporator is reused after recycling without discharging it into the environment. The Framatome type shows the highest concentration value at 4 and 2 times higher than those of the W type and CE type, respectively.

3. Conclusions

We selected three reactor types, namely the Framatome type, CE type, and W type, to evaluate the chemical forms ^{14}C during the process of discharging and purging ^{14}C to the environment at PWR. Every week we took a 20ml sample in RCS and mixed the samples which were taken during five weeks. From these samples, we found that the organic form of $^{14}CnHm$ prevails at over 74.6% and 89.9% in RCS at the W type and CE type, repectively. The Framatome type showed 89.8% possession. As mentioned in the previous chapter, Framatome uses the concentrated water that is produced during the operation of the evaporator to collect the boron in the water in the hold up tank by recycling without discharging it into the environment. These data evaluate the operation characteristic of ^{14}C behavior and can be useful to control the emission of ^{14}C to the environment.

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