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Advanced Operational Practices for Reducing of Carbon-14 Stack Emissions at CANDU Plant

Wook Sohn
[a]*Korea Electric Power Research Institute, 103-16 Munji-dong, Yusung-gu, Daejeon, 305-380, Korea*
[*]*Corresponding author: wsohn@kepri.re.kr*

1. Introduction

Carbon-14 (C-14), a pure β-emitting radioisotope of carbon with half-life of 5730 years, may be found in the radio-waste from nuclear power plants. Among the nuclear power plants, CANDU plants, which use heavy water as moderator and coolant, emit a larger amount of C-14 than LWRs since heavy water contains a higher isotopic abundance of oxygen-17, one of major source materials for C-14 production at nuclear power plants.

The monitoring for stack emission of C-14, which was initiated at four domestic CANDU units in 1998, revealed that during 1998 and 1999, relatively high C-14 emission rates were observed at three units. In order to find measures to reduce those C-14 stack emissions, a special monitoring program had been conducted from July 2000 to August 2002 at Unit W-3.

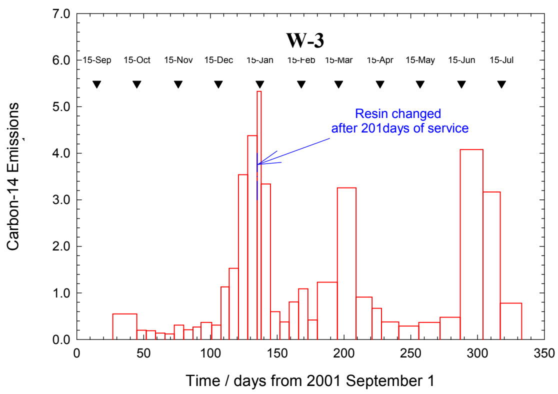

Fig. 1. C-14 stack emissions from Unit 3 during program.

2. Monitoring Program and Results

2.1. Special Monitoring Program

In September 2001, a special monitoring program was initiated at Unit W-3. The program involved i) measurement of C-14 stack emissions over one week (or shorter) periods, ii) measurement of Total Organic Carbon (TOC)/Total Inorganic Carbon (TIC) levels in the bulk moderator water and in the moderator purification return flow at least once a week, iii) measurement of C-14 levels in the bulk moderator water and in the moderator purification return flow at least once a week, iv) detailed chemistry measurements, v) measurement of the dissolved deuterium concentration in the bulk moderator, vi) measurement of deuterium peroxide (D_2O_2) on a routine basis of about 1 week frequency, vii) detailed records of the purging, venting of the cover gas and any other moderator cover gas maintenance, and viii) detailed records of moderator purification IX column service.

2.2. Program Results

The C-14 stack emissions during the program are shown in Fig. 1. After the IX-resin column that had been in-service to 201 days was replaced on day 135 (2002 January 14), the C-14 stack emissions decreased but then peaked to near near day 200 and 300.

The concentration of TIC at the outlet of purification was always less than that at the inlet, i.e., the IX-resin column was always removing TIC from the moderator water (Fig. 2). The TOC concentration in the main moderator water decreased down to about 50 mg kg^{-1} until day about 115 and then remained constant through the IX-column change at day 135. The TOC concentrations in the water exiting the purification system remained essentially constant over the whole reporting period (Fig. 2).

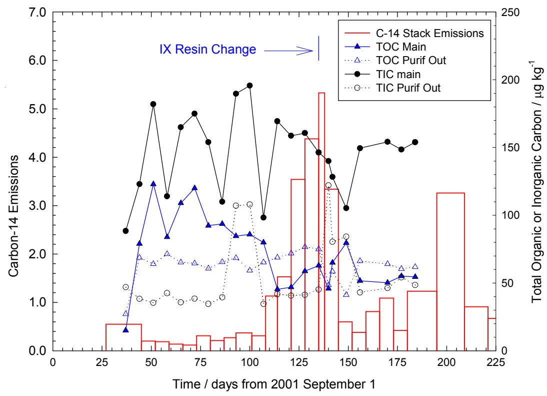

Fig. 2. Correlation of C-14 stack emissions with the moderator TIC/TOC concentrations.

The concentration of dissolved C-14 entering the moderator purification system was always higher than that exiting the system, with the exception of the time period day 115 to day 130 when the peak C-14 stack emissions occurred (Fig. 3). During the time of the peak C-14 stack emissions up to the point when the IX-column was changed, the dissolved C-14 concentrations at the exit of the purification system were always in excess of the concentration in the moderator. Replacement of the IX-resin column had the immediate effect of returning the dissolved C-14 concentrations in the moderator back to the very low levels before the excursion.

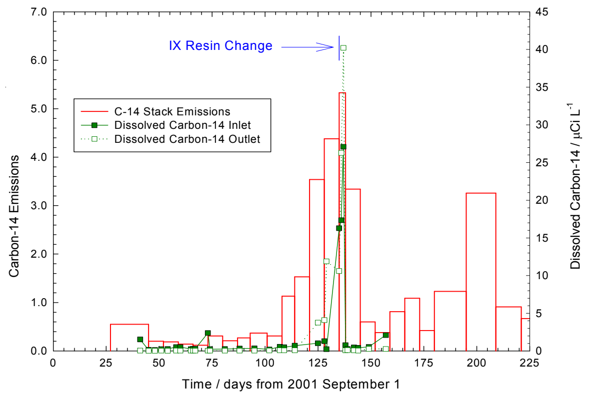

Fig. 3. Correlation of C-14 stack emissions with dissolved C-14 concentrations in the moderator

4. Discussions

Several factors have contributed to a remarkable reduction in the C-14 stack emissions during 2000 (Fig. 1). One of them was the procedure to limit the maximum service time of IX resin in the purification columns to about 80 days to ensure that the weakly absorbing HCO_3^-/CO_3^{2-} was not displaced off the resin by other competing anions. The implications of this practice are that the moderator purification IX columns may be removed from service prematurely, increasing production of radioactive IX resin waste. Therefore, for optimal use of the moderator resins from a C-14 emissions point of view, it is needed to estimate as exactly as possible the exhausting time of the resins with respect to C-14.

Furthermore, columns that had seen previous general purification service were no longer being used for the removal of gadolinium nitrate from the moderator during and following reactor start-up. It had been common practice at a number of CANDU sites to re-use IX columns, which had already seen normal operation service, for gadolinium nitrate removal at reactor start-up, as these columns were not "spent" as far as removal of gadolinium ions and nitrate ions were concerned. However, the IX columns were probably "spent" or close to being "spent" as far as HCO_3^-/CO_3^{2-} were concerned. During the procedure to remove gadolinium nitrate at reactor start-up, the more strongly absorbing nitrate anions displace the HCO_3^-/CO_3^{2-} back into the moderator system.

The IX-column in the moderator purification system was changed out after 201 days of service. The stack C-14 emissions over the last about 175 days of the IX-resin service life indicate that the emissions were acceptably low until about 25 days before the column was replaced. It seems that the only data that provided an operational marker for deciding when to remove the IX-resin column is an observed increase in the C-14 stack emissions themselves. Furthermore, any increase over the rate of 0.4 Ci month-1 for two consecutive weeks may be the indication for an IX-resin column change, especially if the IX-resin column has been in service for more than 80 days. For this, it is needed that the stack emissions continue to be measured on a weekly basis.

As a further measure to reduce releases from the moderator cover gas system, oxygen addition to the cover gas was also carried out to ensure that there is a stoichiometric excess of oxygen over deuterium. This allows for more efficient use of the catalytic recombiner to convert deuterium into heavy water, which has significantly reduced the frequency of purges needed to keep the cover gas deuterium concentration within specifications.

3. Conclusions

The evolution of the moderator chemistry practices at Unit 3 has lead to a significant decrease in the C-14 stack emissions from 1999. These low C-14 emissions have been achieved through: i) changing out the moderator purification IX-resin column after about 80 days of service; 2) discontinuation of the practice of re-using IX-resin columns for removing the gadolinium nitrate from the moderator water at start-up; iii) maintaining an excess of oxygen in the moderator cover gas to ensure the complete recombination of deuterium gas; iv) reducing the frequency of purges required to reduce the deuterium concentration of the moderator cover gas.

In addition, evaluation of the moderator chemistry data suggests that practices of changing the IX-resin column after 80 days may be extended slightly while still maintaining low C-14 emissions from the stack. The data suggests that from a C-14 emissions perspective, the criterion for resin change out can be based on weekly determinations of the stack C-14 emissions at the plant.

REFERENCES

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