A Study on ^{14}C Desorption From Spent Activated Carbon of Air Cleaning Units in Domestic Nuclear Power Plants

HangRae Cho* and Cheon-Woo Kim
Korea Hydro & Nuclear Power Co., Ltd. Central Research Institute, 70, Yuseong-daero 1312beon-gil, Yuseong-gu, Daejeon, Republic of Korea
*hrcho6795@khnp.co.kr

1. Introduction

There are many air cleaning units (ACUs) that purify the exhaust air containing radioactive iodine compounds and micro particles such as organic/inorganic carbon compounds in the radiation controlled area buildings of nuclear power plant (NPP).

The TEDA impregnated activated carbon has numerous pores and adsorbs inorganic carbon dioxide (CO2) and volatile organic compounds (CnH2n+2) containing 14C. The iodine removal efficiency of the activated carbon filters are checked periodically every over-haul of NPP under strict regulation. When the removal efficiency is lower than design value, the activated carbon filters are replaced by new activated carbon and spent activated carbon (SAC) is generated in bulk [1].

It is expected to cost enormous money to dispose of the SAC as intermediate and low level radioactive waste because almost all SAC contains ^{14}C exceeding the severe limit concentration (1Bq/g) for clearance level.

Therefore, we began a study on the ^{14}C desorption from SAC and performed a lot of experiments to seek the best condition for efficient removal of ^{14}C from the pore space and wall surface of SAC.

2. Experiment for ^{14}C desorption

A thermal desorption device of laboratory scale by micro-wave was used with vacuum pump, nitrogen (N_2) purge gas and mass flow controller (MFC, 0~20 SLPM), steam generator and PLC/MMI control system to derive an optimal conditions for almost perfect desorption of hydrocarbon compound containing ^{14}C.

2.1 Physicochemical characteristics of activated carbon

The result of moisture desorption test, thermal gravimetric analysis (TGA), py-GC/MS analysis, SEM-EDS analysis on SAC and new activated carbon showed that the gaseous molecules containing ^{14}C are likely to be adsorbed physically on the surface of pore by van der Waals force [2].

2.2 Preparation of sample for experiment

Three samples among thirty four SAC samples taken from domestic NPPs were selected as representatives of three kinds of ^{14}C concentration level (low, intermediate, high). The ^{14}C concentrations of representative samples were 1.73, 35.2, 128 Bq/g respectively. Each selected sample of 4,000 mL was prepared respectively in consideration of the sample volume of 200 mL per batch and twenty times of experiments for several levels of various parameters such as heating temperature, temperature ramp rate, flow rate of N2 purge, amount of water injection, reduced pressure during heating and after heating.

2.3 Establishment of experimental strategy

Three steps of heating temperature range was decided on the basis of the physicochemical characteristics of activated carbon.

At the first step, sample was heated by near 150℃ and kept for 1 hour under reduced pressure for removal of CO_2 and moisture, various organic compound adsorbed physically in the pores of activated carbon because physically adsorbed material could react with functional group or TEDA at high temperature over 200℃.

At the second step, the heating temperature was increased by the temperature range of 550~600℃ for desorption of the organic compound captured by functional group on the surface of pore and the impregnated TEDA.

At the final step, the heating temperature was increased up to 850℃ with small amount of water oxidant to oxidize partially the wall surface of pores for removal of the ^{14}C bonded chemically to carbon skeleton itself or adsorbed at the edge of carbon skeleton. The water oxidant was injected into the N2 purge gas supply line by the differential pressure between the microwave heating chamber and atmosphere. This step is similar to a reactivation process to regenerate the spent activated carbon.

3. Result of experiment

3.1 Low-concentration sample

The ^{14}C concentration of sample below 2 Bq/g was decreased below 1 Bq/g by single step heating for 60 minutes under the temperature of 400℃ and the reduced pressure of 400 mmHg getting the decontamination factor (DF) of 3.8 higher than the demanded DF of 2 as shown in Fig. 2 and Fig. 3.

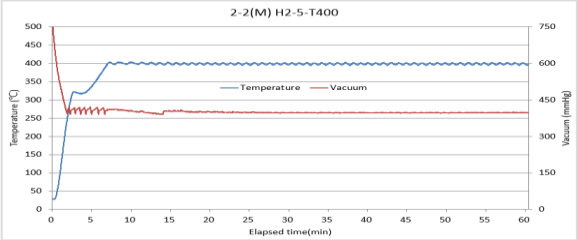

Fig. 1. The graph of single step heating (400℃).

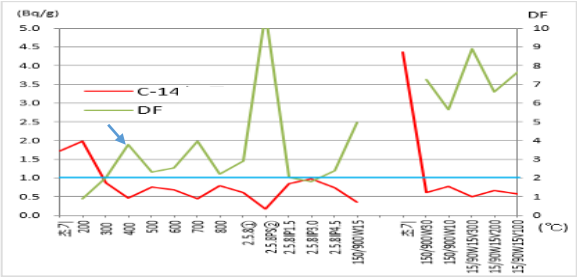

Fig. 2. ^{14}C desorption trend of low concentration sample.

3.2 Medium-concentration sample

In case of medium concentration sample (35.2 Bq/g), the ^{14}C decontamination factor over 50 was demanded to decrease the concentration below 1 Bq/g. However, the DF only reached up to 16 by 3 step heating for 90 minutes under the condition of temperatures of 200℃, 500℃, 800℃ and reduced pressure of 400 mmHg, continuous purge, steam supply at 800℃ as shown in Fig. 3 and Fig. 4.

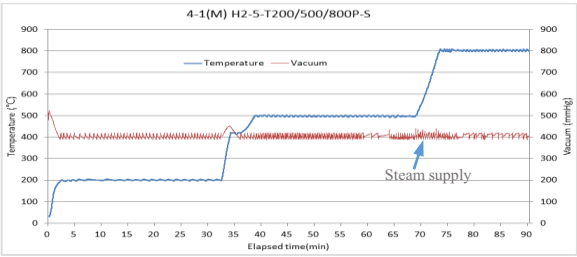

Fig. 3. The graph of 3 step heating (200, 500, 800℃).

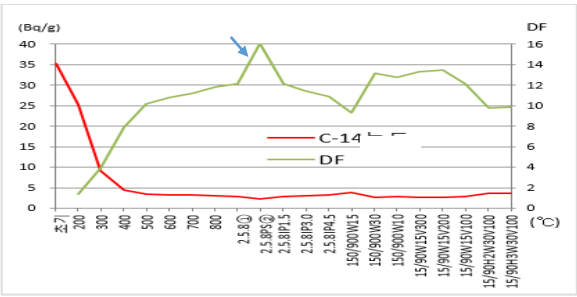

Fig. 4. ^{14}C desorption trend of medium concentration sample.

3.3 High-concentration sample

The ^{14}C DFs of high concentration sample were in the range of 1.5 ~ 19.4 according to the progression of experiment as shown in Fig. 6. In case of high concentration sample (128 Bq/g), the DF of 200 was demanded to decrease the concentration below 1 Bq/g. The maximum DF reached by 2 step heating for 210 minutes under the condition of temperatures of 150℃, 800℃ and reduced pressure of 300 mmHg during heating as shown in Fig. 5 and intermittent purge, water injection at 700℃ and 800℃, reduced pressure of 100 mmHg after heating.

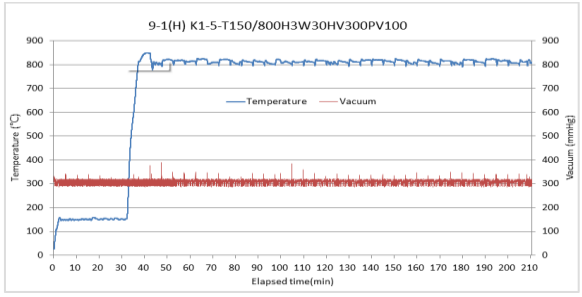

Fig. 5. The graph of 2 step heating (150, 800℃).

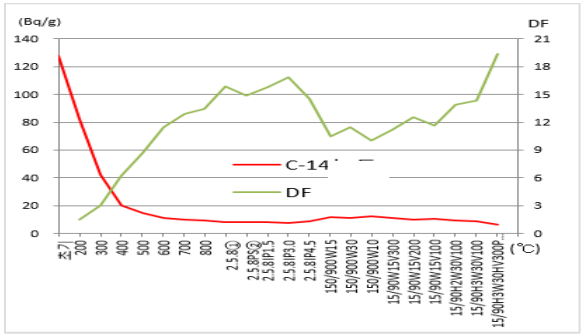

Fig. 6. ^{14}C desorption trend of high concentration sample.

4. Conclusion

The experiments for ^{14}C desorption from SAC were performed with a thermal desorption device by microwave and SAC samples of domestic NPPs to ascertain the possibility to dispose the spent activated carbon as a clearance level waste.

As a result of the experiments, it is expected that SACs below 15 Bq/g in ^{14}C concentration can be changed easily to the clearance level waste by the thermal desorption technology with reduced pressure and appropriate moisture.

However, the last experiment implied that the ^{14}C DF will be more enhanced by the increased heating temperature up to 1,000℃ and more reduced pressure below 200 mmHg and extended heating time.

REFERENCES

[1] U.S. NRC, “Design, Inspection and Testing Criteria for air Filtration and Adsorption Units of Post-accident Engineered-safety Feature Atmosphere Cleanup System in Light-water-cooled NPPs”, Regulatory Guide 1.52 Rev. 4 (2012).

[2] Roop Chand Bansal et al., Activated carbon Adsorption, CRC Press, p.3-10 (2005).