Monitoring of Kr-85 by using BfS-IAR System

Young Gun Ko *, Hyuncheol Kim, Ji Young Park, Sang-Do Choi, Jong-Myoung Lim, Geun-Sik Choi, Wanno Lee
Nuclear Emergency & Environmental Protection Division, Korea Atomic Energy Research Institute, 989-111 Daedeok-daero, Yuseong-gu, Daejeon 34057, Korea
**Corresponding author: ygko@kaeri.re.kr*

1. Introduction

The radioactive noble gas ^{85}Kr is a beta emitter (E_{max}: 687 keV) with a half-life of 10.76 years. Most of the ^{85}Kr in the present atmosphere is derived from anthropogenic sources such as nuclear weapons tests, nuclear-fuel reprocessing and nuclear reactors, although there is natural occurred ^{85}Kr produced by nuclear reactions by cosmic radiation in the upper atmosphere and spontaneous fission of the heavy elements in the Earth's crust [1]. No effective reduction mechanism without radioactive decay is expected because ^{85}Kr is chemically inert [2]. Therefore, monitoring of radioactivity of ^{85}Kr in the atmosphere is a significant and reliable method to detect the nuclear activities of neighboring countries.

A Budesamt für Strahlenschutz - Institute of Atmospheric Radioactivity (BfS-IAR) system is the only commercialized system developed by the Budesamt für Strahlenschutz (Germany) to measure the radioactivity of ^{85}Kr in the atmosphere. In this study, the atmospheric ^{85}Kr concentration of the west of South Korea was measured using the BfS-IAR system. To analyze the atmospheric ^{85}Kr concentration with a high level of accuracy, the trapped amount of ^{85}Kr using by an adsorption module (^{85}Kr sampling module) is very important. One of effective methods to increase the amount of trapped ^{85}Kr is to design the optimum structure of the module. The adsorption/desorption dead zone in the module should be removed to increase the amount of trapped ^{85}Kr. We got the distribution data of pressure, air velocity and temperature in adsorption modules of three cases to investigate the dead zone using a simulation software.

2. Materials and Methods

A Budesamt für Strahlenschutz - Institute of Atmospheric Radioactivity (BfS-IAR) system was used to carry out the process for ^{85}Kr adsorption and analysis after its modification partially. The process is composed of an air sampling system, a pretreatment system and a purification/detection system (Fig. 1).

In the stage of the air sampling, dust, carbon dioxide and water were removed with a filter, a soda lime packed column and two silica gel packed columns before the inflow of air into the activated charcoal packed adsorption module (^{85}Kr sampling module). During the air sampling, the adsorption module is within a Dewar containing liquid nitrogen. The inflow rate of air into the module was 10 m^3/day and the air sampling was carried for 1 day.

In the stage of pretreatment, the desorption process was carried out with helium gas purging by heating the adsorption module at 300 ^{o}C for 1 h. The desorbed gas was transferred to a mini-can (aluminum bottle, 4 bar, 1L).

In the stage of purification/detection, the remained CO_2, N_2 and O_2 gases in the mini-can were removed again with a heater and a liquid nitrogen filled bath, and then Kr in the gas was separated by using preparative gas chromatography (GC). Quantification of the separated Kr was performed by using analytical gas chromatography. Radioactivity of ^{85}Kr was determined using by a gas proportional counter.

The distribution of pressure, air velocity and temperature in the adsorption module was analyzed with an ADINA software (ADINA R&D Inc.). Numerical simulations of air flow into the module was presented. The finite element computational fluid dynamics module of the ADINA software (ADINA-CFD) was used to obtain various data based on the 2D structure of the adsorption module (Fig. 2). ADINA-CFD is a general infinite element code that can be used to model a wide range of fluid-flow problems. Three cases were modeled to select the optimum structure design of the adsorption module (^{85}Kr sampling module).

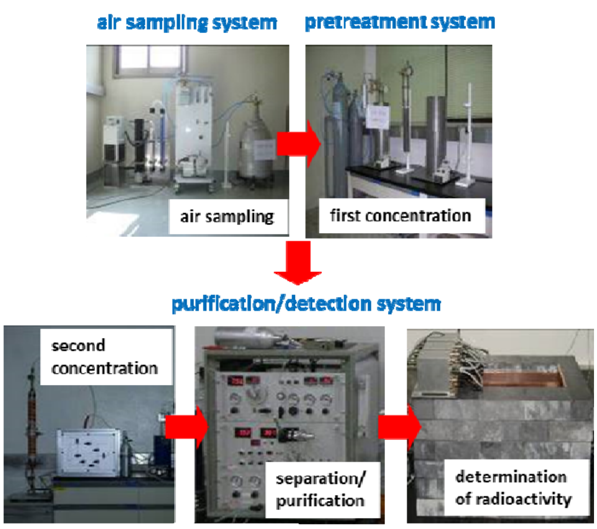

Fig. 1. Composition diagram of the Budesamt für Strahlenschutz - Institute of Atmospheric Radioactivity (BfS-IAR) system.

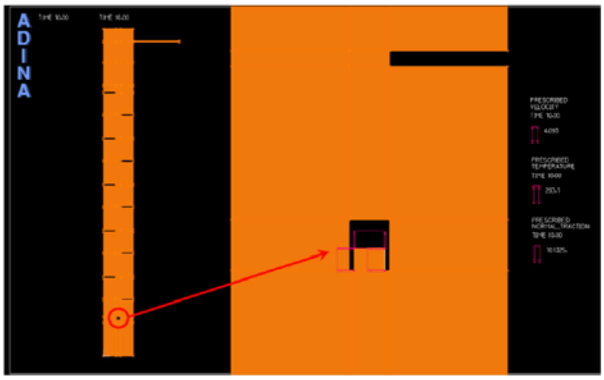

Fig. 2. Basic 2D model for the simulation by using ADINA-CFD.

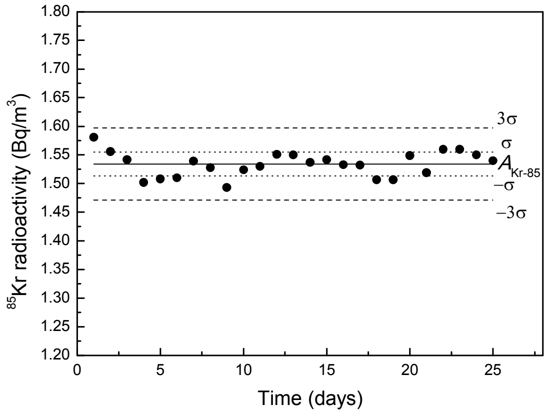

Fig. 3. Atmospheric ^{85}Kr concentration of the west of South Korea measured by the BfS-IAR system.

3. Results and Discussion

The radioactivity of 25 of samples were analyzed. One sample a day was obtained by using the air sampling system of the BfS-IAR system. The obtained mean activity concentration of ^{85}Kr was ca. 1.53 Bq/m^3 at the west of South Korea (Fig. 3). The obtained activity of the west of South Korea is similar with other research groups' data (ca. 1.5 Bq/m^3) [1,3]. The atmospheric concentration of ^{85}Kr is increasing because no recovery operation has been applied to ^{85}Kr in stack gas from reprocessing plants [2]. The standard deviation (σ) was 0.02 Bq/m^3. All data for 25 samples were in the range of 3σ. Therefore, 1.53 Bq/m^3 can be a ^{85}Kr background activity level of the west of South Korea. If the measured value of ^{85}Kr is higher than 3σ, we can guess that nuclear activities such as nuclear weapons tests, nuclear-fuel reprocessing, etc. have been carried out (or were carried out) in the neighboring country near South Korea. The activity of 85Kr is influenced by an air current and generation time. Therefore, the activity of 85Kr at the generation place is not able to be estimated by its detection in Daejeon. On the contrary, the detection time and place of the activity of ^{85}Kr higher than 3σ is not able to be decided owing to the air current and generation time, even though the exact activity of ^{85}Kr at the generation place.

To use the effective the adsorption module (^{85}Kr sampling module) for obtaining low MDA of ^{85}Kr, three structures were designed and simulated with an ADINA-CFD software; (1) flat bottom without a baffle, (2) flat bottom with baffles and (3) cone-type bottom with baffles. The structures were analyzed with data of pressure, air velocity and temperature in the modules. The baffles in the module played an important role for the heat transfer and mass transfer. However, cone-type of the module bottom did not show meaningful results in the mass transfer analysis. The data of combination of pressure and air velocity showed the dead zone in the module clearly.

4. Conclusions

In conclusion, the mean activity concentration of ^{85}Kr in the area of the west of South Korea was obtained (ca. 1.53 Bq/m^3) by using the BfS-IAR system. It required a lot of manpower because all stages to get data should be performed manually although reliable data with a high level of accuracy were obtained by using the system. For this reason, automation of the system is significant and should be rapidly carried out to get continuous data without manpower.

Three cases of the adsorption module (^{85}Kr sampling module) were simulated to select the optimum module with an ADINA-CFD software. The baffles in the module played an important role for the heat transfer and mass transfer while cone-type of the module bottom did not show meaningful results in the mass transfer analysis.

We expect our study will be a stepping stone to develop the automated ^{85}Kr analysis system.

REFERENCES

[1] M. Aoyama, K. Fujii, K. Hirose, Y. Igarashi, K. Isogai, W. Nitta, H. Sartorius, C. Schlosser, W. Weiss, Establishment of a Cold Charcoal Trap-Gas Chromatography-Gas Counting System for ^{85}Kr Measurements in Japan and Results from 1995 to 2006, Technical Reports of the Meteorogical Research Institute No. 54, Meteorogical Research Institute, Japan, 2008.

[2] N. Momoshima, F. Inoue, S. Sugihara, J. Shimada, and M. Taniguchi, An Improved Method for ^{85}Kr Analysis by Liquid Scintillation Counting and Its Application to Atmospheric ^{85}Kr Determination, Journal of Environmental Radioactivity, Vol. 101, pp. 615-621, 2010.

[3] H. Y. Inoue, H. Matsueda, Y. Igarashi, Y. Sawa, A. Wada, K. Nemoto, H. Sartorius, and C. Schlosser, Seasonal and Long-Term Variations in Atmospheric CO_2 and ^{85}Kr in Tsukuba, Central Japan, Journal of the Meterological Society of Japan, Vol. 84, No. 6, pp. 959-968, 2006.