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Elemental Analysis of Metallic and Non-metallic Aerosols Formed by Reaction of Molten Iron with Concrete

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1. Introduction

In the event of a severe accident in a nuclear power plant, many aerosols are formed due to the reaction between the molten reactor core and the basemat concrete, and there is a possibility that radionuclides contact to (or contained in) the molten core are volatilized[1,2]. However, there have been few practical tests confirming this possibility, and published test data are difficult to obtain. On the other hand, it is difficult to use real radionuclides in the tests, because these tests require harsh reaction conditions in which a high-temperature substance and a low-temperature substance are in contact. In addition, the non-radioactive tests require an elemental analysis method with a low detection limit, and there is no prior information about the detection limit required for the molten core and concrete test.

We measured the transportation of metallic and non-metallic components during the reaction of high-temperature molten iron with concrete by using several elemental analysis methods, including the NAA (neutron activation analysis). By comparing the data obtained by each analysis method, each analysis method was evaluated for the analysis of radioactive aerosol behaviors.

2. Experimental and Results

2.1 Experimental

For simulating the molten reactor core, molten iron was prepared using thermite reaction. Concrete containers that react with molten iron were manufactured from Portland cement generally used in nuclear power plants, but coarse aggregates (pebbles) were excluded to form the concrete. Fission products were simulated using non-radioactive isotopes and mainly oxide powders were used. The aerosol formed during the high-temperature reaction of molten iron-concrete was collected through a fiber filter by vacuuming the air around the reactor. In our test conditions, only a fraction of the formed aerosol could be collected in the capture system.

Except for ICP-OES (Inductively coupled plasma - optical emission spectrometry) analysis, aerosol samples were analyzed in the form of aerosol particles. ICP-OES analysis measured the concentration after ashing and acid dissolution of the samples. Elemental analysis was performed by analysis methods such as SEM-EDS, XRF (X-ray Fluorescence), ICP-OES, and NAA.

Fig. 1. Photos of molten iron-concrete reaction test (lower left: filters before and after aerosol capture, lower right: iron and concrete after reaction)

2.2 SEM-EDX analysis

Aerosol samples were analyzed using SEM-EDX (Scanning electron microscope/energy-dispersive X-ray spectroscopy), one of the surface analysis methods. In this elemental analysis, the fiber filter tissue was removed from the sample before the measurement. Table 1 shows the results of EDX analysis of the collected aerosol samples. The relative concentrations of I, Te, and Cs, which are known as volatile elements in fission products, were measured to be relatively high. However, none of the nonvolatile components we put as reactants were detected. The Si, Mn, Fe, and Al components detected here are the major components of concrete and thermite.

Table 1. EDX elemental analysis data of an aerosol sample obtained from molten iron and concrete reactions (filter tissue were removed before EDX analysis)

Element	O	Si	Cl	K	Mn	Fe	Al	S	Na	I	Cs	Te
Normalized wt.%	28.5	3.9	6.2	7.6	8.8	5.1	1.6	1.5	2.6	15.5	6.1	12.6

These results indicate that non-volatile elements are not present in the collected aerosol components or cannot be detected by EDX method. On the other hand, XRF element analysis was performed on the aerosol sample with a fiber filter, but effective components were not detected for metal and non-metal elements.

2.3 NAA (neutron activation analysis)

Among elemental analysis methods, NAA is an elemental analysis method that does not require sample pretreatment and has a low detection limit, but has limitations in its use because it requires a nuclear reactor for neutron irradiation.

Table 2 shows the aerosol elemental analysis results from NAA. In this table, only some of the detected metallic and non-metallic elements are shown due to space limitations in the abstract. A detailed test history and data for all elements are described in the original report[3]. Samples marked in yellow in the Table are samples taken in a test with simulated fission products added, and the rest are blank samples without simulated fission products. According to the NAA results, not only volatile elements such as Te, but also non-volatile elements added as simulated fission products were measured above the NAA detection limits.

Table 2. NAA elemental analysis data of aerosol samples obtained from molten iron and concrete reactions

단위: mg/kg

Sample\Element	Ba	Ce	Nb	Te	Zr	Al	Si	Zn
F-X-09	0.24	-0.02	0.01	2.44	0.18	2.85	16.94	0.54
C-S-01	0.13	0.17	0.02	1.06	0.13	47.88	111.80	22.11
C-X-02	0.62	-0.05	0.07	1.40	0.14	16.34	182.60	31.26
M-X-10	0.76	0.18	0.24	2.94	0.31	4.24	43.11	12.69
I-X-07	0.91	0.03	0.08	9.39	0.16	111.30	339.30	129.90
C-S-03	**10.22**	**0.42**	0.24	**972.20**	0.25	46.38	261.80	113.00
C-S-04	**10.56**	**0.59**	0.27	**885.90**	0.24	108.70	539.50	119.50
C-S-05	4.83	1.18	0.16	8.32	0.30	205.80	576.80	81.67
C-S-06	**0.72**	0.09	**0.30**	**622.80**	0.27	24.69	277.70	91.54
C-S-08	**1.10**	0.09	**0.34**	**885.50**	0.31	36.76	370.60	128.90
C-S-11	**4.66**	**0.88**	**0.26**	**808.00**	0.27	134.10	291.80	134.60
C-S-12	**5.61**	0.15	0.16	**246.30**	0.18	21.93	146.80	81.25
C-S-13	**5.15**	**13.83**	0.12	**805.20**	0.18	124.40	354.40	141.20
C-S-14	**7.25**	**0.83**	**0.33**	**851.10**	0.28	302.30	627.20	109.30

Meanwhile, ICP-OES analysis was performed on the same samples. This method cannot avoid pretreatment such as ashing and dissolution of the sample, but Cs and I are lost in these processes, so they cannot be measured with accuracy. However, for other elements, the analysis results similar to those of NAA were obtained.

3. Conclusions

Various analytical methods were used to analyze the metallic and non-metallic elements contained in the aerosol samples formed by the reaction of molten iron and concrete. Only Cs, I, and Te volatile elements could be detected by SEM-EDX surface analysis. On the other hand, the NAA method was able to measure the relative concentrations of not only volatile elements but also non-volatile elements induced by volatilization at high temperature. For non-volatile elements, the ICP-OES measurement method was able to detect the components of simulated fission products at level similar to NAA results.

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