Aerial spraying to capture released radioactivity from NPP in a severe accident

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1. Introduction

In nuclear power plants, containment envelopes the reactor core and related coolant system to prevent the release of radioactivity into the environment in the event of core melt leading to severe accident. In case of disfunctioning of reactor containment (leakage or rupture) during the accident, radioactivity can be spread out to the environment. The Fukushima nuclear accident in Japan is an example of such calamity. This accident knockedon serious negative impacts on nuclear industry, public health, national economy, and neighboring countries. The radioactive fallout contaminated the land near the nuclear power plant site and the residents in region faced the challenges to clean the contaminated topsoil and foliage. Possibility of future occurrence of such accidents involving the release of radioactive materials to the environment cannot be ignored and therefore, it is very important to have new technological strategies for containing and limiting the spread of radioactivity. The proposed strategy in this paper is the use of aqueous spray (water/foam) mixed with suitable chemical additives to capture, dissolve and stabilize the radioactive gases and aerosol particles released from leaked reactor containment and auxiliary building. The spray system can be approached to the leaked reactor building through the use of a truck with high rising cranes, unmanned aerial vehicles (UAVs, such as helicopters), aerostats, or by installing fixed piping structure around the containment building depending on the accident situation [1].

Laboratory-scale experimental system was setup to examine the performance of such systems. The alkaline water (aqueous NaOH.Na₂S₂O₃) and foam-based spray material (sodium lauryl sulphate) were used to examine capture efficiency of gaseous iodine and aerosol particles.

2. Experimental

A laboratory-scale experimental setup was made inside the hood to evaluate the gaseous iodine and aerosol particle capture efficiencies of water and foam-based sprays. The overall experimental setup was based on the use of iodine gas and aerosol particles to determine the efficiency of capture by the spray. Both alkaline water and foam were used as the material to spray.

The system comprised of five parts based on their functions:

- 1. Spray chamber with nozzle
- 2. Spray solution tank along with spray pump
- 3. Gaseous iodine and aerosol particles sources

- 4. Gas scrubbers to trap un-captured contaminants by sprays
- 5. Sprayed solution collecting tank

The schematic diagram and photographs of experimental arrangement are shown in the Figures 1-2.

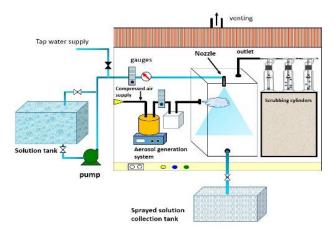


Fig. 1. Schematic diagram of experimental setup

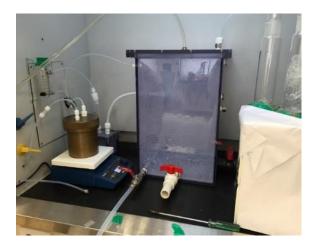


Fig. 2. Experimental arrangement

To enhance the interaction probability of spray droplets with gaseous iodine and particles, the spray chamber design and nozzle selection was based on the goal of spray covering the maximum volume of the spray chamber. Two different spray nozzles (full cone 1/8G3 and hollow cone 1-7N-SS-6 nozzle) were examined to generate water and foam sprays and to evaluate their efficiencies. The nozzles were located at the center of the spray chamber and were oriented down towards the floor of the chamber. To enhance the iodine mass transfer to

the spray droplets and to convert the captured iodine into stable iodine species sodium iodide (NaI⁻), alkaline water of pH 13 was prepared by adding NaOH and Na₂S₂O₃ into water. When iodine reacts with NaOH and Na₂S₂O₃, the following reaction occurs [2]:

$$4I_2 + 10NaOH + Na_2S_2O_3 \rightarrow 8NaI + 2Na_2SO_4 + 5H_2O$$

To generate the foam for the experiment, foaming solution was prepared in a tank by mixing 3 % by weight of foaming agent-sodium lauryl sulphate (NaC₁₂H₂₅SO₄) in water. Homogeneous mixing of foaming agent in water was achieved by circulating the mixture with pump at flow rate of 100 lit/min. A fine grid was placed at the gas outlet port of the chamber to prevent the entrainment of sprayed foam contents to the scrubbing cylinders placed at the outlet. The pressure (Kgf/cm²) and flow rate (lit/min) gauges were installed on spray pipe line to set the desired spray pressure and flow at nozzles.

The amount of gaseous iodine and aerosols captured in the sprayed solutions were determined through UVvisible spectroscopy technique. The UV-Visible spectrometer was first calibrated by obtaining the absorption spectra of standard solutions of NaI⁻ and TiO₂ different concentrations and calibration curves were drawn as shown in the Figures 3 and 4 respectively.

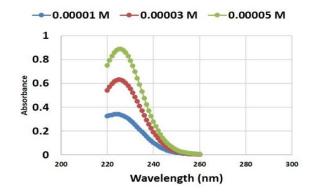


Fig.3. Absorption spectra of NaI⁻ standard solutions

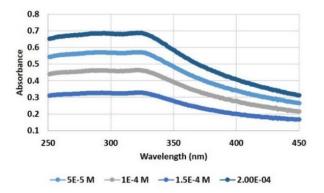


Fig.4. Absorption spectra of TiO $_2$ (0.02 $\mu m)$ standard solutions

In order to develop the understanding of gaseous iodine and particle release and spray behavior in the spray box and to minimize the uncertainties in collection efficiencies, number of pre-experiments were conducted. The procedures for generating and injecting the gaseous iodine and aerosol particles into the spray, spray operation and collection of un-captured iodine gas and particles in the scrubbers placed at outlet of the spray chamber were carefully controlled to be identical in each of the experiments for quality control purposes. The overall mass balance was determined by measuring the iodine and particles amount in the iodine gas and particle generators, supply pipes, sprayed solution in the chamber and scrubbers placed at the outlet of the spray chamber. The residual iodine contents deposited on inner walls of the iodine gas generator and gas supply pipe connected to the spray chamber were determined by carefully rinsing the walls with NaOH solution. The iodine amount dissolved in rinsed solution was determined through the UV-visible spectroscopy.

3. Results and Discussions

3.1 Gaseous iodine removal efficiency by water and foam-based sprays

As radioactive iodine is considered one of the most hazardous source terms that could be released during severe accident [3], the iodine removal efficiency was assessed by comparing the experimental results of alkaline water and foam-based sprays having flow rate of 2 liter/min (Table 1).

Table 1: Comparison of gaseous iodine removalefficiency between alkaline foam and water spray

Nozzle Type	Alkaline foam- based spray efficiency	Alkaline water- based spray efficiency
	(%)	(%)
Full cone	95	74
(1/8 G3)		
Hollow	80	66.7
cone		
(1-7N-SS-6)		

Experimental results showed that the foam-based sprays significantly removed the gaseous iodine in the spray chamber than the alkaline water spray (without foam). As the surface tension of water is large, the ability of capturing gaseous iodine by the water-based sprays is weak On the other hand, the foaming agent reduces the surface tension of water which enhances the capture efficiency of gaseous iodine. Furthermore, in order to attain the high removal efficiency through alkaline water based spray, high spray flow rate and pressure will be required. In this way, water consumption will become higher than foam. Consequently, the foam technology has an advantage over the water based spray in controlling and capturing the gaseous iodine which can minimize the volume of liquid waste. Table 1 also shows the spray removal efficiencies of two different types of nozzles-full cone nozzle and hollow cone nozzle. The iodine removal efficiency using full cone nozzle was greater due to full spray coverage in a round shaped area in the spray chamber as compared to nozzle producing hollow spray pattern.

3.2 Aerosol particles removal efficiency by water and foam-based sprays

The spray efficiencies were also investigated for the removal of aerosol particles (TiO₂) at different volumetric air release rates (30, 45 and 50 liter/min) by fixing the spray flow rate at 2 liter/min. Two different types of spray liquids-water (H₂O) and aqueous sodium lauryl ether sulphate (NaC₁₂H₂₅SO₄) foam were introduced to spray chamber through a full cone nozzle (1/8 G3). The results revealed that water-based spray removal efficiency was increased from 52 to 81.2 % when the total particles release rate was decreased from 50 to 30 lit/min (Figure 3). Whereas the efficiencies obtained from foam-based spray were significantly improved from 70 to 98 % by decreasing the total particle release rate from 50.

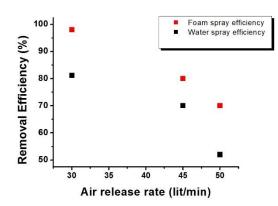


Fig.5. Comparison of aerosol removal efficiency by water and foam-based sprays

The maximum removal efficiency of 98 % was achieved through the foam based spray, whereas the removal efficiency using the water (H2O) based spray was 81.2 % at total gas release rate of 30 lit/min. The reduction in the surface tension of water by adding sodium lauryl sulphate (NaC12H25SO4) in water enhanced the capture efficiency of aerosols. The forces acted by the spray droplets were dominant at lower particle carrier gas release rates and longer residence time of particles in spray region lead to flow rate the high probability of diffusion into the spray droplets. On the other hand, at higher particle carrier gas flow rate, the contact time between the particles and the spray droplets was short and insufficient for the diffusion of particles into spray droplets. Therefore, at high aerosol carrier gas flow rate, most of the aerosol particles escaped from the spray streams and vented through the outlet of the spray

chamber without interaction with the spray droplets. These experimental observations show that in real accident situation of leaked reactor containment or other buildings, spray operation will be more effective for low release rates of radioactive contaminants and spray will be less effective for large releases as a result of large sized hole/crack containment structure.

3.3 Aerosol size impact on spray removal efficiency

As the radioactive aerosols particles released from a nuclear power plant during a severe reactor accident consist of various sizes, it was decided to study the impact of aerosol size on the spray removal efficiency. The aerosols of sizes 0.02, \leq 0.15 and <5 µm were selected in the experiments based on their high mobility in the atmosphere. The spray removal efficiency for the large sized particles having high inertia was higher as compared with small sized particles (Figure 6).

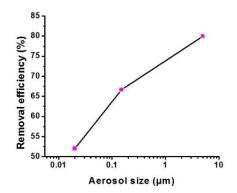


Fig.6. Spray removal efficiency as a function of aerosol size (μm)

4. Conclusion

In this research, we investigated the removal efficiencies of gaseous iodine and particles by alkaline water and foam-based sprays. The removal efficiencies were also assessed as function of particles release rate because the release rate would affect the mitigation performances of spray.

The following observations were made:

- 1. The gaseous iodine and aerosol removal efficiency of foam-based spray is higher when compared with alkaline water-based spray.
- The nozzle producing full cone spray provides the better removal efficiency than nozzle producing hollow cone spray patterns.
- 3. The spray removal efficiency for large sized particles is higher due to dominant inertial impaction mechanism.
- 4. The spray application can contribute significantly in reducing the radiation dose levels near plant site.

REFERENCES

[1] Irfan Younus, Man-Sung Yim, "Out-containment mitigation of gaseous iodine by alkaline spray in severe accident situation," *Progress in nuclear energy.* **83**, 167-176, 2015.

[2] Row, T.H., Parsly, L.F., Zittel, H.E., Design Consideration of Reactor Containment, 1969.

[3] Jungsook Clara Wren , Joanne M. Ball and Glenn A.

Glowa, The chemistry of iodine in containment,

Nuclear Technology, Vol 129, 2000.