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The Localization of Finely Dispersed Caesium Radioaerosols from Off-Gases

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Abstract

Based on fundamental research, we developed the basic scheme of a decontamination setup to remove acid gases (HCl, SO_2, NO_x) from steam-air flows with simultaneous localization of radioactive aerosols, including aerosols with a particle size of less than 0.1 μm and volatile radioactive iodine compounds. The decontamination process is based on the method of the agglomeration cocrystallization of finely dispersed and other aerosols in the gas phase by including the aerosols in the inner structure of large-particle non-radioactive aerosols of ammonium salts, for example, NH_4Cl or $(NH_4)_2SO_3$. For ^{137}Cs, the most ecologically dangerous long-lived radionuclide, the setup ensures a decontamination factor of 10^2 to 10^3 in one decontamination stage. Because the setup consists of three consecutive stages of the same type, the cumulative decontamination factor can reach to 10^6 - 10^7. To localize radioactive iodine as I_2, HI, HOI, HIO_3, and CH_3I from vapor-air flows, the setup uses a unit containing special granulated sorbents based on inorganic compounds. Developed at the Institute of Physical Chemistry, Russian Academy of Sciences, these sorbents effectively remove various radioactive iodine species (inorganic species and methyl iodide) from steam-air flows, ensuring decontamination factors of at least 10^4. The proposed technological scheme will allow vapor-gas flows to be cleaned of radioaerosols and all volatile radioactive iodine species at decontamination factors of not less than 10^4.

Keywords

Aerosols; Caesium; Agglomeration; Cocrystallization

1. Introduction

At present, one of the most important ecological problems is environmental contamination with radioactive substances formed as a result of civil and defense activities of people. A potential source of radioactive contamination is nuclear wastes of nuclear fuel processing enterprises. Treatment of these wastes to obtain final waste forms involves high-temperature processes (calcination, vitrification, ceramic matrix processes, incineration) and formation of off-gases, which can contaminate the environment with radionuclides. The sources of the contamination of off-gases during this process are materials entrained by a gas flow (dust, molten glass particles,

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secondary aerosols), semivolatile materials (Cs, Ru, Tc and the like), and volatile gases (Kr, Xe, I_2). The size of the particles entrained by gas flows from furnaces changes over a wide range (from fractions of nm to 20 μm). ^{137}Cs exhibiting the highest level of activity in the off-gas is a radioactive contaminant of the greatest concern. It is evolved from a glass melter in the form of semivolatile compounds, which are condensed in the off-gas system as nanometer aerosols. It is therefore one of the most difficult contaminants to remove and, as such, controls the decontamination factors (DF) requirements in the off-gas systems. The caesium release to the off-gas largely depends on the melter plenum temperature, on the poll coverage, and on the melter design. Typical values of caesium losses range between 1% and 10% for liquid-fed ceramic melters [1]-[3].

A suitable combination of equipment is required for the removal of all contaminants from the process of solidification of off-gas, so that permissible emission values should be reached, while the choice and the arrangement of individual cleaning devices must depend upon the characteristics of a given off-gas system (e.g., slow rate, temperature composition etc.), but the basic concept is generally similar to that of a multistage washing procedure followed by filtration.

A multistage washing procedure involves, in both already operating and projected waste solidification plants, a range of facilities such as dust scrubbers, demisters, quenchers, wet Venturi scrubbers of various designs, high-efficiency mist eliminators, etc. The number of such units in these systems varies from 4 (AVM Process, France) to 5 (AECL, Canada; China; Pamela Process, Germany) and even to 7 (DWPF, USA) [4]. The DF at each of these stages fall within a wide range from 6 to 200 and ensure (or for projected facilities, specify), including filtration from high-efficiency particulate air filters (HEPA), a total DF of about 10^7 - 10^{14} for various particles of high-level wastes [4] and about 10^5 - 10^8 for low-level wastes [5]. However, all operating waste processing facilities have much lower DFs for caesium, which, as noted above, is due to the formation of nanoparticles and hydrophobic aerosols. HEPA filters trap nanoparticles with a relatively high DF (up to 10^3) [4] [6], leading to a high total DF of the entire facility. However, these filters either have significant disadvantages decreasing the reliability of these filters (they cannot be used repeatedly, and their efficiency decreases in the presence of moisture; resistance to a gas flow is very high), or are at the stage of development and introduction.

The main principle of the current dust traps is based on the precipitation of suspended particles on various surfaces (gratings, drops or liquid films, fiber and electrode surfaces), using such phenomena as gravitation, inertia, diffusion, electricity, magnetism, and supersound. Some of these phenomena have been used to advantage for a long time, some are at the stage of development (diffusion in condensation-type dust traps and vapor-ejection facilities, electrostatic forces in cyclones, scrubbers, and filters using the effect of the precharging of suspended particles) and some are the object of scientific research (catalytic diffusion phoresis, supersonic treatment) [7]. A full review of modern off-gas treatment technologies for application to thermal treatment of wastes was made in [8]. All the processes and the related equipment are based on the above-mentioned principle of precipitation of particles. For the most promising plasma torch process, the efficiency of the separation of toxic metals from off-gases is expected to be less than 35%. Moreover, this process will also use the traditional Venturi scrubber/packed-bed scrubber combination. This low decontamination factor is due to the fact that in the plasma torch process, a large part of the particles formed in off-gases (up to 70 wt %) have a size less than 0.7 μm. In the rotary kiln process, the content of these particles is less than 60 wt % [8].

The choice of the type of a dust trap is based on the properties of trapped particles. However, the particle size is still the deciding factor. Larger particles require less effort for their precipitation and consequently, simpler equipment. The particle size significantly increases during condensation of steam from a vapor-gas mixture on these particles, and this one of the factors of effective purification in condensation-type dust traps. These dust traps are able to trap particles with a size less than 0.2 μm (supersonic Venturi scrubber) and 1 μm (jet scrubber) [4]. Nevertheless, the DF value provided by these facilities is only equal to 10 and 20, respectively.

Thus, the currently used and developed off-gas treatment facilities are comparatively large, energy-consuming, expensive, and not effective enough. According to the [9], the cost of each can of vitrified high-level wastes is more than US$ 1,000,000. Therefore, decreasing the cost of the decontamination of off-gases from ecologically hazardous releases will significantly decrease the cost of the nuclear waste treatment and disposal.

2. Results and Discussion

We suggest a basically new method of localization of caesium radioaerosols, which is based on the physicochemical processes occurring in the gas phase [10]-[12]. This new approach to the protection from radioaerosols

uses unconventional methods based not on the filtration of the gas flows, but on the extraction of radioaerosols from them by the formation of mixed agglomerates with man-made hydrophilic nonradioactive macroaerosols.

We studied the interaction of radioactive caesium iodide aerosols with ammonium chloride and ammonium sulphite under various conditions of the crystallization of these compounds using the set-up described early [13]. Caesium iodide radioaerosols were produced using a stainless steel generator, on which a solution ^{137}CsI was deposited. The fundamental investigations show that aerosols, which were prepared by evaporating CsI from a metal surface heated up to 1300 K in air or argon, contain the three groups of particles (1 - 10 nm, 0.1 - 5.0 μm, and 50 - 200 μm) [14]. The number of the particles of the first group is comparatively large and is 2 orders of magnitude higher than that of the other two.

In order to establish the possibility of the formation of mixed agglomerates of radioactive caesium with non-radioactive ammonium salt aerosols, we studied the behaviour of ^{137}Cs during the formation of NH_4Cl and/or $(NH_4)_2SO_3$ from the steam-gas phase [15]. We suggested that under specific conditions, caesium radioaerosols could be the cocrystallization center for ammonium salts and thereby form mixed aerosol agglomerates.

Microscopic studies proved our assumption about the formations of mixed ^{137}Cs-NH_4Cl or ^{137}Cs-$(NH_4)_2SO_3$ agglomerates. The agglomerate of the mixed aerosols measures 2.0 μm and includes different ^{137}Cs aerosols (less than 0.1 μm) [15]. The most stable mixed agglomerates, whose crystallization centre is the radioactive aerosol, are formed, if the mixing of gaseous HCl (and/or SO_2) with the vapor-gas mixture is carried out separately from their mixing with NH_3. The same order of mixing is also necessary for the full cocrystallization of caesium aerosols with ammonium compounds. Thus, in the new set-up, the mixing of the air mixture with HCl (and/or SO_2) should be followed by the mixing of the resulting mixture with NH_3. The efficiency of the set-up will be determined primarily by the completeness of the mixing of the air mixture with HCl (and/or SO_2).

Based on the results of our fundamental studies, we have the patent [16]. Also, in collaboration with the Zhukovskii Central Institute of Hydrodynamics (Moscow), we have designed a new off-gas cleaning facility which uses this principle (**Figure 1**). The facility consists of 2 - 3 sections. Each section of this facility consists of a zone of gas mixing, where a man-made aerosol is formed, and a zone of the aerosol precipitation. After the off-gas is passed through these sections, the radioactive caesium decontamination coefficients are about $10^{2.5}$ - 10^3. Because at each stage, particles with the same parameters (particle size and their character) are produced, after the off-gas flow is passed through three consecutive sections, the decontamination coefficient increases up to 10^7 - 10^8.

The mixer is designed for the injection and mixing of the components added to the off-gas flow. This mixer is in the form of a cylinder with net partitions inside (**Figure 2**). Two nets (No. 1 and 4) are “active” and are made of tubes. Compressed air with the components (net no.1, hydrogen chloride and/or sulphur dioxide and net no.4, ammonia) are fed into the tubes through a ring collector, into which air with appropriate components is fed from an ejector-mixer with the vortex of the flow. From the tubes of the active nets, compressed air with the components is injected through a system of holes (diameter ~1 mm) into the off-gas flow passed through this net. Thus, a uniform distribution of the components along the cross-section of the mixture is achieved, and a sufficiently high uniformity of the distribution of the components added to the off-gas flow will be attained by the instant of the injection of ammonia near net No.4. The interaction of ammonia with hydrogen chloride and/or sulphur dio-

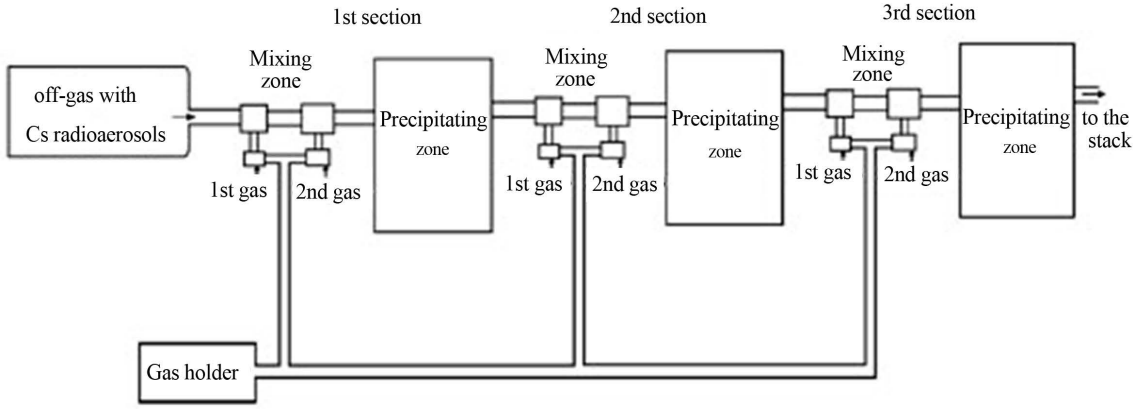

Figure 1. New decontamination set-up.

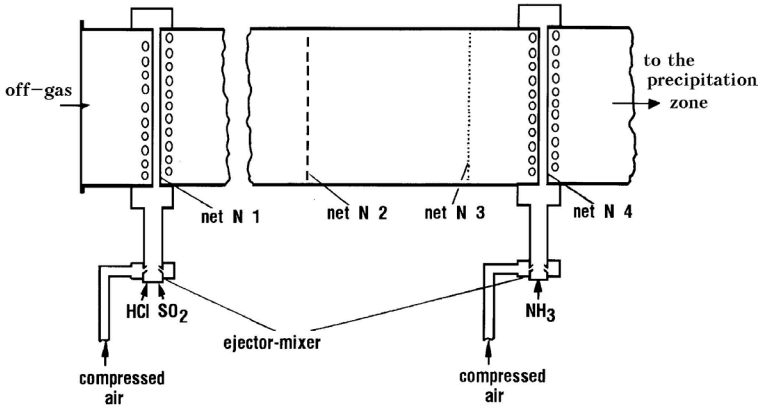

Figure 2. The mixer.

xide behind net No.4 results in the formation of a solid suspension of ammonium chloride and/or ammonium sulphite. This suspension traps the different forms of caesium radioaerosols. The two latter entered in the structure of NH_4Cl or $(NH_4)_2SO_3$ as crystallization centres; *i.e.*, these compounds are in the most stable state. From the mixer, the off-gas flow with the solid suspension of ammonium chloride and/or ammonium sulphite goes to the precipitation zone (for example, scrubber-type dust trap or a foam gas-cleaning apparatus [6]), where an efficient precipitation of the solid particles takes place. At this stage, excess ammonia, which was added to the system for the precipitation of NH_4Cl and/or $(NH_4)_2SO_3$, is also removed from the off-gas mixture.

Note that, in contrast to familiar filtration systems, this facility also solves the problem of its decontamination, because the resulting precipitate composed mainly of readily soluble inorganic salts can be washed from the walls with water and directed to a disposal.

Moreover, this facility can be used either as an independent unit or in addition to already existing filters. The design of this facility allows for its autonomous operation without electric energy supply and at any gas flow velocity. A direct-flow operation of this facility (as compared to traditional filters) ensures its long-term operation at any concentrations of aerosols in the gas phase without an increase in the resistance to the flow passing through this facility.

The main advantages of the proposed method and design facility are:

1. high-efficiency localization of radioactive nanoparticles (with a size smaller than 0.1 μm);
2. possible operation in a closed cycle;
3. use of individual the gas flow components (NH_3, SO_2, HCl) for the formation of mixed agglomerates with finely dispersed radioaerosols;
4. simplicity and low cost of the equipment and possibility of using typical equipment.
5. possibility of including this technology in any technological off-gas treatment line.

Note that an analogous process was suggested in Japan (Chin-Nagoya thermal power plant, Chubu Electric Company [8] [17]) for an additional cleaning of coal-fired flue gas from acid gases.

3. Conclusion

In conclusion, it should be noted that for the final estimate of the efficiency of the proposed facility and the refinement of its geometrical and hydrodynamic parameters, it is necessary to carry out experimental tests on a model and a pilot set-up.

Announcement

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