

APPENDIX 14A

CHEMICAL SPRAYS

INTRODUCTION

A chemical additive has been added to the reactor building spray for the purpose of removing any radioactive iodine which may be released following a loss-of-coolant accident. The chemical spray solution is a boric acid solution containing sodium thiosulfate and enough sodium hydroxide to form an alkaline pH buffer with the boric acid. This solution composition is commonly referred to as "alkaline sodium thiosulfate."

The sodium thiosulfate in the alkaline sodium thiosulfate solution reacts rapidly, completely, and irreversibly with iodine, and was, in fact, suggested by Griffith⁽¹⁾ in his 1963 study on the use of sprays for removing iodine from reactor building atmospheres. The sodium hydroxide is not intended to play an active role in the absorption of iodine; its function is merely to preserve the long-term stability of the alkaline sodium thiosulfate solution by maintaining the desired pH.

Data presently available ⁽²⁻⁷⁾ indicate that the alkaline sodium thiosulfate solution is satisfactory when used in a properly engineered system.

IODINE REMOVAL EFFECTIVENESS

ORNL has conducted a number of spray tests in the Nuclear Safety Pilot Plant (NSPP) facility. These tests, as reported in ORNL-4253⁽⁷⁾, have demonstrated that elemental radioactive iodine is rapidly removed by chemical sprays.

Using an NSPP run made at accident conditions closely approximating those predicted for Midland Units 1 and 2, the measured iodine half-life was 31 seconds; that is, half of the radioactive iodine was removed from the steam-air atmosphere in 31 seconds after starting the sprays. These data have been scaled to the Midland Units 1 and 2 design. They result in an iodine half-life of 28 seconds with the full spray installed capacity operating (2350 gpm) and a half-life of 56 seconds at half capacity. The iodine half-life reported in Section 14.2.2.3.7 is 61 seconds at full capacity and 121 seconds at half capacity.

A large number of confirmatory tests ⁽⁷⁻¹²⁾ have been made which demonstrate that chemical sprays are effective for iodine removal. These spray tests have been made using a wide range of variables - spray distributions with droplet sizes ranging from 100 to 1200 microns, fall heights ranging from a few feet to approximately 50 feet, temperature and pressure conditions varying from ambient to maximum accident conditions, iodine concentrations ranging from 1 to 130 mg/cubic meter, single and multiple spray nozzle installations, spray fluxes ranging from 0.007 to 0.2 gpm per square foot vessel cross section, and condensing and noncondensing conditions. With this wide range of confirmatory test conditions, we are confident that the Midland Units 1 and 2 chemical spray systems will perform as predicted.

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ORGANIC IODINE

The organic iodine consists primarily of methyl iodide but includes a small, almost insignificant, fraction of other organic iodides.

Experimental data obtained under a wide variety of conditions on the amount of methyl iodide released from overheated fuel are reported in numerous publications. Iodine release experiments using irradiated Zircaloy-clad UO_2 fuel in a PWR accident environment show less than the 5 percent organic iodine assumed in Section 14.2.2.3.7. Six tests⁽¹³⁾ were performed at Battelle-Northwest Laboratories. They found that 1 percent or less of airborne iodine was in the organic form. Thirteen other experiments were performed in England⁽¹⁴⁾; all but two show less than 0.2 percent as methyl iodide. The highest result was 3 percent. There are a number of other experiments reported in the literature which deal with the amount of nonreactive iodine released from overheated fuel. Some of these experiments have observed greater than 5 percent nonremovable iodine; however, these experiments were conducted under conditions that are not applicable to the PWR accident environment. It is on the basis of all the data above that we concluded that 5 percent nonremovable iodine was a conservative value for use in the accident analysis.

Spray tests at Oak Ridge National Laboratory^(7, 15) and Battelle-Northwest Laboratory⁽¹⁶⁾ have demonstrated that alkaline sodium thiosulfate spray is effective for removal of methyl iodide. While the removal rate is not as dramatic as that for elemental iodine, the methyl iodine removal rate is sufficient to make a significant reduction in the airborne iodine concentration and thus in the off-site doses.

Spray test A-12 in the Containment Systems Experiment (CSE)⁽¹⁶⁾ demonstrated that alkaline thiosulfate spray removes methyl iodine from a steam-air environment with a half-life of 80 minutes. Scaling this to Midland Units 1 and 2 predicts a methyl iodide half-life of 52 minutes.

PARTICULATE IODINE

A small fraction of the iodine assumed to be released following the LOCA may attach to particles and dust to form particulate aerosols. Several studies have shown that, should the particulate aerosols be formed, they will be rapidly removed from the reactor building atmosphere.

Stinchcombe and Goldsmith^(17, 18) have shown that submicron particles are removed with 95 to 99 percent efficiency by condensing steam. They also show that the thermal and vapor pressure gradients, which exist in the condensing steam environment, drive the submicron particles toward the cooler surfaces where they are removed from the atmosphere with the condensing water vapor.

Particles greater than 0.2 micron size are removed efficiently by impaction with the reactor building spray; those particles less than 0.2 micron size are not. However, the smaller particles serve as condensation nuclei which grow until gravitational and inertial forces result in rapid deposition of these particles.^(17, 19) The absence of particles, after aging, demonstrates the effectiveness of this removal mechanism. Likewise, very small thermal gradients act as driving forces which cause migration of particles to the spray drop surfaces and thereby enhance removal.⁽¹⁷⁾

High concentrations of small particles are very unstable and rapidly agglomerate into larger particles.^(20, 21) These are effectively removed by impaction with spray drops, by washout from the condensing steam, and by settlement. As a result, nearly all particulates are expected to be removed from the reactor building atmosphere.

Experiments in the CSE⁽⁸⁾ have demonstrated rapid removal of particulates by spraying. These tests indicate removal rates in a condensing steam environment - similar to that in the reactor building following an LOCA - of one third to one half as fast as for elemental iodine.

HYPOIODOUS ACID

Hypoiodous acid is a product of elemental iodine hydrolysis by alkaline aqueous solutions.⁽²²⁾ The addition of sodium thiosulfate to an alkaline solution precludes formation of hypoiodous acid in any significant amount. Sodium thiosulfate reacts instantaneously with elemental iodine, reducing the iodine to iodide. Thus, no elemental iodine is available for hydrolysis to the hypoiodous form.

REFERENCES

- (1) Griffith, V., The Removal of Iodine From the Atmosphere by Sprays, AHSB(S)R 45, 1963, pp 7-12.
- (2) Cottrell, Wm. B., ORNL Nuclear Safety Research and Development Program, Bimonthly Report for July-August 1967, ORNL-TM-1986, p 35.
- (3) Cottrell, Wm. B., ORNL Nuclear Safety Research and Development Program, Bimonthly Report for March-April 1968, ORNL-TM-2230, p 81.
- (4) Cottrell, Wm. B., ORNL Nuclear Safety Research and Development Program, Bimonthly Report for May-June 1968, ORNL-TM-2283, pp 64-73.
- (5) Cottrell, Wm. B., ORNL Nuclear Safety Research and Development Program, Bimonthly Report for July-August 1968, ORNL-TM-2368, Section 3.5.
- (6) Cottrell, Wm. B., ORNL Nuclear Safety Research and Development Program, Annual Report for 1967, ORNL-4228.
- (7) Parsley, L. F. and Franzreb, J. K., Removal of Iodine Vapor From Air and Steam-Air Atmospheres in the Nuclear Safety Pilot Plant by Use of Sprays, ORNL-4253, June 1968.
- (8) Nuclear Safety Quarterly Report; November, December 1967, January 1968; for Nuclear Safety Branch of USAEC Division of Reactor Development and Technology, by the Staff of Battelle-Northwest, BNWL-816.
- (9) Nuclear Safety Quarterly Report; February, March, April 1968; for Nuclear Safety Branch of USAEC Division of Reactor Development and Technology, by the Staff of Battelle-Northwest, BNWL-885.

- (10) Parker, G. W., Reaction of Molecular Iodine and of Methyl Iodide With Sodium Thiosulfate Sprays, ORNL-4076, p 169.
- (11) Nishizawa, Y., et al., "A Study of the Removal of Iodine From the Atmosphere by Sprays," Journal of the Japan Society of Nuclear Power, Vol 8, No. 11, November 1966, pp 598-602.
- (12) Maekawa, T., et al., "A Study on Removal of Iodine From Atmosphere by Spray I - Removal of Iodine by Sprav Under Atmospheric Pressure," Journal of the Japan Society of Nuclear Power, Vol 7, No. 10, pp 563-569, 1965.
- (13) Hilliard, R. K., Coleman, L. F. and McCormack, J. D., Comparisons of the Containment Behavior of a Simulant With Fission Products Released From Irradiated MO_2 , BNWL-581, March 1968, Table 4, p A.5.
- (14) Collins, R. D. and Hillard, J. J., Some Experiments Relating to the Behavior of Gas-Borne Iodine, TRG-R-983(W) UKAEA, Windscale, England, April 1965.
- (15) Parker, G. W., Creek, G. E., and Horton, N. R., Dissolution and Hydrolysis of Methyl Iodine in Misting Spray Solutions, Nuclear Safety Program Annual Progress Report for Period Ending December 31, 1967, ORNL-4228, April 1968.
- (16) Nuclear Safety Quarterly Report; May, June, July 1969, for Nuclear Safety Branch of USAEC Division of Reactor Development and Technology (to be published).
- (17) Stinchcombe, R. A. and Goldsmith, P., "Removal of Iodine From Atmosphere by Condensing Steam," AERE-M-1214.
- (18) Stinchcombe, R. A. and Goldsmith, P., "Removal of Iodine From Condensing Steam," Journal of Nuclear Energy Parts A/B, Vol 20, pp 261-275, 1966.
- (19) Goldsmith, P. and May, F. G., "Diffusiophoresis and Thermophoresis in Water Vapor Systems," Aerosol Science, C. N. Davies, Ed., Academic Press, Inc., New York, New York, pp 163-194 (1966).
- (20) Collings, D. A., et al., "Experience in Trapping Iodine-131 and Other Fission Products Released From AGR-Type Fuel Elements," TID-7677, p 113.
- (21) Keilholtz, G. W., "Filters, Sorbents and Air Cleaning Systems as Engineered Safeguards in Nuclear Installations," ORNL-NSIC-13, p 139, October 1966.
- (22) Parsley, L. F. and Wantland, J. L., "General Considerations F-guarding the Use of Sprays," pp 256-264, Nuclear Safety Program Annual Progress Report for Period Ending December 31, 1968, ORNL-4373.

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