

Proceedings of the Korean Nuclear Society Spring Meeting
Cheju, Korea, May 2001

Introduction of New Decontamination Techniques Using Carbon Dioxide

Kwangheon Park*, Hakwon Kim*, Hongdu Kim*, Moonsung Koh*, Jeongdong Ryu*,
Yangeun Kim**, Bumsik Lee**, and Hyuntaek Park**

¹⁾ Kyunghee University

1 Sochen-ni, Kihung-eop, Yongin-shi Kyongki-do, 449-701, Korea

³⁾ Wolsung Power Plant, Korea Electric Power Cooperation
Yangnam-myun, Kyungju-shi, Kyungsangnam-do, Korea

ABSTARCT

Green technology is being developed up to a point that is feasible not only in an environmental sense, but also in an economical viewpoint. This paper introduces two case studies that applied green technology into nuclear industry. 1) Nuclear laundry: A laundry machine that uses liquid and supercritical CO₂ as a solvent for decontamination of contaminated working dresses in nuclear power plants was developed. The machine consists of a 16 liter reactor, a recovery system with compressors, and storage tanks. All CO₂ used in cleaning is fully recovered and reused in next cleaning, resulting in no production of secondary nuclear waste. Decontamination factor is still lower than that in the methods currently used in the plant. Nuclear laundry using CO₂ looks promising with technical improvements - surfactants and mechanical agitation. 2) CO₂ nozzle decontamination: An adjustable nozzle for controlling the size of dry ice snow was developed. Using the developed nozzle, a surface decontamination device was made. Human oils like fingerprints on glass were easy to remove. Decontamination ability was tested using a contaminated pump-housing surface. About 40 to 80% of radioactivity was removed. This device is effective in surface-decontamination of any electrical devices like detector, controllers which cannot be cleaned in aqueous solution.

1. Introduction

Nuclear power is intrinsically a clean energy source due to its high energy density with less generation of waste. However, the future of nuclear energy does not look bright to the general public in spite of its intrinsic cleanness. The reasons for the low public acceptance may come from the following; the safety of nuclear power and the production of radioactive wastes. Large investment on the increased safety of the plants over a generation makes the nuclear power plant safer in both design and operation. Radioactive wastes are inevitable, since fundamentally energy is released from the fission of nuclei. It is hard to reduce the total radioactivity generated during operation; however, the quantitative amount of radioactive wastes can be reduced by improving operation processes like decontamination. Nowadays, green technology is emerging because of the increased concern on the environment, which already becomes a level of threatening the existence of human being. The final goal of the green technology is to obtain an environmental equilibrium for the comfortable survival of current life forms on the earth. The process in green technology is more active in introducing an environmentally friendly nature into the operating process, rather than adding up additional processes into the conventional process for the improvement on the environmental impact. One possible introduction of environmentally friendly nature is the use of CO₂ as a processing medium. CO₂ is nontoxic, nonflammable, inexpensive and environmentally friendly, and CO₂ is essential for the survival of current life forms in the earth. We can make CO₂ the gas, liquid, and supercritical states relatively easily. The usage of CO₂ as a process medium becomes expanding in many technically important fields due to its environmentally friendly nature. This paper introduces two case studies of applying CO₂ as a cleaning medium in decontamination.

2. CO₂ as a Processing Medium

The Montreal Protocol and its subsequent modifications mandate a phase-out of the manufacture and use of chloro-fluorocarbons (CFCs). Hydro-fluorocarbons(HFC), viewed as potential replacements, can generate corrosive compounds if subjected to high temperatures, and it is not clear whether these compounds satisfy toxicity issues. Carbon dioxide is a nontoxic, nonhazardous, nonflammable material with no ozone depleting potential. If compressed, carbon dioxide becomes a non-polar liquid. In a supercritical state, which is easy to access, its surface tension becomes negligibly small with low viscosity. The diffusivity of supercritical carbon dioxide is as high as that of the

gas, while having the same (or even higher) solubility power as liquid. The solubility of a substance in liquid (especially supercritical) carbon dioxide is very sensitive to the pressure during the process, hence, the system becomes a simple one with high efficiency[1].

The use of carbon dioxide becomes essential in the green technology. In addition to its inherent environmentally friendly nature, carbon dioxide is easy to be purified and recycled due to low latent heat of evaporation. Pure carbon dioxide is currently used as a processing solvent for extraction of caffeine from coffee beans, spices and flavor extraction from the basic food materials, and for medicine production [2-4]. Its limit for non-polar organic solute dissolver can be got over by additive agents in it. Now, one of the most active use of this technique comes to dry cleaning using carbon dioxide. DeSimone in University of North Carolina developed a detergent for liquid CO₂ dry cleaning[5]. Dry cleaning systems using CO₂ like MicellTM and DryWashTM, are in the market for the commercial use. Metal extraction using chelating agent in carbon dioxide is also being developed nowadays. Actinide metal ions in liquid solution can be collected to carbon dioxide flowing through the solution as chelates. Based on this mechanism, an environmentally friendly process, e.g., a dry PUREX process, can be possible[6].

Another field of use of carbon dioxide is dry ice. Dry ice is solid, but it evaporates in air leaving no trace. Dry ice pellet blasting machines are available in the market already. This technique is currently used in cleaning the surface of a structure, and pulling off a thin layer, like paint. Main advantage of this cleaning method is no generation of secondary wastes.

3. CO₂ Nuclear Laundry

The first case study of possible application of green technology into nuclear industry is nuclear laundry. The laundry system using CO₂ does not make secondary waste due to the recycle use of CO₂. Decontamination of oily contaminants is the better than that using water. And, purification of CO₂ is easier.

A decontamination washer for working dresses using liquid and supercritical carbon dioxide were designed and manufactured. The size of reactor for decontamination is about 16 liter. There is an agitator giving mechanical impact to dresses for cleaning particles attached to them. The system is a closed one with purifying and recycling ability of carbon dioxide. CO₂ used in washing is

collected and purified, then reused with negligible loss. The system was made to use either liquid or supercritical carbon dioxide for cleaning. A control system of the carbon dioxide flow was set in a control panel. Figure 1 shows a diagram of the system and Figure 2 shows a CO₂ washer manufactured in this study.

The manufactured decontamination washer was brought to Wolsung nuclear power plants, and installed to check the efficiency of decontamination and the feasibility of usage in nuclear power plants. The elimination of oil from the contaminated dresses turned out to be good. However, the decontamination factor for contaminated dresses was lower than the design goal value. The surface radioactivity of about 12 $\mu\text{Ci}/\text{m}^2$ went down to about 7 $\mu\text{Ci}/\text{m}^2$. It's due to the low removal rate of radioactive particles attached on the dresses. In this study, mainly pure carbon dioxide was test as a cleaning solvent. Usage of Carbon dioxide only does not give satisfactory cleaning in this case. Most of radioactive nuclides in the contaminated dresses were in the form of particles, and efficient particle removal needs two important mechanisms, i.e., detachment of attached particles from the fabrics and holding them in cleaning solvent. The choice of cleaning solvent does not seem critical in the case of particle removal. Cleaning efficiency can be improved by increasing the mechanical impacts (e.g., increased agitation or ultrasonics), and by applying surfactants for increased holding power of solvent. Right now, water cleaning with detergent is the better than pure CO₂ cleaning. However, with improved design and use of detergent can make cleaning using environmentally friendly CO₂ competitive to traditional water cleaning. Reduction of radioactive wastes and recycles of processing solvents become important in nuclear industry in the future, and CO₂ has a strong edge on traditional cleaning solvents in these viewpoints.

4. Dry Ice Snow Jet Cleaning

Decontamination technology is essential in the operation of nuclear power plants. Dry ice snow cleaning is one of the newest cleaning method, which does not make any secondary waste, leaving no or negligible damage on the working surface[7, 8]. We applied dry ice snow cleaning method to decontamination of radioactive contaminants.

A converging/diverging nozzle for producing dry ice snow jet was developed. We put a nucleation/growth volume in the nozzle and made it adjustable. Dry ice snow particle size was measured to be 1 to 10 μm depending on the nucleation/growth volume. Jet penetration depth (the maximum length to which leading edge of jet reaches in the stagnant air) of nozzle is in normal operating

condition was measured to be about 60 cm. Figure 3 shows the jet penetration depth of this nozzle. The removal mechanisms of surface contaminants are momentum transfer, aerodynamic drag force, and partial dissolving and sublimation. When momentum transfer from the incident snow to the surface particles overcomes the adhesive forces, the particles are removed from the surface and removed by the high velocity gas. Momentum transfer is indifferent to the surface particle size, but proportional to the size of incident snow. Aerodynamic drag force is proportional to the diameter of surface particles. Generally, particles which have less than 0.5 mm diameter cannot be removed by only viscous flow. When the carbon dioxide snow impacts the surface, the force on the solid carbon dioxide snow causes formation of a transient liquid phase at the snow-particle surface interface. Surface oily-residues are dissolved by the liquid carbon dioxide and are removed when trapped during re-solidification by the rebounding snow particle. After removing the particle, dry ice snow sublimates.

Using the developed nozzle, a decontamination device was manufactured (Figure 4). A heat supply system is added at the dry-ice-snow gun for the prevention of ice layer formation on the working surface. To see the cleaning power of this device, fingerprint removal test was done. A glass specimen-containing fingerprint was prepared. The fingerprint on the surface was successfully removed. It took about 2 minutes to remove the fingerprints. Work surfaces are usually contaminated with oil and also with dust. A plastic specimen having scratches (mesh size 180) was prepared and contaminated with mixtures of mineral oil and dusts. This specimen was cleaned by the device developed in this study. After 2 minute cleaning, the specimen recovers the original cleanness. This cleaning method seems very effective to targets with complex geometries like scratches, crevices, and holes.

To check the applicability of this device to radioactivity decontamination, moderator pump housing from Wolsung nuclear power plant was chosen and tested (see Figure 5). Three points on the inner housing surface were selected. Radioactivities of before and after cleaning process were measured by the smear method. Cleaning time was set equal at each point. The surface radioactivity certainly decreases after cleaning process. The reduction of radioactivity was about 40 to 80%. The removal rate at each point, A, B, and C was somewhat different, and recontamination was suspected (Table 1). A suction facility is recommended for the prevention of recontamination. This device can be especially useful in surface-decontamination of any electrical devices like detector, controllers which cannot be cleaned in aqueous solution.

5. Conclusion

Green technology becomes essential for the continuous survival of current life forms in the earth. In this study, the applicability of green technologies using carbon dioxide to nuclear industry was examined. CO₂ dry cleaning laundry machine was build and tested in the nuclear facility. Decontamination factor is still low. However, with improvements like increased agitation and use of detergent, it can be competitive to conventional methods. Cleaning using dry ice snow jet was applied to surface decontamination. It turned out to be effective to surface-decontamination of any electrical devices that cannot be cleaned in aqueous solution. These green technologies have a strong edge on traditional cleaning methods in radioactive wastes reduction and in the recycle of resources.

References

1. M.A.McHugh and V.J.Krukonis, 'Supercritical Fluid Extraction - Principles and Practices', Butterworth-Heinemann (1994)
2. E.Kiran and J.M.H.Levelt Sengers, 'Supercritical Fluids - Fundamentals and Application', Kluwer Academic Publishers (1993)
3. E.Kiran and J.F.Brennecke, 'Supercritical Fluid Engineering Science - Fundamentals and Applications', A.C.S. (1993)
4. S.S.H. Rizvi, 'Supercritical Fluid Processing of Food and Biomaterials', Blackie Academic and Professional (1994)
5. J.B.McClain, D.E.Betts, D.A.Canelas, E.T.Samulsk, J.M.DeSimone, J.D.Londono, H.D.Cochran, G.D.Wignall, D.Chilura-Martino, and R.Triolo, 'Design of Nonionic Surfactants for Supercritical Carbon Dioxide, Science 274 (1996) 2049
6. C.L.Phelps, N.G.Smart, and C.M.Wai, 'Past, Present and Possible Future Applications of Supercritical Fluid Extraction Technology', J. Chem. Edu., 73 (1996) 1163
7. Walter H. Whitlock, William R. Weltmer, Jr., and James D. Clark, 'U. S. Patents - 4,806,171', (1988)
8. Applied Surface Technologies Web site, <http://www.co2clean.com>, 1998.

Table 1. The reduction of radioactive nuclides after CO₂ cleaning.

Nuclide	Before (Bg/g)	After (Bg/g)	%Reduction
Ce-144	19.37	10.9	43.5
Ba-140	3.74	2.80	25.0
Cs-137	0.579	0.373	35.6
I-131	0.598	0.273	54.3
Nb-95	316.8	184.3	41.8
Zr-95	245.2	139.7	43.0
Cr-51	40.2	19.7	50.9
Zn-65	2.70	1.20	55.3
Co-60	1.42	0.136	90.5

Table 2. Surface radioactive dose of each point before and after cleaning, and reduction of radioactivity

Surface	Before Cleaning (μ Ci/m²)	After Cleaning (μ Ci/m²)	Redection of Radioactivity (%)
A	0.28	0.05	82
B	0.26	0.15	42
C	1.21	0.65	46

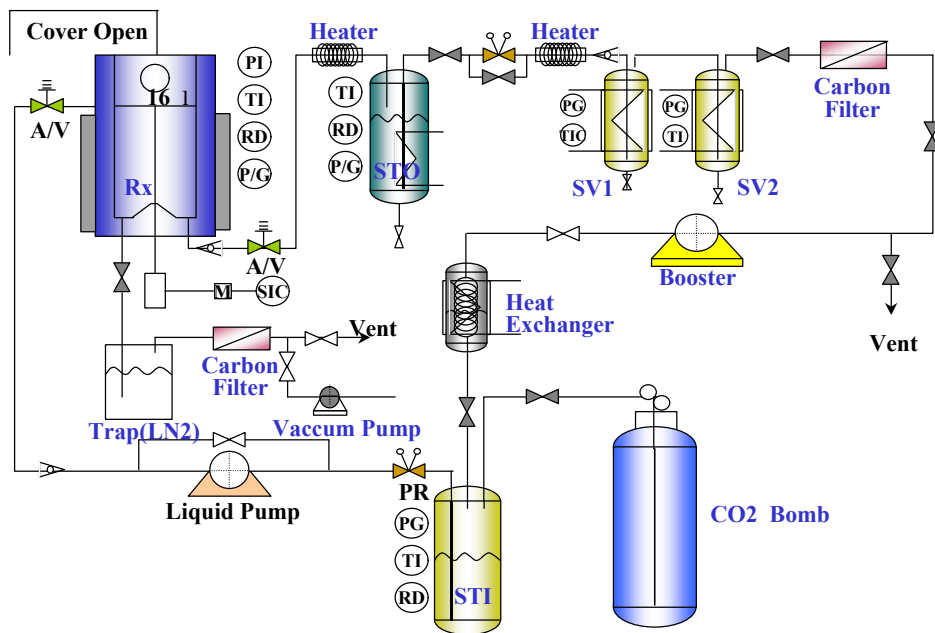


Figure 1. Schematic diagram of carbon dioxide washer.



Figure 2. Manufactured carbon dioxide washer

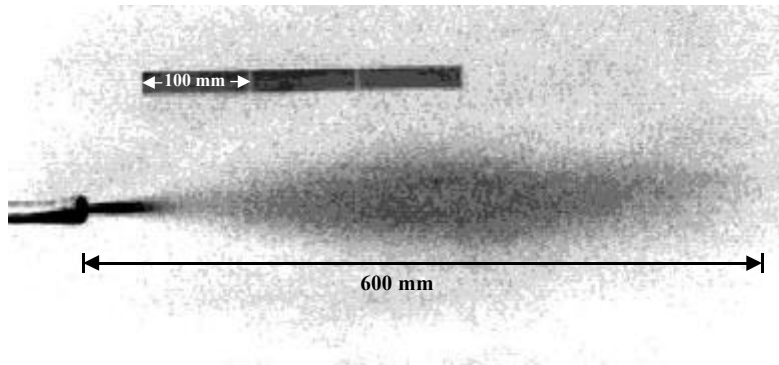


Figure 3. Penetration depth of the developed nozzle



Figure 4. Dry ice snow jet cleaning machine

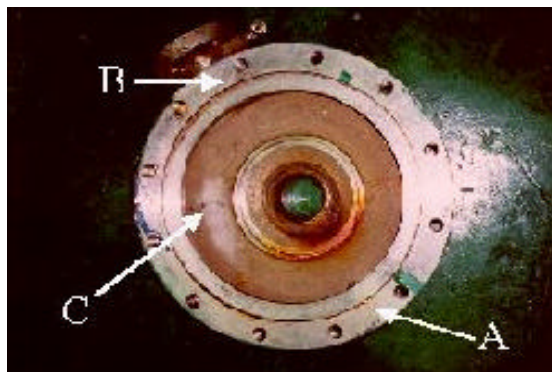


Figure 5. Pump housing after dry ice snow cleaning