

## **PFC Decontamination of a Metal Surface and the Recycling of a Spent PFC Solution**

C.H. Jung, H.J. Won, W.Z. Oh, J.K. Moon, J.H. Park  
Korea Atomic Energy Research Institute  
P.O.BOX 105, Yuseong-gu, Daejeon, 305-600  
Korea

### **ABSTRACT**

PFC(perfluorocarbon) ultrasonic decontamination behavior of loosely contaminated metal specimens such as a plate, pipe, welding and a crevice specimen in a mixed solution of PFC and an anionic surfactant was investigated. Perfluoroheptane( $C_7F_{16}$ ) was used as a PFC ultrasonic media. The contaminants were completely removed for almost all of the tested specimens except for the longest pipe length specimen. For the 6-cm long specimen, 98.5 % of the contaminants were removed. For the recycling of the PFC solution, a distillation test for the spent PFC solution was also performed. The results show that 97.5 % of the PFC was recycled without a loss of the decontamination efficiency.

### **INTRODUCTION**

Chlorofluorocarbons(CFCs) and hydrochlorofluorocarbons(HCFCs) with a potential to deplete the ozone layer should be replaced with alternative compounds which do not deplete the ozone layer nor enhance the global warming problem[1]. PFCs are extremely stable compounds with unique physical and chemical properties that make them particularly suited for some specialized applications. The major applications of PFCs are: (1) semiconductor manufacturing processes[2], (2) fire suppression agents[3], (3) precision cleaning solvents[4], (4) heat transfer fluids or coolants[5], (5) atmospheric tracers[6]. It is also noted that the aluminum-smelting process is also the major generation source of  $CF_4$  and  $C_2F_6$  in industrial applications[7].

The Sonatol process was developed under a contract with DOE[8]. The Sonatol process uses an ultrasonic agitation in fluorinated surfactant solutions to remove radioactive particles from surfaces. Filtering the suspended particles allows the solutions to be reused indefinitely. Their work applies the Sonatol process to the decontamination of heterogeneous legacy Pu-238 waste that exhibits an excessive hydrogen gas generation, and prevents a transportation of the waste to the Waste Isolation Pilot Plant.

Korea Atomic Energy Research Institute is developing the dry decontamination technologies applicable to the decontamination of highly radioactive surfaces, loosely contaminated with radioactive particles. As a part of this project, PFC ultrasonic decontamination technology development has been performed since 2004.

The main objective of the present study is to evaluate the ultrasonic decontamination efficiency of several shapes of type 304 stainless steel specimens in a PFC solution as an ultrasonic medium. The feasibility study of the reuse of the PFC solution by a distillation was also performed.

## EXPERIMENTAL

### Reagents

The PFC solution used in this study was perfluoroheptane(PFC-5070, 3M). Table I shows the physical properties of the perfluoroheptane and water. Anionic fluorinated surfactant containing the carboxylic group was mixed with the perfluoroheptane solution and it was used as an ultrasonic medium.

Table I. Physical Property of PFC and Water

	Water	PFC- 5070
Molecular Formula	H <sub>2</sub> O	C <sub>7</sub> F <sub>16</sub>
Molecular Weight	18	388
Boiling Point,	100	80
Dynamic Viscosity, 10 <sup>-6</sup> m <sup>2</sup> /s	1.06	0.55
Surface Tension, dyne/cm	73	13
Latent Heat, kJ/kg	539	80
Specific Heat, kJ/kg.K	4.2	1.1
Density, g/cm <sup>3</sup>	1.0	1.7

### Experimental Procedure

After weighing the specimen, it was contaminated with an alcohol containing Eu<sub>2</sub>O<sub>3</sub> powder and a fluorescent material. It was then dried in a dark place and photographed, weighed again and dipped into a PFC ultrasonic decontamination chamber. The schematic diagram of the PFC ultrasonic decontamination test equipment is shown in Fig. 1. PFC ultrasonic decontamination medium is a mixed solution of perfluoroheptane and an anionic surfactant. The frequency of the transducer is 28 KHz and the power is 30 W. The decontamination is performed as a dual mode; one is the static mode and the other is the rotation mode. After 5 minutes of an ultrasonic application, the specimen is pulled out, dried and weighed again. To investigate the decontamination surface, it was photographed once more. After a multiple application of the PFC solution, the solution containing the Eu<sub>2</sub>O<sub>3</sub> powder and the fluorescent material was distilled. Before and after a distillation, the turbidity of the solution was measured(Model, DRT 15-CE HF Scientific, Inc.). The FT-IR spectrum for the PFC solution containing an anionic surfactant was obtained by FT-IR spectrometer(Model MB-102, Bomem Co.).

## RESULTS AND DISCUSSION

### Decontamination of a Rectangular Type Metal Plate Specimen

Decontamination efficiency according to a variation of the ultrasonic media was investigated. A rectangular type metal specimen (4 cm X 1.5 cm X 0.1 cm) was used as a surrogate contaminated specimen. Fig. 2 shows a plot of the remaining portion of the contaminants against the application time for water, PFC, and PFC + 0.1 vol% surfactant. For all the three kinds of solutions, most of the contaminants were removed during an early stage of the ultrasonic application. Decontamination factor after a 5 minutes application increased from 20 and 50 to 200 as the ultrasonic media changed from water and PFC to a PFC + 0.1 vol% surfactant solution. The difference in the decontamination factor between water and PFC is explained by the 1) surface tension of water, and 2) a trace amount of an oily component on the metal surface. As the surface tension of water (73 dyne/cm) is larger than that of PFC (13 dyne/cm), PFC is more attractive for the contaminants. From the measurement of the specimen dipped into the water, we found that a trace amount of an oily component remained on the metal specimen.

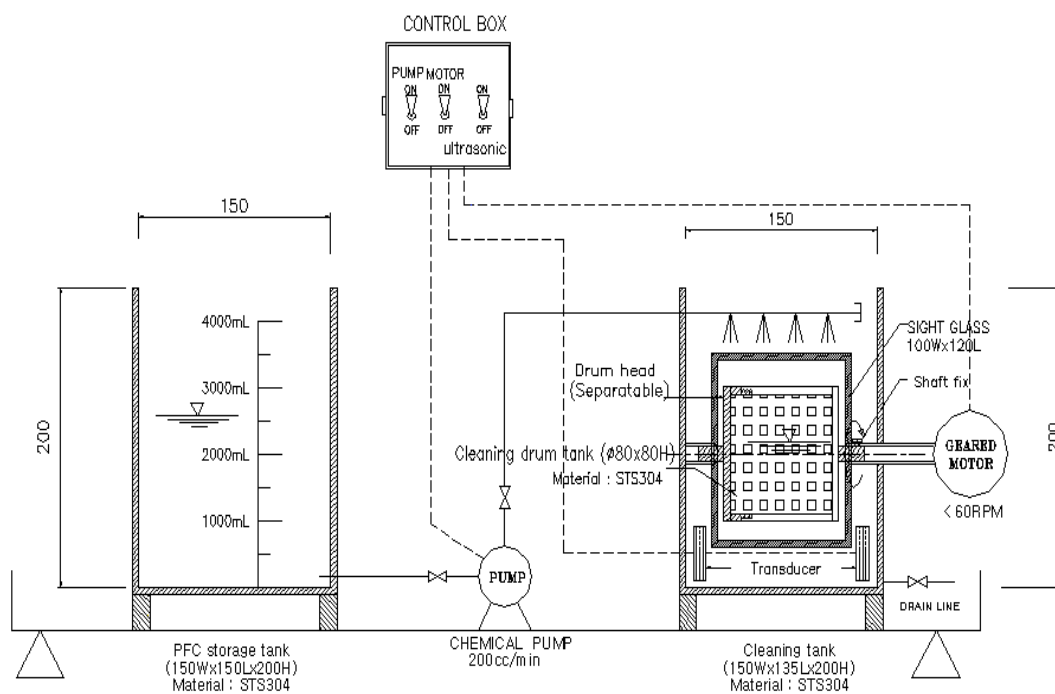


Fig. 1. Schematic diagram of the ultrasonic decontamination equipment

Fig. 3 shows the FT-IR spectra for the PFC and the PFC+ 2.0 vol% surfactant. In the spectrum (b), we can observe the carboxyl group at a wave number of 3300  $\text{cm}^{-1}$ . Surface potential of the metal oxide varies with a change of the solution pH[9]. From the measurement of the zeta potential for the  $\text{Eu}_2\text{O}_3$  in an aqueous solution, we found that the surface potential of the  $\text{Eu}_2\text{O}_3$  powder had a positive value in the decontamination condition in this study. The difference in the decontamination efficiency between the

pure PFC and the PFC + surfactant solution is explained by the interaction between the positively charged  $\text{Eu}_2\text{O}_3$  powder and the carboxylic group of an anionic surfactant in a PFC solution.

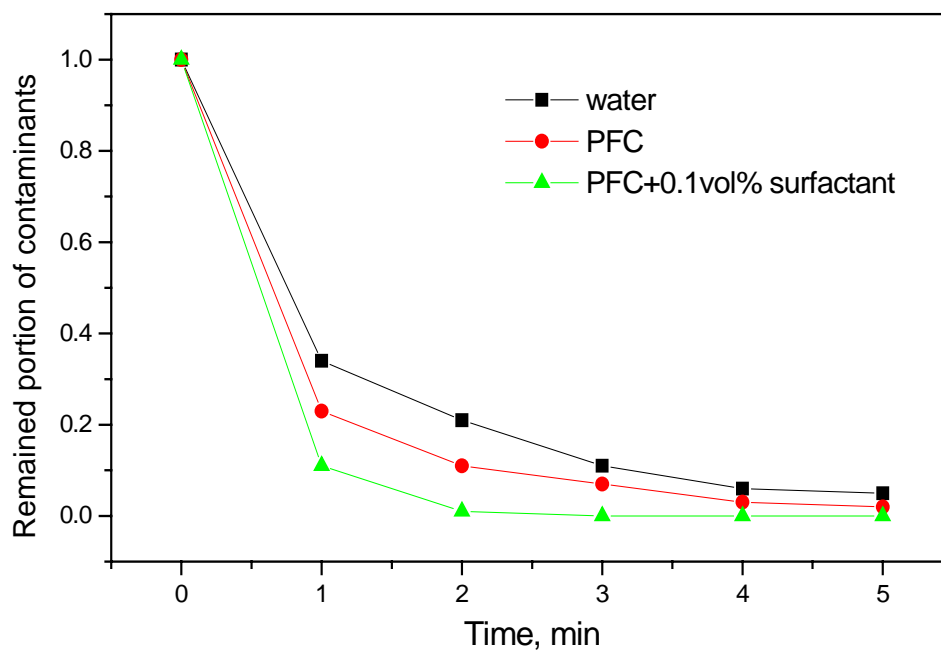


Fig. 2. Remained portion of contaminants according to the application time for 3 kinds of solutions

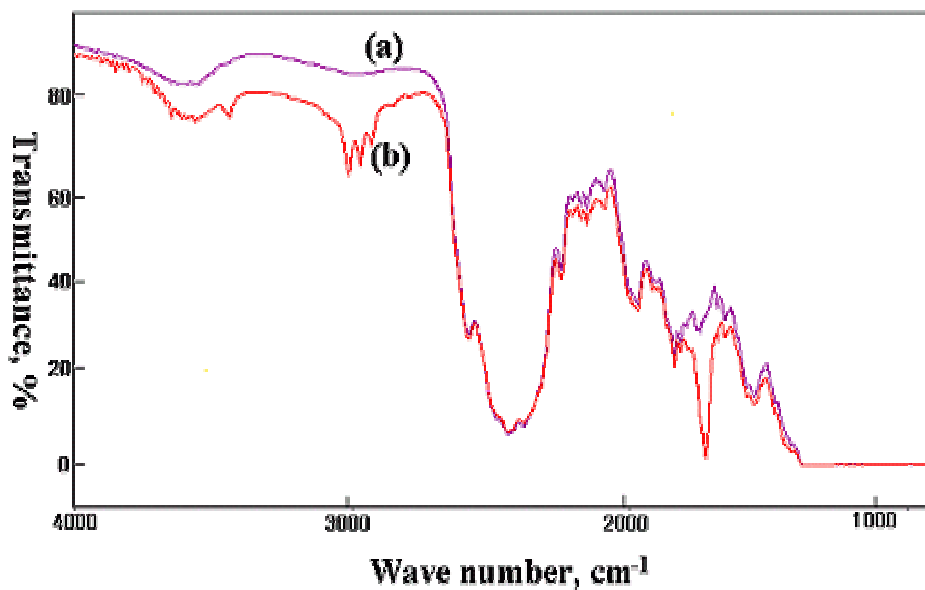


Fig. 3. FT-IR spectra, (a) pure PFC, (b) PFC + 2.0 vol% surfactant

### Decontamination of a Pipe, Welding and a Crevice Specimen

In order to examine the ultrasonic decontamination efficiency for the internal part of a metal specimen, type 304 stainless steel pipe specimens were fabricated. The diameter of a pipe specimen was 2 cm. Fig. 4 shows the photographs of the pipe specimens. As shown in Fig. 4, all the contaminants on the internal parts of the pipe specimens were completely removed. However, a small portion of the contaminants remained on the surface of the pipe specimen with a length of 6 cm after 5 a minute ultrasonic application. This was removed by a rotation of the specimen chamber during the application of an ultrasonic wave.

Several shapes of the metal specimens were simulated and fabricated according to the shapes of the metal specimens in a hot cell. Fig. 5 shows the photographs of the specimens. For all the test specimens, the contaminants on the surfaces were removed satisfactorily. However, a small portion of the contaminants remained on the edge of the crevice specimen after a 5 minute ultrasonic application. This was also removed by a rotation of the specimen chamber during the application of an ultrasonic wave.

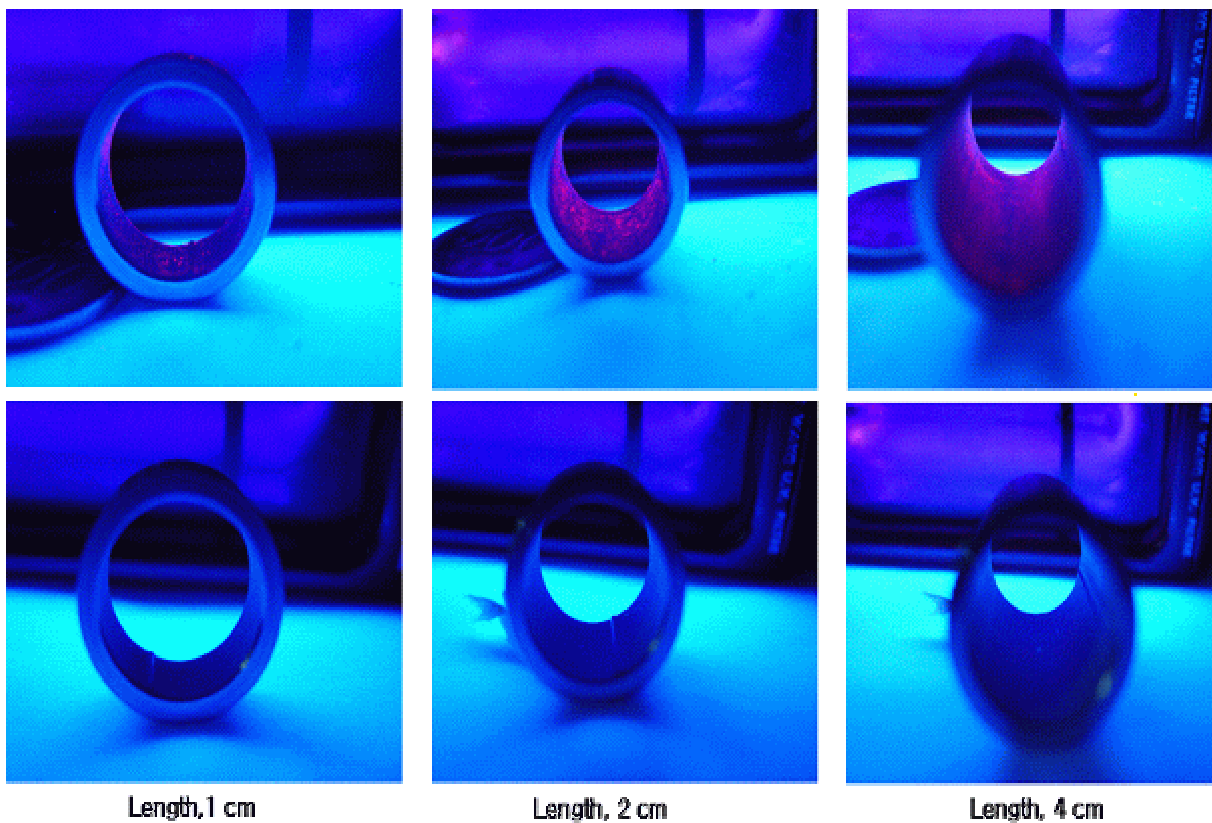


Fig. 4. Photographs of a surrogate pipe a specimen (upper) before a decontamination (lower) after a decontamination FT-IR spectra, (a) pure PFC, (b) PFC + 2.0 vol% surfactant

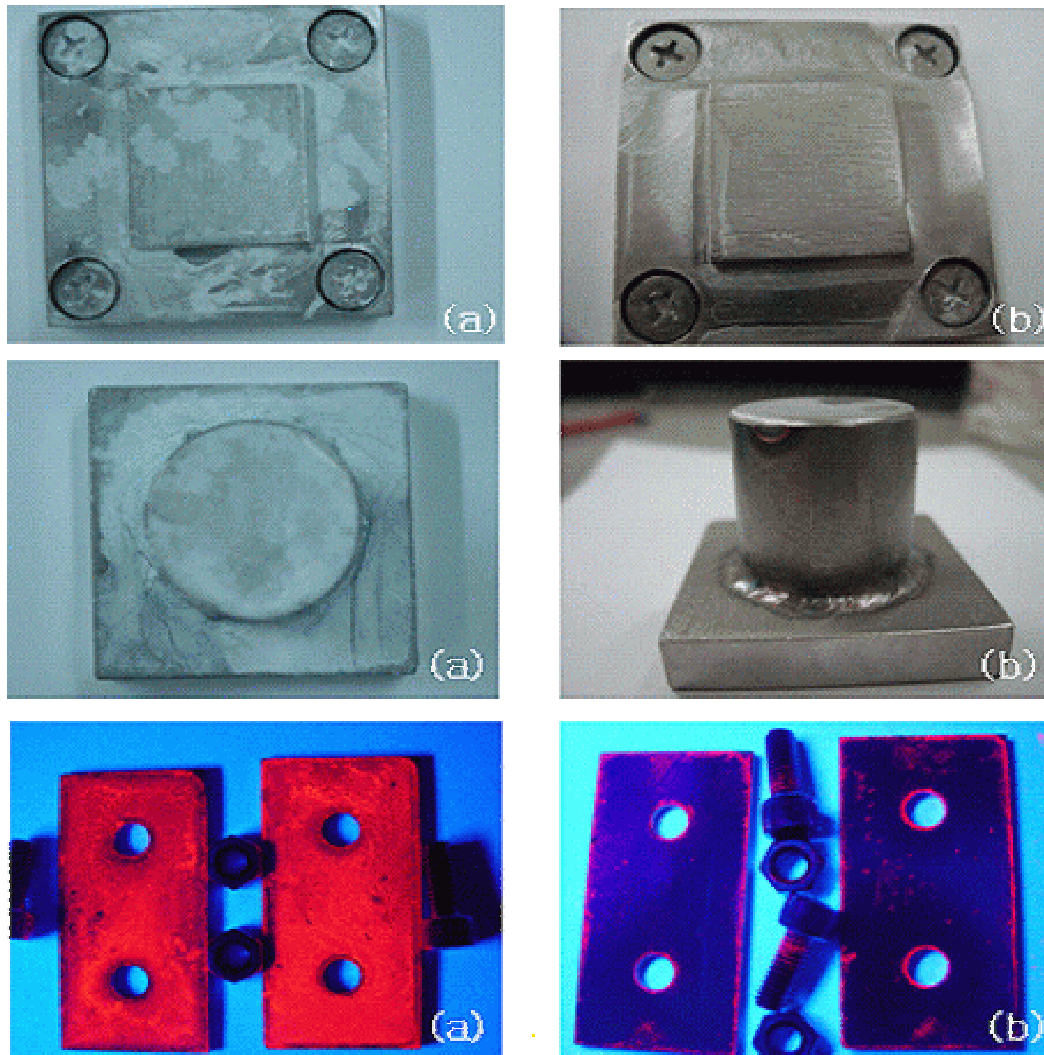


Fig. 5. Photographs of the 3 kinds of surrogate specimens, application time 5 minutes, PFC + surfactant, (a) before a decontamination, (b) after a decontamination

### Recycling of PFC by a Distillation

As a result of multiple applications, the PFC solution becomes to dirty. The contaminants in the solution are removed by a distillation and the solution can be recycled. Fig. 6 shows the plot of a variation of the turbidity of the PFC solution against the distillation cycle. The turbidity of the contaminated solution is significantly reduced by a distillation. After seven distillations, the turbidity of the PFC solution is almost the same as that of the original solution. By a distillation, more than 97.5 % of the PFC was recovered.

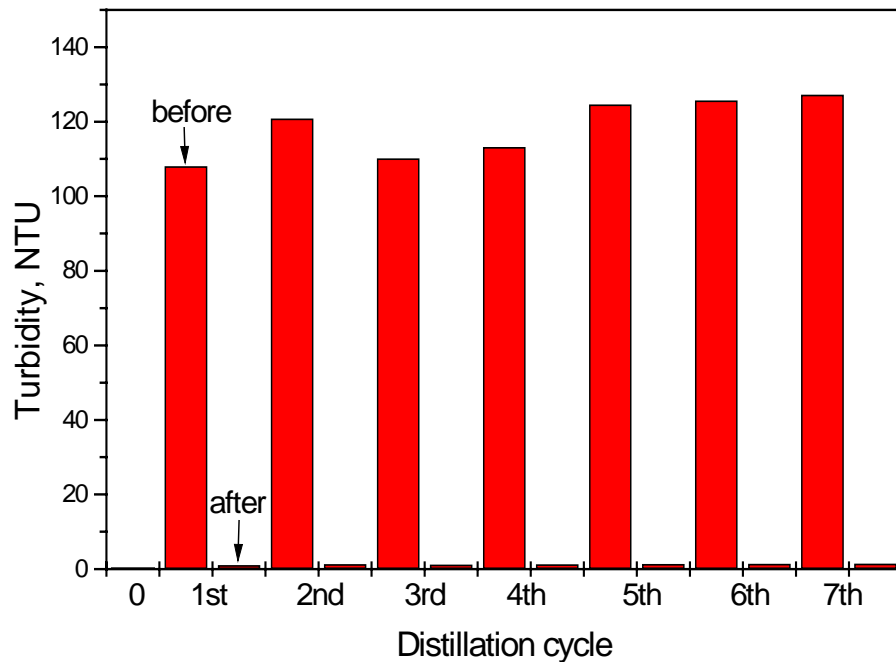


Fig. 6. Variation of the turbidity of the PFC solution against the distillation cycle

## CONCLUSION

A study on a PFC ultrasonic decontamination by using several shapes of metal specimens was performed in a PFC solution. For all the tested specimens, we found that the ultrasonic decontamination was satisfactorily applied. A recycling of the PFC solution by a distillation makes the process more reliable. The decontamination work was performed with a little loss of the main decontamination agents. As the PFC solution is a nonconductive substance and easily separated from the contaminants, the PFC ultrasonic decontamination process is a promising method to decontaminate metal specimens loosely contaminated with radioactive particles. Contamination removal can be enhanced by rotating specimen. Based on the test results, we are developing the PFC spray decontamination process. This process will be used to decontaminate the hot cell and its materials in the Institute.

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## REFERENCES

1. Tsai, W. T., Chen, H. P. and Hsein, W. Y., "A review of uses, environmental hazards and recover/recycle technologies of perfluorocarbons(PFCs) emissions from the semiconductor manufacturing processes", *J. Loss Prevention in the Process Industries*, 2002, 15, 65-75.
2. Hawkinson, T. E. and Korpela, D. B., : Chemical hazards in semiconductor operations, in R. A. Bolmen, *Semiconductor Safety Handbook*, pp 163. Westweed(USA): NOYES, 1998, Chap. 3.
3. Robin, M. L., : Halogenated fire suppression agents, in A. W. Miaiolek and W. Tsang, *Halon replacements: Technology and Science*, pp. 85, Washington, DC: *American Chemical Society*, 1995, Chap. 9.
4. Owens, J. G., "Low GWP alternatives to HFCs and PFCs", *Proceedings of 1999 Taipei International Conference on Atmosphere Protection*, Taipei, Taiwan 1999.
5. Smart, B. E. and Fernandez, R. E., : Fluorinated aliphatic compounds. In J. I. Kroschwitz and M. Howe-Grant, pp. 499, *Kirk-Othmer encyclopedia of chemical technology*, 4<sup>th</sup> ed., 1994, Chpt.11.
6. Straume, A. G., Dietz, R. N., Koffi, E. D., and Nodop, K., "Perfluorocarbon background concentrations in Europe", *Atmospheric Environment*, 1998, 32, 24, 4109-4122.
7. Weston, R. E. Jr., "Possible greenhouse effects of tetrafluoromethane and carbon dioxide emitted from aluminum production", *Atmospheric Environment*, 1996, 30, 16, 2901-2910.
8. USDOE: Separation and extraction of plutonium in mixed waste, Work performed under DOE award number: DE-AC26-01NT 41308, 2002.
9. Hunter, R. J., *Zeta Potential in Colloid Science- Principles and Applications*, Academic Press, Australia, 1981.