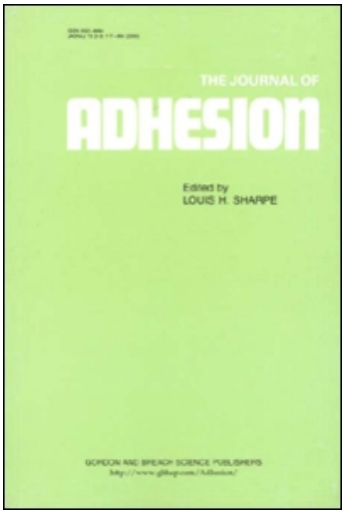

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Enhanced Particle Removal in Fluorinated Liquids – An Application in Nuclear Decontamination*

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An experimental cleaning system has been developed to demonstrate the decontamination of model electronic circuit boards by this cleaning process. The media used in this process are a wash solution of a high molecular weight fluorocarbon surfactant in a perfluorinated carrier liquid which results in enhanced particle removal, followed by a perfluorinated carrier liquid rinse. The perfluorinated liquids of interest, which are recycled in the process, are inert, nonflammable, generally safe to use, and do not present a hazard to the atmospheric ozone layer. The cleaning experiments were performed at the MIT Nuclear Reactor Laboratory. The radioactive particles removed from the circuit boards were captured by 0.22 μm filters with a filtration efficiency of 99.5% per stage. Compatibility tests were performed on these model electronic circuit boards. The results obtained show that neither the process fluids used nor the maximum level of the ultrasonic agitation applied harmed the circuit boards or the circuit components. All the circuit boards used in these tests were still functional after the cleaning experiments. A three log reduction in contamination was obtained in 1 hour. These data indicate that this process, once implemented on a large scale, will offer users in the nuclear industry a practical and cost effective means of decontaminating and recovering a wide variety of tools and instruments.

KEY WORDS: Cleaning system; inert perfluorocarbon liquids; enhanced particle removal; fluorocarbon surfactant; sonification; radioactive iron oxide particles; nuclear decontamination; electronic circuit boards.

1. INTRODUCTION

The application of Entropic Systems Inc.'s enhanced particle removal process to the nondestructive decontamination of nuclear equipment is discussed. None of the decontamination methods now available can be used to decontaminate electronic/electrical equipment without irreversible damage. For this reason ESI received support from the Nuclear Regularity Commission to develop this process further.

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A laboratory scale ultrasonic decontamination system has been developed to demonstrate the application of the enhanced particle removal process[1] to the decontamination of radioactive electronic circuit boards.[2] The process uses inert perfluorinated liquids as the working media; the liquids have zero ozone depletion potential, are inert, nonflammable, and are generally recognized as non-hazardous materials. The parts to be cleaned are first sonicated with a dilute solution of a high molecular weight fluorocarbon surfactant in an inert perfluorinated liquid. The combination of ultrasonic agitation and liquid flow promotes the detachment of the particles from the surface of the part being cleaned, their transfer from the boundary layer into the bulk liquid, and their removal from the cleaning environment, thereby reducing the probability of particle redeposition.[3] After the cleaning step, the parts are rinsed with the pure perfluorinated liquid to remove residual surfactant and dried. The process is operated in a closed flow loop, thereby minimizing the consumption of the process liquids.

2. DECONTAMINATION SYSTEM

A laboratory scale decontamination system has been designed and constructed with a process layout as outlined in Figure 1. There are two separate cleaning loops in this system: loop#1 is a washing loop which contains a surfactant solution in perfluorinated liquid as the working medium, and loop #2 is a rinsing loop which contains pure perfluorinated rinse liquid. As shown in Figure 1, the cleaning liquid is drawn

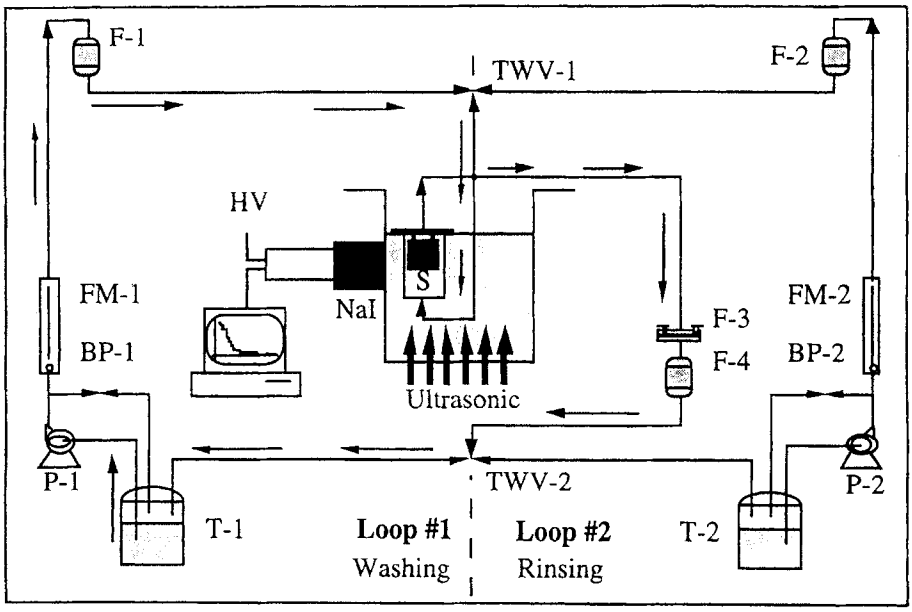

FIGURE 1 Schematic of the decontamination system.

from storage tank (T-1) by pump (P-1) through a closed flow loop #1 consisting of a flow meter (FM-1), a 0.2 μm membrane filter assembly (F-1), a test cell (S), a 0.2 μm membrane particle capture filter (F-3), a redundant 0.2 μm membrane filter assembly (F-4), and finally returns to the storage tank (T-1). The test cell, containing the test part to be cleaned, is placed in a temperature-controlled ultrasonic bath, filled with 2 gallons (7.6 liters) of water as a coupling liquid for ultrasonic propagation. Ultrasonic agitation in the bath is generated by a square wave generator with a power range from 15 watts to 285 watts. The "detached" radioactive particles are removed from the cell by the perfluorinated liquid, and then captured on a disposable 0.2 μm membrane filter (F-3). In this laboratory scale cleaning system, the radioactive parts are placed in a small test cell as shown in Figure 1. This test cell is a thin walled, electropolished, stainless steel cylindrical chamber, 6 cm in diameter and 10 cm deep. The cylinder has an internal volume of 250 ml.

3. PREPARATION OF RADIOACTIVE TEST PARTS

3.1 Model Circuit Boards

Small circuit boards, 3.81 cm square, were designed and built to fit into the test cell as test parts. These boards are functional and are tested before and after each decontamination run. The circuitry of these boards is shown in Figure 2.

3.2 Contamination of Circuit Boards

The experiments were performed with a number of fine natural iron oxide powders. The particle size distribution of the principal powder used in the experiments is shown in

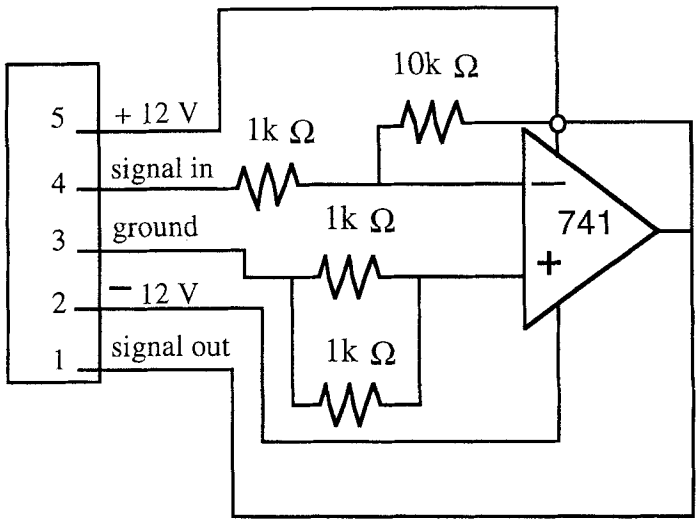

FIGURE 2 Circuitry of model circuit board.

Figure 3. The mean diameter, D_{50}, for this fine powder is 0.75 μm. After irradiation, the sample powder (principal isotope = Fe-59) was suspended in isopropanol. The test circuit boards were contaminated by dipping them in this suspension.

The suspension was first sonicated at 120 Watts for 15 min, to ensure its homogeneity. A circuit board was then dipped into the suspension for 15 min and air dried for 15 min. The activity of the board increased as the number of "dips" increases. Since the activity of the radioactive Fe-59 iron powder is a function of the decay time, the activity levels of the boards were different between experiments. The activity level was ranged from 60,000 to 480,000 disintegrations per minute.

4. CAPTURE OF RADIOACTIVE PARTICLES

4.1 Filtration Efficiency

Before initiating the particle removal studies, it was necessary to demonstrate that any suspended particles could be captured by simple filtration. The purpose of these experiments was to determine suitable filters for capturing the contaminants. The filtration efficiency of a series of 47 mm diameter membrane filters was evaluated. Seven filters with pore size range from 0.05 to 5.0 μm (0.05, 0.1, 0.2, 0.45, 0.65, 1.0 and 5.0 μm) were tested. 100 ml of the isopropanol which contained irradiated iron oxide powder was filtered by each membrane filter. The activity of the Fe-59 isotope was measured before and after each filtration. Filtration efficiency was obtained by comparing these two measurements. Figure 4 shows the filtration efficiency *versus* filter pore size.

The results presented in Figure 4 show that a capture efficiency of 99.95% can be obtained with a 0.2 μm filter. To ensure adequate capture of the contaminants during the actual experiments, two 0.2 μm membrane filters were placed in series. This results in an expected capture efficiency of 99.9975%.

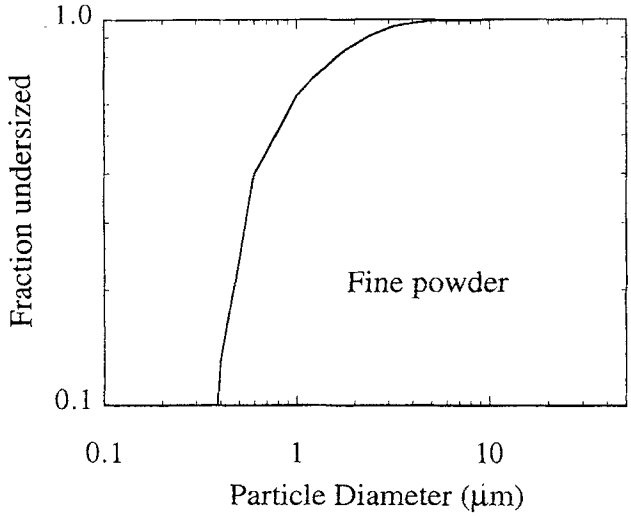

FIGURE 3 Particle size distribution of iron oxide powder.

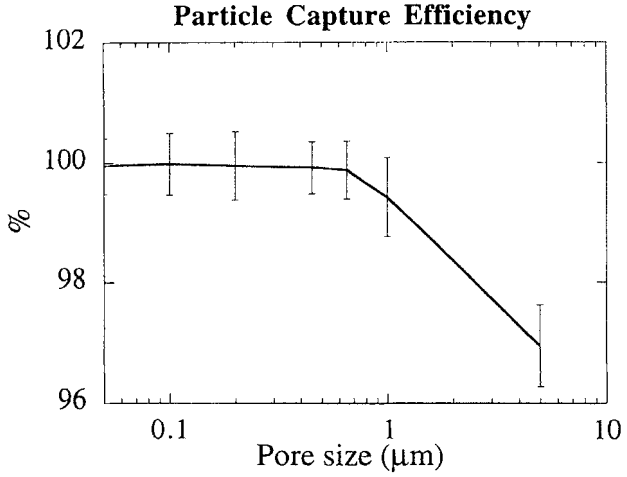

FIGURE 4 Filtration efficiency of iron oxide powder.

4.2 Permeability

Filtration times were also recorded during the filtration experiments. The permeability was observed to increase with increasing pore size as shown in Figure 5. At 0.2 μm, the permeability of the fluid is about 0.0083 ml/cm^2/sec/psi. At a flow rate of 300 ml/min., this results in a pressure drop of about 25 psi (172 KPa) across a single 47 mm diameter filter, which interferred with the ultrasonic agitation. For this reason, a large surface area cartridge filter (1000 cm^2) was used, resulting in negligible pressure drop at experimental flow rates.

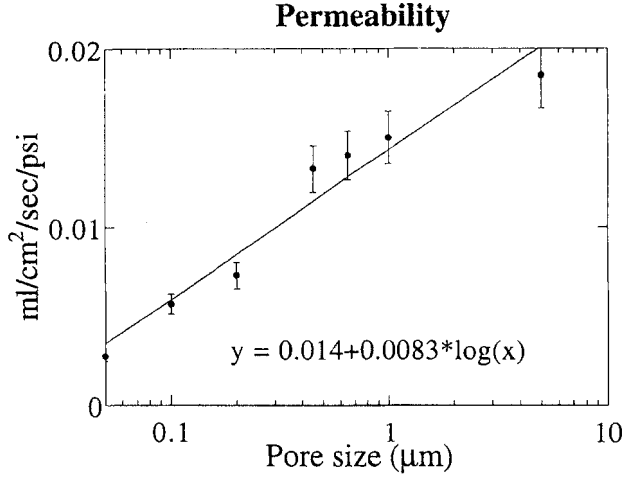

FIGURE 5 Permeability of 3M's PF-5070 per fluorocarbon liquid as a function of pore size with 47 mm Filters.

5. ON-LINE ACTIVITY MONITORING SYSTEM

A radiation detection system consisting of a 3 × 3″ (7.6 × 7.6 cm) NaI scintillation gamma detector, a pre-amplifier, an amplifier, a high voltage power supply, a counter and a computer data acquisition system, was used to monitor the radioactivity level of the test parts during the decontamination process, as shown in Figure 6. The Fe-59 emits gamma rays with energy peaks of 1.09 and 1.29 MeV. At these energy levels, the attenuation due to the process liquid, the thin steel wall or the water bath is insignificant. The extent of removal expressed as a decontamination factor (DF), is calculated by comparing the residual activity of the time, t, with the initial activity at time 0, *i.e.*,

$$\mathrm{DF}(t) = [\text{initial activity } \mathrm{A}(0)]/[\text{final activity } \mathrm{A}(t)]. \quad (1)$$

To reduce errors in calculating DF, the background radioactivity has to be minimized. A two-inch (5.1 cm) thick lead shield was installed around the system surrounding the water bath and the gamma detector. Furthermore, when the final count rate is comparable to the background level (when a high DF is achieved), a long period post-cleaning counting is also required to minimize statistical errors in the net activity.

6. RESULTS

6.1 Characteristics of Particle Removal Rate Using Nuclear Measurements

More than 100 cleaning experiments were performed with different operating conditions – liquid type, flow rate, temperature, particle size, sonication power, etc. Typical

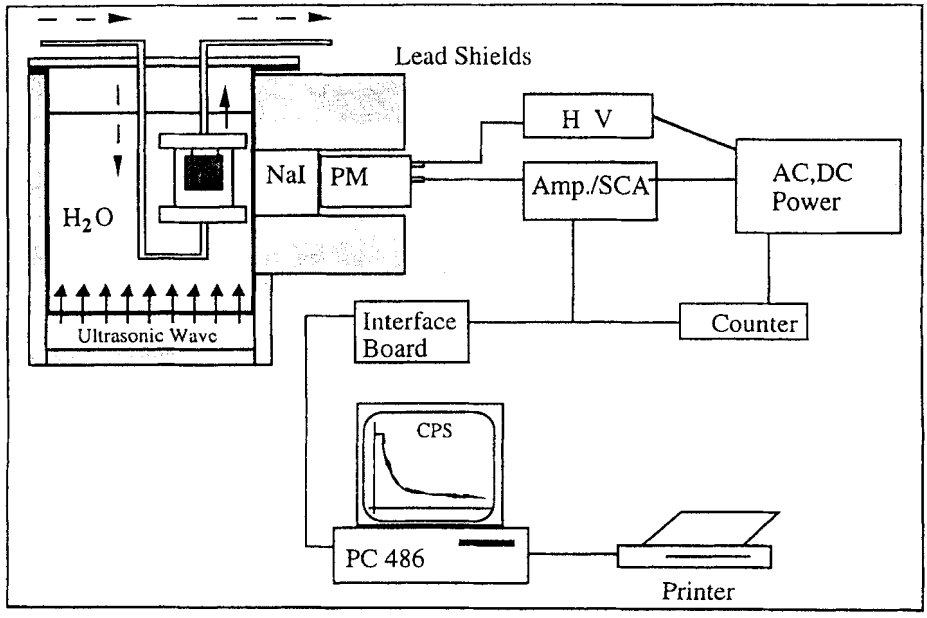

FIGURE 6 An on-line radiation detection system for monitoring the decontamination level.

results are presented in Figure 7. (For details and a complete listing of the experiment performed, see Ref. 4) As can be seen in this figure, a combination of ultrasonic agitation and flow is required to achieve particle removal. As is evident from Figure 7, no significant change in activity is noted during the first 5 min. of the experiment when no ultrasonic power was applied. Once the ultrasonic power was applied (60 W), a very rapid change in activity was then noted, with the rate of removal decreasing exponentially.

Particle removal is also a function of liquid properties. Different process liquids were used in the cleaning experiments. The results show that perfluorinated liquids have better particle removal capability than CFC-113 (Freon-113®). Addition of 0.3 wt.% of a fluorinated surfactant further enhances the cleaning capability of the perfluorinated liquids. A summary of the DF results obtained with different process liquids in 1 hr is presented in Table I. This table presents the best results obtained under the following process conditions: sonication power 60 W, flow rate 75 ml/min. and temperature 110°F.

The removal rate is also a function of flow rate. Initially, the rate of particle removal is higher at a higher flow rate of 150 ml/min than at 75 ml/min. However, after

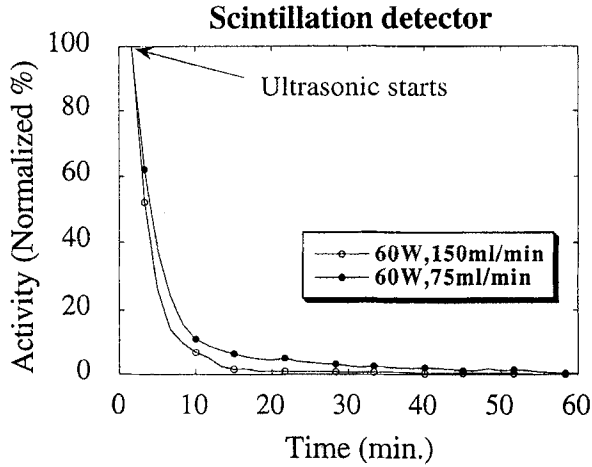

FIGURE 7 Cleaning of radioactive circuit boards with fluorinated surfactant solution at 60 W scintillation measurements.

TABLE I
Comparison of DF for Different Process Liquids. Best Results Obtained at 60 Watts Sonication Power, 75 ml/min., 110°F (43°C) and Cleaning Period of 1 Hour

Process Liquid	DF	Error
CFC-113	64	5%
PFC-Vertrel® (Dupont)	93	8%
PFC-3M's PF-5070	250	15%
PFC-3M's PF-5070 + (0.3 w/o surfactant)	1213	30.5%

Surfactant: Dupont's Krytox® 157 FSL Lot-12; PFC: Perfluorocarbon.

approximately 60 min, flow rate has little further effect on the continuing rate of decontamination. Initially, particle removal appears to be a function of the rate of mass transfer of the particles into the flowing liquid stream. The larger the flow rate, the higher the rate of removal. At the end, when most of the particles are removed, removal is no longer a function of mass transfer, but is a probabilistic surface event independent of fluid flow. At 60 min., at both flow rates, value of DF > 1200 were obtained with the surfactant solutions used. The particle removal rate at the end of the 1 hour experiment is not zero. The DF increases from 1200 after 1 hr to 1800 after 2 hours.

6.2 Monitoring of Decontamination by Optical Measurements

The rate of particle removal from circuit boards contaminated with nonradioactive iron oxide powders was measured by the optical counting method used by ESI to evaluate the cleaning of precision parts using fluorocarbon solutions. Test circuit boards were contaminated with nonradioactive iron oxide powder in exactly the same manner previously used to contaminate test boards with the same iron oxide powder that had been rendered radioactive by irradiation at the MIT reactor.

The contaminated boards were cleaned in ESI's cleaning system setup in its clean room in Woburn, MA. This system is a duplicate of the decontamination system setup at the MIT Nuclear Reactor Lab, except for the sensors. The system at MIT is equipped with a NaI scintillation detector to monitor the rate of removal of radioactivity from the board. The system in Woburn is equipped with optical particle sensors which measure the concentration of particles in the cell's effluent stream. In these experiments, both light blocking and light scattering sensors were used with a detection range from 0.5 to 100 μm. These sensors were placed in series to measure the particle population in the effluent stream as a function of time. Particle removal (counts/ml) was measured during the cleaning experiments at sonication power levels of 60 W and 240 W and at a flow rate of 75 ml/min. Particle concentration signals of the effluent measured by the optical sensors at the exit of the test cell are presented in Figure 8 and 9.

The optical measurements provide interesting corollary information to the nuclear measurements. Both sets of experiments indicate that both fluid flow and ultrasonic agitation are required for particle removal. The optical measurements differ from the nuclear data; in fact, they provide information on the effect of particle size on the rate of decontamination. The results shown in Figure 8 and 9 clearly indicate that the small particles are the last ones to be detached. This is to be expected since the ratio of the sum of the shear energy (due to ultrasonic agitation and flow) and of thermal agitation to that of van der waals' attraction decreases with decreasing particle size.[5,6] For long cleaning periods (> 1 hr.), the values of the DF obtained are controlled by the number and the rate of removal of the smallest particles present.

The combination of low acoustic velocity and high density exhibited by perfluorocarbons results in an effective coupling between these liquids and the applied acoustic field.[7] In this application, this results in effective particle removal. The addition of a soluble surfactant further enhances the ability of particle removal because of the formation of a solvated film on the surface of the particle and the solid substrate being cleaned.

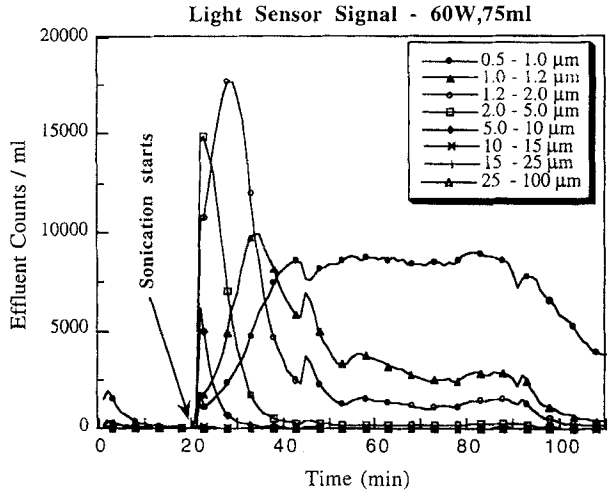

FIGURE 8 Cleaning of nonradioactive circuit boards with fluorinated surfactant solution at 60 W-optical measurements.

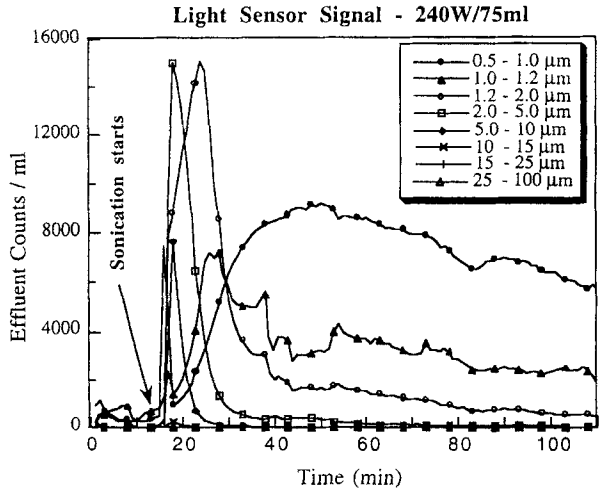

FIGURE 9 Cleaning of nonradioactive circuite boards with fluorinated surfactant solution at 240 W-optical measurements.

The particle concentration signals obtained by optical sensors were converted to mass removal data. These results are presented in Figure 10. In order to compare the results between optical and scintillation measurements, the final DF of the optical measurement was normalized to the scintillation result. By comparing the decontamination curves from Figure 7 and Figure 10, a strong correlation is noticed between the scintillation and optical measurements.

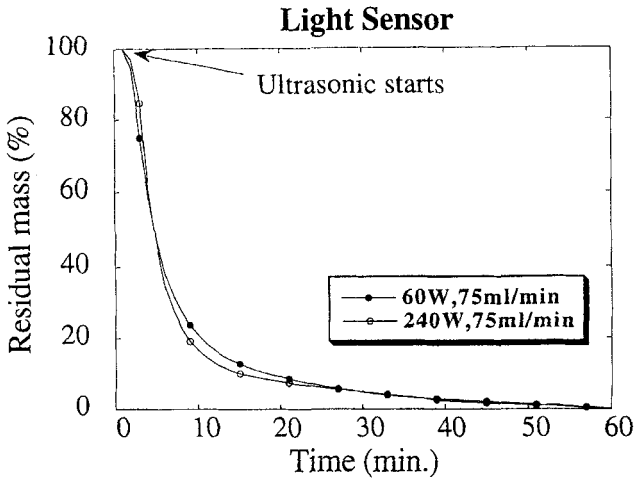

FIGURE 10 Normalized cleaning results of nonradioactive circuite boards with florinated surfactant solution.

7. CONCLUSIONS

Based on the experimental data obtained, including the data in Ref. 4, it is concluded that:

(i) The particle removal ability of perfluorinated liquids is better than that of CFC-113 (Freon-113®).
(ii) Enhanced removal is obtained with the addition of 0.3 wt.% of a perfluorinated surfactant.
(iii) High Decontamination Factors (DF > 1200) are obtained in 1 hour.
(iv) The removal rate after 1 hour is non-zero.
(v) This cleaning process does not harm the electronic circuit boards.

Acknowledgement

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