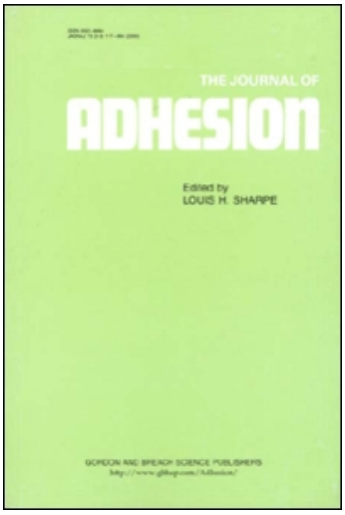

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Ahmed A. BUsnaina[a]; Fen Dai[a]
[a] Microcontamination Research Laboratory, Clarkson University, Potsdam, NY, USA

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The Removal of Submicron Particles in Liquid-based Cleaning*

AHMED A. BUSNAINA** and FEN DAI

Microcontamination Research Laboratory,
Clarkson University, Potsdam, NY 13699-5725, USA

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Megasonic cleaning is one of the most widely used wet-cleaning processes in the semiconductor, hard disk and flat panel display industries. Presented results involve different and new techniques for introducing the high frequency ultrasonic energy in the cleaning bath. The effects of power, temperature and time on the removal efficiency of Si_3N_4 particles in the size range from 0.1 μm to 1.0 μm from silicon wafers are presented. Results show that removal efficiencies near 100% for silicon nitride particles using deionized water could be achieved under the right conditions. The megasonic input power has a greater effect on the removal efficiency than does temperature.

Keywords: Megasonic cleaning; surface cleaning; particle removal; acoustic streaming; removal mechanisms; submicron particles

1. INTRODUCTION

Liquid-based cleaning is extensively used for removal of particulate contamination in the semiconductor and other industries affected by contamination. One of the widely used wet-cleaning processes is the high frequency ultrasonic cleaning (known as megasonic cleaning) [1]. Ultrasonics refers to the frequencies above what is audible to humans (approximately 18 kHz and higher). The megasonics term is used in

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**Corresponding author.

the industry to refer to ultrasonic frequencies near 1 MHZ. Ultrasonic cleaning has been used for more than three decades. Rosenberg [2] utilized ultrasonic cleaning for the removal of particulate contaminants and films. Olaf [3] showed that ultrasonic cleaning using high and low frequency (15 kHz to 2.5 MHZ) can be used to clean glass substrates. More recently, McQueen [4, 5] identified the effect of the acoustic boundary layer and its role in the removal of small particles at high frequency. However, megasonic cleaning techniques used today in the industry were first presented by RCA scientists [1, 6, 7]. Kashkoush, Busnaina *et al.* [8–11] studied ultrasonic and megasonic particle removal, focusing on the effects of acoustic streaming. They showed that the removal percentage increased with power. Their results also indicated different removal efficiencies for polystyrene latex (PSL), silica (SiO_2) and silicon nitride (Si_3N_4) particles. Megasonic cleaning using SC1 and SC2 (Standard Clean 1 and 2) chemistry has been shown to be very effective by Syverson *et al.* [12]. They also showed that the removal efficiency increased with power up to a 150 W (maximum power available). Wang *et al.* [13] also showed that power had the greatest influence on the removal efficiency up to a maximum power available (150 W). These results are consistent with what Kashkoush, Busnaina and Gale [12, 13] observed. However, Gale and Busnaina [14–17], using higher power megasonics up to 800 W, showed that the highest removal efficiency occurs at an optimum power (500–600 W), above which it decreases slightly. They also showed that the megasonic input power has the greatest influence on particle removal efficiency as compared with solution temperature, both in water and in SC1 solution. They also showed that SC1 removes particles more efficiently than DI water, particularly at lower megasonic powers. But they also showed that it was still possible to achieve 100% removal in DI water under the proper conditions.

This paper introduces the recent results that involve different and new techniques for introducing the high frequency ultrasonic energy in the cleaning bath. The results show that removal efficiencies near 100% for silicon nitride particles using DI water could be achieved under the right conditions. The megasonic input power has a greater effect on the removal efficiency than does temperature. This is consistent with previous results generated using different megasonic equipment [14–17].

1.1. Ultrasonic Cleaning

Ultrasonic cleaning tanks typically have piezoelectric transducers at the bottom vibrating at a prescribed frequency. Piezoelectric substance will mechanically deform when electrically polarized. This flexing or vibrating of the transducer produces a pressure (sound) wave that propagates through the tank. The speed of the sound wave in water at ambient temperature is 1481 m/s. The sound speed is a strong function of temperature, decreasing as the latter increases in most liquids [18]. In water, however, it increases with temperature up to 73°C, after which it decreases. The given temperature is for pure water with no air or vapor bubbles. A typical temperature at which the maximum speed of sound occurs will be between 38–49°C [9, 10].

In the tank, the wave loses energy as it travels away from the transducer. This is called wave attenuation. This is mainly caused by scattering, diffraction, and absorption [18]. Wave scattering takes place when the wave encounters a significant number of particles or bubbles which reflect the sound beam. Diffraction occurs in the region beyond the distance $D^2/4\lambda$ from a circular source (where D is the source diameter and λ is the wave length). Attenuation near the transducer is primarily caused by absorption. Absorption results mainly from interaction with the fluid medium which includes viscous losses, heat conduction losses, and losses associated with molecular energy exchange [19].

1.2. Acoustic Streaming

There are three types of acoustic streaming that occur in a sound field. Rayleigh streaming occurs outside the acoustic boundary layer due to a standing wave in a tube or channel. The vortices in Rayleigh streaming are of the scale of the acoustic wavelength. Eckart streaming occurs in a free, nonuniform sound field and is characterized by streaming patterns on the scale of the volume of the flow field (larger than the acoustic wavelength). Boundary layer streaming occurring due to interactions with obstacles in an acoustic flow is referred to as Schlichting streaming [20].

Streaming velocity increases with the increase of frequency and power, and decreases with the increase of kinematic viscosity [21]. For

example, at 850 kHz the streaming velocity (Eckart-type streaming) at high intensity (7.5 W/cm^2) is about 4.8 m/s. At commonly used lower intensities (2.5 W/cm^2), the streaming velocity is also lower at 1.5 m/s. The streaming velocity decreases at lower frequencies. At 360 kHz, the streaming velocity at high density (7.5 W/cm^2) is about 0.78 m/s. For lower intensity (2.5 W/cm^2), the streaming velocity decreases to 0.25 m/s.

One of the most important aspects of ultrasonic and megasonic cleaning is the acoustic boundary layer which is very small as compared with a typical hydrodynamic boundary layer at the same velocity. This exposes small particles on the surface to much larger velocities and increases the particle removal efficiency. The acoustic boundary layer thickness is a function of the frequency ω (in rad/s, $\omega = 2\pi f$) and the bulk viscocity ν of the fluid and is defined as $\delta_{ac} = (2\nu/\omega)^{1/2}$ [22]. For example, the acoustic boundary layer thickness in water at 40 kHz is about 2.82 μm and about 0.59 μm at 900 kHz. Experimental measurements of acoustic boundary layer thickness in air at low frequencies from 1–4 kHz using a hot-wire anemometer had been reported [23]. The boundary layer thickness under ultrasound was found to be about two orders of magnitude less than the hydrodynamic boundary layer thickness for a similar flow with the same velocity. For example, the hydrodynamic boundary layer ($\delta = 0.16\,[\nu/U\,x]^{1/7}$ where U is the fluid velocity and x is the distance from the leading edge of the wafer) for a flow with a velocity of 4 m/s (maximum streaming velocity for the considered equipment) is about 1500 microns at the center of the wafer (considering a turbulent boundary layer). The inverse square-root dependence of the acoustic boundary layer thickness on frequency has also been verified [23].

Streaming that occurs near bubbles in the field is called micro-streaming. The bubble surface vibrates as a result of negative and positive pressure in the sound waves [24]. This generates vortices and currents that contribute to the removal of small particles from surfaces.

1.3. Particle Removal

Particle removal in megasonic cleaning relies on the reduction of the boundary thickness on the substrates and three types of acoustic

streaming in the tank; Eckart streaming (visible upward flow motion in the tank), Schlichting (boundary layer) streaming and microstreaming. Acoustic streaming has been shown to be the particle removal mechanism in high frequency ultrasonics [8–17]. Other particle removal mechanisms that take place are cavitation and radiation force. Cavitation is considered an effective particle removal mechanism for low frequency ultrasonic cleaning (20–100 kHz). Radiation pressure force has been shown to be significant only at very high intensity sound fields. The resulting force is responsible for the observed levitation and "dancing" of bubbles in the ultrasonic tanks. For very high intensity of 10 W/cm^2, the radiation pressure force on a 1.0 μm particle is 5.24×10^{-11} N. The van der Waals force of adhesion for a silicon particle of this size on a silicon substrate is on the order of 10^{-8} N. For small particles the radiation force is of a very small magnitude relative to the adhesion force [17]. Using the radiation pressure force is only practical at very high intensities. Brereton *et al.* have moved micron-sized particles using a very intensely focused beam ($\langle E\rangle\approx390$ kN/m^2) at frequencies above 1 GHz [25]. Another type of mechanism is identified by Olson [26] and Hasheminejad [27] is the so-called resonance cleaning where resonant response of the particle to a normally incident wave is induced by sweeping through natural frequencies of attached particles at relatively low intensities. However, for submicron particles this requires ultrasonic cleaning in the range of 0.1–1.0 GHz.

2. EXPERIMENT

All the experiments in this study were performed in the Class 10 cleanroom of the Microcontamination Research Laboratory at Clarkson University. The equipment used consists of a quartz megasonic tank (made by PCT, Inc.), which has a maximum input power of 640 Watts (intensity of 7.75 Watts/cm^2) and a frequency 750 kHz. There are two arrays of five transducers each located on the bottom of the tank. The transducers are multiplexed. The first transducer in each array is activated for one second, then the second in each array is activated and so on. Therefore, only two transducers are

active at any given time to avoid overheating. The tank volume is 15 liters. The recirculation (*via* overflow) was kept to about 5 liter/min. A laser surface scanner (made by Particle Measuring Systems, Inc.) was used to scan and determine surface particle counts in the experiment. This instrument uses laser light scattering to detect, locate and size the particles. The scanner has a minimum resolution of 0.1 microns and a size range of 0.1 μm to 10 μm. The scanner provides particle count, location, size distribution and surface roughness.

A spin rinser/dryer, by SemiTool Inc. (STI), was used to dry the wafers in this study. The number of particles added by the rinser/dryer was measured before every experiment and was found to be negligible. The 125 mm silicon wafers used in this study were new, unetched, polished, and *p*-type ⟨100⟩ (single crystal). The resistivity is between 11~18 Ω-cm, and the thickness is between 560 ~ 650 μm. The particles used were silicon nitride (Si_3N_4, supplied by Alfa Aesar, Inc.). The particle size was mixed, from 0.1 ~ 3.0 μm, but most of the particles were in 0.1 ~ 0.3 μm range. A nebulizer was used to deposit particles on the wafer surface. The particles were suspended in isopropanol alcohol (IPA). Particles were deposited on the wafers using a nebulizer with an air filter attached. A point-of-use filtration utilizing a 0.02 μm Millipore filter was used for the DI water.

2.1. Experimental Procedure

(1) The clean silicon wafers were pre-scanned. If the number of particles on the wafer was more than 80, a pre-cleaning process was used to lower the particle count.
(2) The Si_3N_4 particle suspension was prepared using IPA. Particles were deposited onto the clean wafer surfaces using a nebulizer. The wafers were scanned to get the particle counts before cleaning, N_{before}. The average number of particles deposited per wafer surface was kept under 1000.
(3) The wafers were then immersed and cleaned in the megasonic tank using the desired power, temperature and time.
(4) After cleaning, the wafers were dried using the STI rinser/dryer. The wafers were then scanned using the surface scanner and the post particle count, N_{after}, was obtained.

(5) The particle removal efficiency was then calculated using the following relation.

$$\eta\% = \frac{N_{\text{before}} - N_{\text{after}}}{N_{\text{before}}} \times 100$$

2.2. Design of Experiments

A statistical design of the experiment, JMP software by SAS, was used in this study. The response surface design is the most popular method to search for the optimum on a curved surface. A uniform precision central composite rotatable design was chosen for this study. The particle removal efficiency as a function of power, temperature and time was studied. A power range from the minimum 0 to the maximum 640 Watts (an intensity of 7.75 Watts/cm^2) was used. The temperature range was from room temperature, 19°C, to 43°C. The process cleaning time was from 10–22 minutes. The design of experiments matrix is shown in Table I along with the measured removal efficiency.

TABLE I Design of experiments matrix

	Temperature	*Power*	*Time*	*Removal Efficiency*
1	24	250	13	0.96
2	24	250	20	0.79
3	24	541	13	0.98
4	24	541	20	0.84
5	38	250	13	0.85
6	38	250	20	0.92
7	38	541	13	0.99
8	38	541	20	0.95
9	19.2	395.5	16.5	0.96
10	42.8	395.5	16.5	0.88
11	31	150.8	16.5	0.99
12	31	640.2	16.5	0.94
13	31	395.5	10.6	0.96
14	31	395.5	22.4	0.95
15	31	395.5	16.5	0.96
16	31	395.5	16.5	0.99
17	31	395.5	16.5	0.96
18	31	395.5	16.5	0.95
19	31	395.5	16.5	0.98
20	31	395.5	16.5	0.97
21	31	0	16.5	0.7

3. RESULTS AND DISCUSSION

Figures 1–2 show that, at a low power, the particle removal efficiency is very low (about 70%) over the whole temperature range. As the power increases, the removal efficiency increases gradually. The contour plots of the power and temperature show that the maximum removal (higher than 97%) is reached when the power is between 400 Watts and 500 Watts and the temperature between 28 and 35°C. As the power increases in excess of 600 Watts, the removal efficiency begins to decrease gradually. Similarly, as the temperature exceeds 37°C, the removal efficiency decreases. The plots indicate that at about 400 Watts the removal efficiency reaches 99% in a wide range of temperature, 27–34°C. Figure 2 shows that after 22 minutes of cleaning, the particle removal efficiency decreases.

The contour plots of time and temperature, Figures 3 (at a power of 250 W) and 4 (at a power of 395 W), indicate that at about 400 Watts the removal efficiency reaches 99% in a wide range of temperature, 27–35°C, and time, 11 min ~ 16 min. Figures 5 and 6 show the

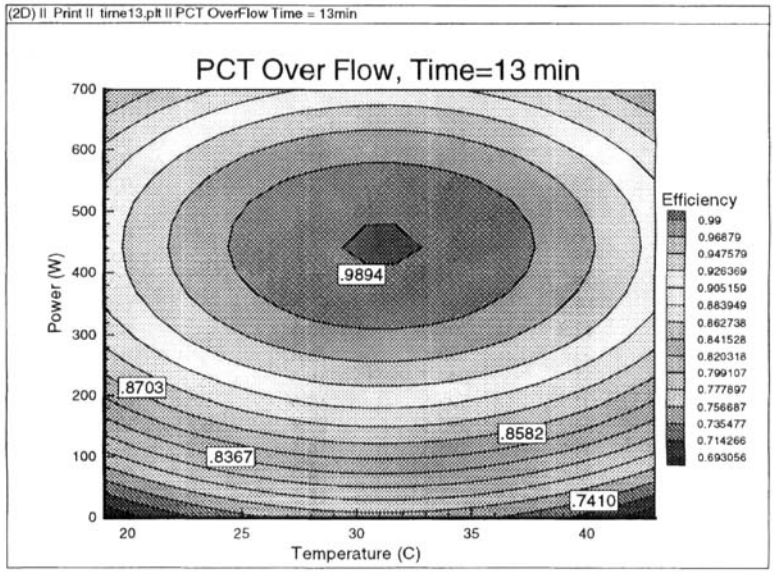

FIGURE 1 Removal efficiency of silicon nitride particles after 13 minutes.

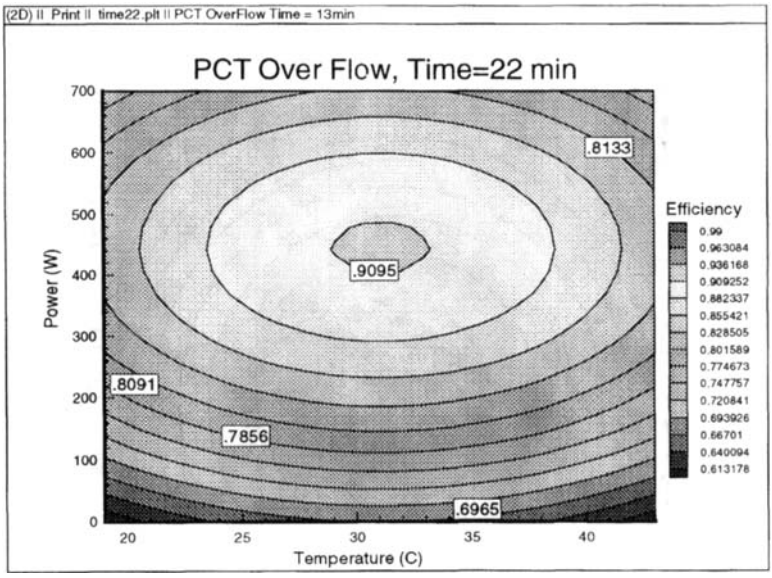

FIGURE 2 Removal efficiency of silicon nitride particles after 22 minutes.

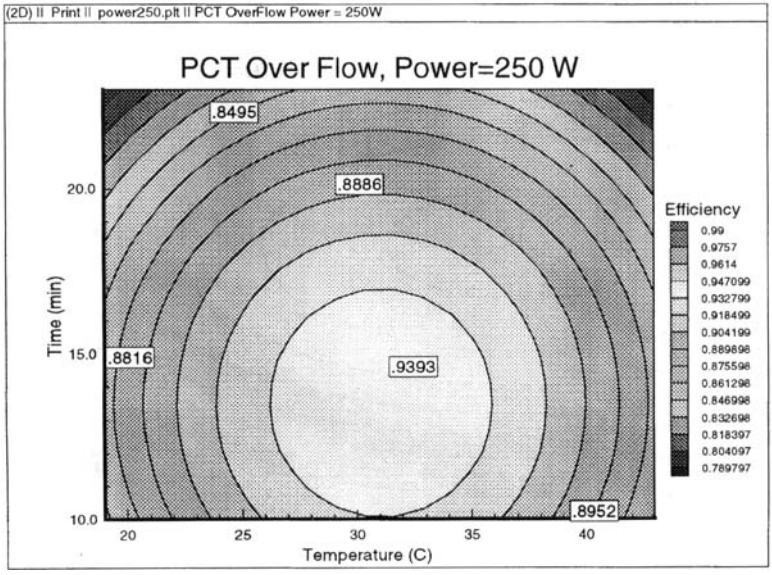

FIGURE 3 Removal efficiency of silicon nitride particles at an input power of 250 W.

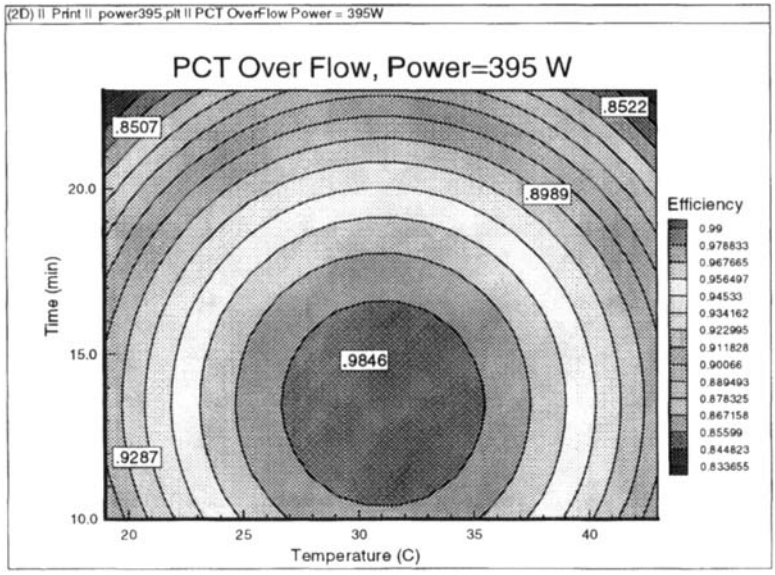

FIGURE 4 Removal efficiency of silicon nitride particles at an input power of 395 W.

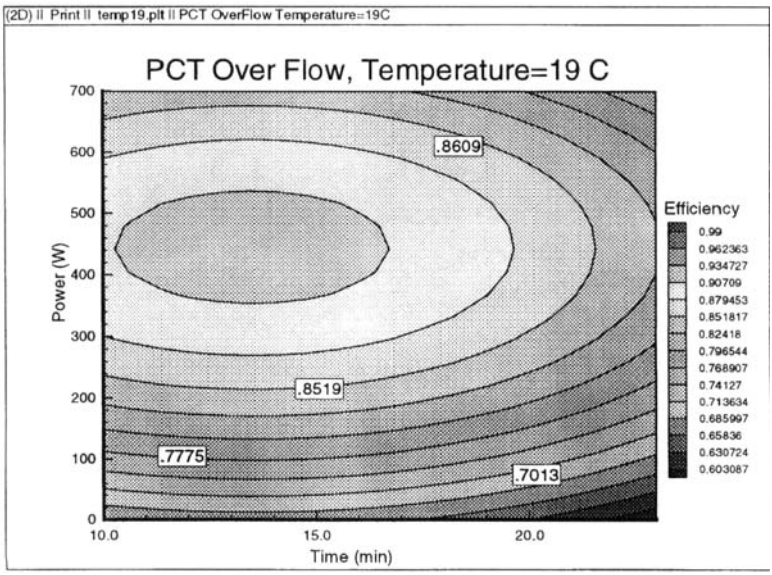

FIGURE 5 Removal efficiency of silicon nitride particles at 19°C.

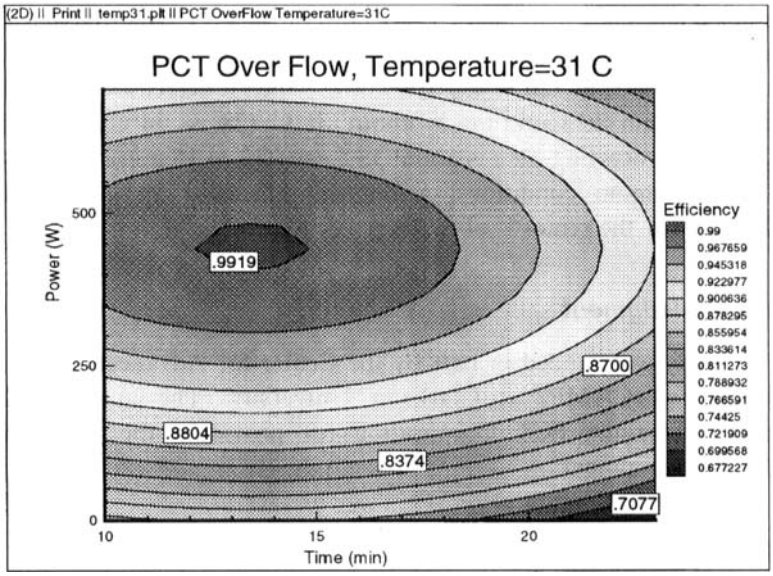

FIGURE 6 Removal efficiency of silicon nitride particles at 31°C.

contour plots of time and power with fixed temperatures (19°C and 31°C, respectively). The figures show that the power has the greatest effect on the particle removal efficiency. When the temperature is at 31°C, more than 97% removal efficiency can be achieved over a wide range of power, 350 W ~ 550 W, and time, 10 min ~ 17 min. The plot also shows that a 99% removal efficiency can be obtained over a power range of 420 Watts–480 Watts and a time range of 13–15 minutes.

4. CONCLUSIONS

These results show the effects of power, temperature and time on the removal efficiency of Si_3N_4 particles in the size range from 0.1 μm to 1.0 μm. The results show that power has the greatest effect on the particle removal efficiency. This agrees with a previous study (using different equipment by Submicron Systems and continuously powered transducers) which showed that power has a greater effect on the

removal efficiency than temperature [6]. For the PCT tank, the particle removal efficiency increases with power until about 450 Watts (optimum value at 420 $\sim$ 480 W), above which it decreases gradually. The optimum temperature is about 31°C (28 $\sim$ 34°C), and the optimum processing time is about 13.5 min (12 $\sim$ 15 min). Using the above optimum conditions, a removal efficiency 99% has been achieved for the removal of Si_3N_4 particles.

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