Coupled high and low-frequency ultrasound remediation of PFAS-contaminated soils

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ABSTRACT

Solids such as soils and sediments contaminated with per- and polyfluorinated alkyl substances (PFAS) from exposure to impacted media, e.g., landfill leachate or biosolids, direct contaminated discharge, and contaminant transport from atmospheric deposition, have caused significant environmental pollution. Such solids can act as secondary sources of PFAS for groundwater and surface water contamination. There are currently no proven technologies that can degrade PFAS in soil and sediments in a cost-effective, environmentally-friendly, and energy-efficient manner. This study examines the use of coupled high and low-frequency ultrasound in desorbing and degrading PFAS in soil, thereby achieving concurrent treatment and destruction of PFAS in soil. Two common PFAS, namely perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS), were used to evaluate treatment performance in soils with both low and high organic matter contents. The test results showed that the ultrasound treatment could significantly reduce PFAS concentrations in artificially contaminated soil; however, no significant degradation was achieved. Ultrasound treatment did improve desorption of PFAS from solid particles, particularly from the highly absorbent organic soil; 68.8 ± 1.8% of PFOA and 45.4 ± 4.1% of PFOS were leached from the soil after ultrasound treatment compared to only 28 ± 0.2% of PFOA and 1 ± 3.1% of PFOS after desorption in water. This work shows that sonication treatment is an effective technology for the removal of PFAS from solids, however, the presence of solids in the solid-liquid slurry can negatively impact ultrasonic cavitation, inhibiting the sonolytic degradation of desorbed PFAS.

1. Introduction

Per- and Polyfluoroalkyl substances (PFAS) are a large family of anthropogenic, fluorinated organic compounds whose widespread use and recalcitrant nature have led to their frequent detection in the environment as well as in the human body, causing growing concern over their impact on human and environmental health [1–3]. The carbon-fluorine C-F bond is the shortest and strongest single covalent bond and is responsible for the thermal and chemical stability of PFAS [4]. This property, in conjunction with the ability to lower water surface tension, makes PFAS ideal for a wide range of industrial and commercial applications such as firefighting foams, non-stick coatings, textiles, etc. [1]. These same properties also render PFAS bio-accumulative, toxic, and ubiquitous in the environment [4]. Thus, the scientific community is rapidly recognizing the presence and impact of PFAS in the environment and looking for innovative destruction technologies.

Sources of PFAS in solids include direct discharge from manufacturing industries, areas utilizing aqueous film forming foams (AFFF), landfill leachate, and biosolids, as well as contaminant transport from atmospheric deposition [5–12]. PFAS are transported from these sources to surrounding soils and waters through leaching and run-off [13–17]. Anderson et al. (2016) evaluated PFAS concentrations in soils and other media for hundreds of samples collected from 40 sites across ten military installations in the USA where AFFF was used. The results demonstrate the widespread presence of several PFAS tested [5,18]. Four PFAS, perfluorooctane sulfonic acid (PFOS), perfluorohexane sulfonic acid (PFHxS), perfluorooctanoic acid (PFOA), and perfluorohexanoic acid (PFHxA), had high concentrations with PFOS being the predominant PFAS detected in soils having a maximum concentration around 373,000 µg/kg [19,20]. More information on the distribution of PFAS in soil can be Brussau et al. 2020, Rankin et al. 2016, and Anderson et al. 2016 [5,18,21].

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In the most commonly detected PFAS in the environment, the carbon–fluorine tail is both hydrophobic and lipophobic, while the non-fluorinated functional head group is polar and hydrophilic. These competing tendencies of the head and tail lead to a wide distribution of PFAS in the environment [22–24]. Two mechanisms govern the sorption of PFAS onto solids such as soils and sediments: (1) the electrostatic interaction of the PFAS polar head group with charged clay and organic matter surfaces and (2) the hydrophobic interaction of the PFAS fluorinated carbon tail with soil organic matter [7,22,24,25]. Equilibrium processes controlling these two mechanisms determine the PFAS partition and distribution in soil environments.

The sorption of PFAS in soils has been shown to increase with an increase in the chain length of PFAS, and also with an increase in the fraction of organic components ($\text{i}_{\text{so}}$) in soils and sediments [22,23]. The distribution of PFAS in soil and sediment is complex and dependent on a variety of factors such as PFAS source properties (PFAS type, source input conditions, co-contaminants), soil properties, and meteorological conditions [5,26,27]. More information on the sorption of PFAS in soil can be found in Li et al. 2018 and Li et al. 2019 [22,23,28]. PFAS contaminated soil and sediments can act as a significant reservoir and a secondary source of groundwater and surface water contamination through leaching and percolation [7,9,18–20,26,29,30]. Hence, the treatment of PFAS contaminated soils and sediments is of great importance because this can affect the overall PFAS flux from source areas, control dissolved plume longevity, and impact the efficiency of groundwater remedial approaches.

Most currently available PFAS remediation technologies involve the treatment of PFAS contaminated drinking water, wastewater, and groundwater, whereas the treatment of soils and sediments has received relatively little attention [31]. Solid (soils, sediments and/or sludge) remediation is challenging, expensive, and time-consuming. Traditional containment technologies (capping and off-site disposal) are useful methods for reducing PFAS mobilization and bioaccumulation. However, with these technologies, complete isolation is impossible, continuous monitoring is needed, and long-term liability may be a concern [32,33]. Solidification/stabilization techniques using amendments (e.g., granular activated carbon, organoclays) can effectively immobilize PFAS, reducing leaching and bioavailability, yet long-term efficiency still needs further research [32–34]. Thermal treatment, soil flushing, and soil washing techniques have also been tested at the field scale, however, they are associated with high costs and energy usage requirements. Thermal treatment must overcome the high boiling point of PFAS while soil washing and soil flushing need a large volume of washing solvent and post treatment processing to remove PFAS from contaminated water/regenerant solution [19,20,30]. Other mobilization technologies (e.g., phytoremediation and electrokinetic) and destruction technologies (e.g., bioremediation, chemical oxidation, ball milling, smoldering combustion, and electron beams) are currently being evaluated at the laboratory scale [36–39]. Detailed reviews on the on the remediation of PFAS in soil can be found elsewhere [32–34].

A new and innovative approach is needed to remove and destroy these compounds from soils and sediments. In this research, the contemporary methodology of coupled sonochemistry is explored for on-site remediation of PFAS contaminated soils and sediments [40]. This process relies on acoustic cavitation to both mobilize/desorb PFAS from solids and subsequently mineralize them. Acoustic cavitation, as driven by high-frequency ultrasound (between 500 kHz and 1000 kHz), is shown to be both an efficient and relatively fast method for the complete destruction and mineralization of PFOS and PFOA and is effective over a wide range of concentrations from 10 nM to 10 $\mu$M [41–44]. For a solid–liquid system, to date, most researchers have utilized low-frequency ultrasound for removal of contaminants (primarily hydrophobic compounds like PAHs and PCBs) from sediments [37,45–49] (see table S1). In these studies, the low-frequency ultrasound caused only sonophysical effects which serve in promoting the desorption of organic pollutants from solids, with little to no degradation of contaminants. To the best of the authors’ knowledge, only one study has evaluated the use of ultrasound for treatment of PFAS-contaminated soil, but that also used low-frequency ultrasound coupled with persulfate [37].

In this research, a novel ultrasound delivery method is used to generate acoustic cavitation by combining two different ultrasonic devices to produce an intense ultrasonic field: a horn (or probe) type low-frequency transducer and submersible (or plate) type high-frequency transducers. A schematic of the coupled high and low-frequency ultrasound system for remediating contaminated solids is shown in Fig. 1. The low-frequency horn transducer provides agitation in the system, stirring up solids and bringing them into suspension; (c) The plate transducers induce acoustic cavitation in the system which initiates sonochemical degradation through pyrolysis and radical reactions.

![Fig. 1. (a) A schematic of the coupled high and low-frequency ultrasound system for remediating contaminated solids; (b) The probe transducer provides agitation in the system, stirring up solids and bringing them into suspension; (c) The plate transducers induce acoustic cavitation in the system which initiates sonochemical degradation through pyrolysis and radical reactions.](image-url)
frequency transducer plate.

2. Experimental procedure

2.1. Materials

PFOA (>98.0%) was purchased from TCI America. Potassium perfluorooctanesulfonate (K-PFOS) (>98.0%) was obtained from Sigma-Aldrich. Ammonium acetate, laboratory-grade sodium hexametaphosphate, sodium hydroxide, potassium iodide (>98%), luminol (5-amino-2,3-dihydro-1,4-phthalazinedione), HPLC-grade methanol, total ionic strength adjustment buffer (TISAB II), and fluoride with TISAB II standards were purchased from Thermo Fisher Scientific. Standards of PFOA (>98%) and K-PFOS (>98%) were bought from Wellington Laboratories. Milli-Q water (>18MΩ-cm) was used throughout the experiment.

2.2. Soil preparation and characterization

Two different soils were used to prepare PFAS contaminated soil to reflect the geochemical parameters influencing anion PFAS sorption. Soil 1 (lab soil) was obtained from NJIT civil engineering geotechnical lab and soil 2 (garden soil) was purchased from a local gardening shop. The soils were air-dried and then sieved with 2-mm stainless steel sieve before the experiments.

Percentages of sand, silt, and clay were determined by using ASTM standard D2487 and USCS soil classification [61]. The pH values of the lab and soil 2 (garden soil) was purchased from a local gardening shop. The pH values of the lab and soil 2 (garden soil) was purchased from a local gardening shop.

2.3. Reactor configuration

The coupled sonochemistry technique examined in this study combines two different ultrasonic devices to generate acoustic cavitation: a high-frequency ultrasonic bath reactor equipped with the 700 kHz plate transducer (provided by PCT systems) and a 20 kHz low-frequency probe transducer (Versonics). The 7.5 in. × 6.75 in. × 4 in. ultrasonic bath fabricated from stainless steel consisted of two arrays of ultrasonic transducers attached to a vibrating plate at the bottom of the bath with each array containing two 1 in. × 5 in. piezoelectric elements operating in multiplex mode. Electrical power was supplied to the first piezoelectric elements in each array for one second, followed by a shift in power to the second piezoelectric elements in each array for another second, and so forth. A maximum electrical power of 45 W/sq inch can be applied to each piezoelectric element surface. Therefore, the total maximum power that can be applied to the plate/ultrasonic transducers with two active piezoelectric elements at a given time is 450 W. The probe transducer had a 1/2-inch horn-tip and a maximum operating power of 475 W power (Versonics). Experiments were conducted at room temperature. The reactor contained a stainless-steel cooling coil to dissipate excess heat generated during the sonication process, maintaining a temperature below 45 °C. All experiments were conducted in an air atmosphere.

2.4. Preparing PFAS spiked soil

Stock solutions of PFOS and PFOA (approx. 200 mg/L) were prepared from solid K-PFOS and PFOA. 350 g of soil was mixed with sufficient DI water to prepare the slurry followed by the addition 8.75 mL of stock solution to achieve a final concentration of 5000 µg/kg of PFOS and PFOA. To uniformly distribute PFAS in the soil, a steel stirrer was used to uniformly mix the slurry. The slurry was then allowed to air-dry to remove water.

2.5. Batch desorption

4 g of PFOA and PFOS contaminated soils 1 and 2 were mixed with 40 mL of DI water in separate 50 mL vials. After capping, the vials were vortexed for 30 s and kept in an orbital shaker for 48 h at a speed of 150 rpm. To separate the soil and water, the vials were centrifuged at 3000 rpm for 18 min. Soil samples were dried in an oven at 50 °C before PFAS analysis. Triplicate bottles were prepared for each soil.

2.6. PFAS contaminated soil treatment, analysis, experimentation

To prepare the soil slurry, 150 g of PFAS contaminated soil was mixed with 1.5 L of water in the reactor. Before initiating the treatment process, triplicate samples of the soil slurry were collected in 50 mL vials to evaluate PFAS desorption from soil after mixing with water. The probe transducer was immersed in the reactor near the reactor center, approximately 1.5 in. below the liquid surface. Treatment occurred in one-hour cycles. The solution was sonicated for ten minutes, with the 700 kHz plate transducers operating continuously and the 20 kHz probe transducer operating for only 1 min every 5 min. To dissipate the heat generated during sonication, the solution was cooled for the remainder of the hour. Treatment lasted for 12 cycles, having a total sonication treatment time of 120 min. After treatment, triplicate samples of the soil slurry were collected in 50 mL vials. To separate the soil and water, the vials were centrifuged at 3000 rpm for 18 min. Soil samples were dried in an oven at a temperature of 50 °C before analysis.

Targeted PFAS analysis of solid and liquid samples was completed following a modified method of EPA 8327 using liquid chromatography with tandem mass spectrometry (Agilent 6470 Triple Quadrupole LC/ MS System). The instrumental detection limit was 0.0001 mg/L, with method detection limits varying based on matrix and sample processing requirements. All water samples were diluted with methanol and with a dilution ratio such that the final concentration was below 100 ppb. Diluted samples were filtered using a 0.25 µm polyether sulfone (PES) syringe filter before analysis by LC/MS. Analysis of soil samples used a solid–liquid extraction similar to that previously described in Higgins et al., 2005, 2006 [26,64]. In brief, a 1 g of the dried soil was transferred to a 15 mL polypropylene vial containing 8 mL MeOH, then vortexed for 60 s, shaken on an orbital shaker (ELMI™ 20 mm Amplitude, Fisher

| Soil Description | Clay size (%) | Silt size (%) | Sand size (%) | fom (%) | TOC (%) | pH | Ka,PFOA (L/kg) | Ka,PFOA (L/Kg) |
|------------------|---------------|---------------|---------------|---------|---------|----|---------------|---------------|
| 1 Lab Soil       | 20            | 20            | 60            | 1.03    | 1.1     | 8.625 | 3.273         | 1.896         |
| 2 Garden soil    | N/A           | N/A           | N/A           | 84.3    | 23.2    | 5.558 | 312.32        | 66.558        |

fom denotes percent organic matter, Ka denotes the soil–water partition co-efficient.

Table 1
Soil Properties.
scientific, Pittsburgh, PA, USA) at 300 rpm for 30 min, sonicated in a 20 kHz ultrasound bath for 30 min, and centrifuged at 2500 rpm for 15 min. The aqueous supernatant was collected, the extraction process was repeated for one more cycle, and finally the two extractions supernatant volumes were combined. Due to the detection limitation and upper range of LC/QQQ analyzation, the blank samples were concentrated by drying under a gentle nitrogen stream in a 50 °C water bath and reconstituted in 1.0 mL methanol. Other samples were diluted with methanol to bring the concentration within the detection limits of the instrument. To test the concentration of inorganic fluoride in solution, the F-ISE (fluoride-ion selective electrode) (Thermo-scientific) was used [42,57]. Liquid samples were diluted with TISAB II in a 1:1 ratio. Triplicate readings were taken for each sample to ensure instrument precision.

2.7. Zeta (ζ) potential measurement

The measurements of the Zeta (ζ) potential were carried out on a Malvern Zetasizer Nano ZS. 5 g of PFAS contaminated soil (passing No. 200 sieve (75 μm)) were mixed with 125 mL of the dispersing agent (sodium hexametaphosphate (40 g/L)) solution for one hour until the soil was thoroughly wet. The soil was allowed to soak for at least ten minutes. The soil slurry was then transferred to a 1 L graduated cylinder, filled to volume with DI water, mixed, and left to settle [65]. Clay particles (<2 μm) were pipetted from 5 cm below the water surface after having settled for 24 h [66–69]. To measure the zeta-potential of the nanobubbles generated during the sonication process, the reactor chamber was filled with 1.5 L distilled water followed by ten minutes of sonication [48,70,71]. Samples were collected 5 min after sonication completed and zeta potential measurements were performed.

2.8. Particle size measurement

3.0 g of each air-dried sample was quantified by the Laser Diffraction Method (LDM) and analyzed using a Malvern Mastersizer 3000 with Aero S attachment in three replications, with the size calculated according to the percentage volume distribution based on the Mie theory [74,75]. This laser diffraction particle-size analyzer adopts a full Mie theory and can effectively measure particles ranging between 10 nm and 3500 μm in diameter.

3. Results and discussion

3.1. PFAS contaminated soil treatment using coupled high and low frequency ultrasound

The coupled high and low frequency ultrasound process was evaluated for the treatment of PFAS contaminated soil. Tests were conducted on two types of soils: (1) a lab inorganic soil consisting of clay, silt, and sand; and (2) a highly organic garden soil. Fig. 2a shows the PFOA and PFOS concentrations in both soils before mixing with water, after mixing with water (before treatment), and after 120 min of ultrasound treatment in the reactor. For each soil, PFOA and PFOS concentrations in the liquid phase after mixing soil 1 and soil 2 with water (before treatment) and after 120 min of treatment.

Fig. 2. (a) PFOA and PFOS concentrations in soil 1 and soil 2 before mixing with water, after mixing contaminated soil and water (before treatment), and after 120 min of treatment; (b) Residual PFOA and PFOS concentrations in the liquid phase after mixing soil 1 and soil 2 with water (before treatment) and after 120 min of treatment.

Fig. 3. PFOA and PFOS mass in soil and water before and after treatment for soil 1.
Nevertheless, the treatment method did result in further desorption of PFAS from the soil. Mass balances of PFOA and PFOS in soil and water (both before and after the experiment) are shown in Figs. 3 and 4 for soils 1 and 2, respectively. For soil 1, $92.5 \pm 6.6\%$ of the total mass of PFOA and $78.8 \pm 3.4\%$ of the total mass of PFOS were recovered from both the aqueous and solid phases. For soil 2, these figures were $129.8 \pm 7.7\%$ for PFOA and $80.8 \pm 5.3\%$ for PFOS. Results from both tests suggest minimal to no PFAS degradation was achieved. In addition, the release of fluoride following potential PFAS degradation was monitored, and negligible fluoride release was detected before or after treatment, also indicating no PFAS degradation was achieved (Fig. S2).

Previous studies have shown that the presence of solids in heterogeneous systems can enhance cavitation activity due to an increase in the number of nucleation sites [76, 77]. This can be explained further by considering the roughness of the solid surface which facilitates the entrapment and stabilization of gas pockets. Not only does this increase the number of nucleation sites but also leads to a reduction in the tensile strength of the liquid. However, sand-sized particles specifically can cause wave scattering and attenuation leading to a reduction in sonochemical activity and less energetic bubble implosion [77, 78]. Thus, the presence of soil may have the inhibited the mineralization of PFAS via pyrolysis in the solid–liquid system.

### 3.2. Desorption of PFAS contaminated soil

A batch desorption experiment was performed to evaluate the impact of the proposed ultrasound treatment on the desorption of PFAS from soil. The percentages of PFOA and PFOS leached from soils 1 and 2 are shown in Fig. 5 and are compared with the percent PFOS and PFOA leached from the soil before (after mixing with DI water) and after the 120-minute ultrasound treatment. PFOA contains 8 carbons but only 7 comprise the fluorinated carbon chain, whereas all 8 carbons of PFOS are fluorinated, resulting in higher hydrophobicity and lower leaching potential [79, 80]. These properties explain the larger percentage PFOA being removed from the soil compared to PFOS as shown in Fig. 5. As mentioned earlier, the higher organic matter content and lower pH of soil 2 must have led to a lower desorption of PFOA and PFOS compared to soil 1. 90.6 ± 2.74% of PFOA and 91.6 ± 1.17% of PFOS were leached from soil 1 in the desorption experiment, whereas for soil 2 these values were only $28 \pm 0.2\%$ of PFOA and $1 \pm 3.1\%$ of PFOS. Comparing PFAS removal in the desorption experiment with the ultrasound treatment, it is clear that ultrasound enhances the percentage removal, particularly for soil 2 with its higher $K_d$ value. For the soil 2, $68.8 \pm 1.8\%$ of PFOA and $45.4 \pm 4.1\%$ of PFOS were leached from the soil after ultrasound treatment compared to only $28 \pm 0.2\%$ of PFOA and $1 \pm 3.1\%$ of PFOS leaching from the soil in the desorption experiment. This suggests that the ultrasound treatment method enhanced the removal of PFOA and PFOS from soil compared to soil washing using water.

Previous studies of PFAS have shown that soil characteristics have a strong influence on partitioning/sorption capacity [22–24, 80]. This was consistent with our results, where soil 1, which had a low organic carbon content and high pH, had the lower sorption capacity and greater leaching potential as compared to soil 2 with a higher organic carbon content and low pH. To the best of the authors’ knowledge, there are limited studies exploring ultrasound enhanced desorption of PFAS. Lei et al., 2020 investigated the direct sonication of PFAS contaminated soil using dual frequency treatment at 20 and 43 kHz coupled with persulfate and reported 100% PFOA degradation but only partial degradation for PFOS [37]. However, this study did not present a mass balance or an initial fluoride release, suggesting the possibility that minimal to no PFAS degradation was achieved [37].

### 3.3. Zeta potential for contaminated soil and nanobubbles

In the case of the soil-liquid slurry, PFAS are adsorbed to the surface of the solid particle through electrostatic interaction and hydrophobic interactions. To further investigate the fate of the electrostatic interaction between nano bubbles and PFAS, zeta potential measurements for contaminated soil were carried out. The zeta potential for soil 1 and 2 were $-48.68 \pm 2.44$ and $-83.12 \pm 12.49$, respectively. Organic matter in soil 2 would have decreased the zeta potential of the particles [68]. In comparison, the zeta potential of the nanobubbles generated in sonication treatment was $-14.92 \pm 1.34$. The surface of the soil minerals is usually negatively charged; similar results has been reported in several other studies [68–71]. Concurrently, the surface of the nanobubbles is also usually negatively charged [52, 72, 73]. These negative charges on the bubble surface and the soil particle will have competing tendencies, resulting in electrostatic repulsion between nanobubbles surfaces and the PFAS adsorbed on soil surface. This would have inhibited the accumulation of PFAS at the bubble–liquid interface, preventing their mineralization via pyrolysis. Thus, the electrostatic repulsion between nanobubbles and soil particles can limit the sonochemical degradation of PFAS in soil slurries.

### 3.4. Change in organic matter, total organic carbon content and particle size before and after the treatment

Ultrasound at both low and high frequencies can promote the leaching of contaminants from soils. The mechano-acoustic effects of
ultrasonic waves accelerate the desorption and facilitate the removal of entrapped contaminants by increasing the percolation rate and the erosion/porosity of soil [30,43,44,46,73–75]. The sonication energy can also result in a reduction in the particle size and the soil organic matter [84]. Total organic matter content in the soils decreased following ultrasound treatment. Organic matter and total organic carbon content tests were performed on soils collected after ultrasound treatment. Following treatment, the organic matter in soil 2 reduced from 84.3% to 72.7% and the total organic carbon content in soil 2 reduced from 23.2% to 21.7%. Since organic matter content correlates with PFAS sorption onto the soil, the decrease in total organic content might also explain the enhanced PFAS desorption after ultrasonic treatment.

The effect of ultrasound on the soil particle size was also investigated. The particle size analyzer test was performed on both soils before and after treatment by laser diffraction using Malvern Mastersizer 3000, dry S. Results are shown in Fig. 6. The particle size distribution for both soils 1 and 2 is shifted toward the left for soil treated after ultrasound as compared to soil before treatment, suggesting that soil particles have been eroded and size of soil particles have been reduced by ultrasound treatment. This can be explained by considering the different mechano- acoustic effects of ultrasound on heterogeneous media, explained elsewhere [35,46,47,50,81–83]. In the present study, the mechanical effects of ultrasound, such as microjet formation upon bubble collapse and microstreaming around oscillating bubbles (also known as acoustic streaming), must have enhanced desorption of PFAS. Microjets are responsible for the erosion/cleaning effect of ultrasound and microstreaming can disrupt inter-molecular bonds [19,85,86].

4. Summary and conclusion

PFAS have been detected in soils and sediments exposed or impacted by landfill leachate or biosolids, direct contaminated discharge, and contaminant transport from atmospheric deposition. There are a limited number of techniques to remediate PFAS contaminated soils, and most of the methods are based on the PFAS stabilization strategy. A new and innovative treatment method for achieving concurrent treatment of soil and destruction of PFAS was evaluated in this research.

This paper presents a detailed investigation of the use of high and low frequency ultrasound for the treatment of the PFAS contaminated soils. It was demonstrated that PFAS concentrations in artificially contaminated soil can be significantly reduced by ultrasound treatment, particularly for soils with high organic matter contents. The concentrations of PFOA and PFOS in soil 2 (23.2% TOC) were reduced by 68.8 ± 1.8% and 45.4 ± 4.2%, respectively, after ultrasound treatment compared with 28 ± 0.2% and 1 ± 3.1% after mixing soil with water. In addition, results indicate that PFOA and PFOS can easily desorb from sediments with low organic matter into the aqueous phase. For soil 1 (1.1% TOC), the concentrations of PFOA and PFOS were reduced by 90.6 ± 2.7% and 91.6 ± 1.1% just after mixing with water. While the ultrasonic transducers were capable of desorbing PFAS from soil, neither a decrease in total PFAS mass in the system or an increase in fluoride ion concentrations were observed, suggesting that little to no sonochemical degradation of PFAS occurred. The presence of nanobubbles and hydroxyl radical formation as evidenced by KI dosimetry, calorimetry, and SCL tests show that acoustic cavitation and sonochemical reactions occurred in the reactor when a liquid medium was sonicated. This indicates that the absence of PFAS degradation in the reactor was likely due to the presence of soil. It is probable that the solids in the system caused wave scattering and attenuation of the high frequency ultrasound, decreasing sonochemical activity. It is also possible that the electrostatic repulsion between the negatively charged soil and nanobubble surfaces inhibited the accumulation of PFAS at the bubble–liquid interface, preventing their mineralization via pyrolysis. Thus, the sonochemical degradation of PFAS in soil slurries is limited by soil–nanobubble interactions.

CRediT authorship contribution statement

Jitendra A. Kewalramani: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft. Boran Wang: Investigation, Formal analysis, Writing – review & editing. Richard W. Marsh: Investigation, Formal analysis, Writing – original draft. Jay N. Meegoda: Conceptualization, Supervision, Methodology, Writing – review & editing. Lucia Rodriguez Freire: Supervision, Methodology, Validation, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
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Appendix A. Supplementary data

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