ABSTRACT: For uranium extraction from seawater, development of new stable and reusable sorbents with high affinity and good selectivity for U(VI) is required. Herein, a new phosphate-functionalized polyethylene (denoted PO₄/PE) was synthesized via a simple Ar-jet plasma treatment of PE in concentrated H₃PO₄ and was employed in U(VI) extraction from seawater. The prepared PO₄/PE shows superior performance in the extraction of trace U(VI) from seawater. The adsorption process followed the second-order kinetics model and the Langmuir model. The maximum adsorption capacity of PO₄/PE for U(VI) reaches 173.8 mg/g at pH 8.2 and 298 K. PO₄/PE can be effectively regenerated by 0.1 mol/L Na₂CO₃ and reused well even after eight cycles. Experimental results offer a new approach for U(VI) uptake from seawater.

INTRODUCTION

The widely used fossil fuels (petroleum and coal) are the main source of environmental pollution and greenhouse effect. The serious environmental impact and the limited reserves of fossil fuels have stimulated the search for alternative energy sources. Nuclear energy is widely considered as a good substitute for fossil fuels because of its unparalleled energy density (about 2.7 million times that of coal in theory) and low emission of CO₂.¹

The development of nuclear energy leads to a significant increase in the demands for uranium, which will be the basic material of nuclear plants in the future. However, the natural resource of uranium is limited and recycling of uranium from radioactive wastewater is difficult. The 4.5 billion tons of U(VI) in the ocean can be considered as a sustainable and environmentally friendly substitute of the uranium terrestrial supply to sustain the demands of the nuclear power industry in the future.²⁻⁴ The use of traditional adsorbents to recover U(VI) from seawater is very challenging because the concentration of U(VI) in seawater is very low (3.3 mg/L) and abundant competing ions (such as alkaline metals, alkaline earth metals, and heavy metals) coexist in seawater. Thereby, the development of adsorbents with a high affinity, excellent selective property, and simple preparation process is critical.⁵ From the initial inorganic adsorbents to the most studied polyamidoxime (PAO)-based adsorbents and to more recent metal–organic frameworks, great efforts have been devoted to developing efficient adsorbents for extracting U(VI) from seawater.

The properties of surface functional groups on sorbent surfaces determine its adsorption affinity and capacity. Among the various functional groups, phosphate groups (−PO₄) have attracted increased attention and play an important role in actinide separation because of their excellent stability, strong irradiation resistance, and high complexion ability with actinide element ions (such as Pu(IV), Am(III), and U(VI)).¹⁻⁶⁻¹¹ Over the past years, various −PO₄-functionalized adsorbents have been widely applied in U(VI) extraction from solution.¹⁻⁸⁻¹⁹ However, −PO₄-functionalized adsorbents are usually synthesized by chemical modification¹⁻¹¹⁻¹⁵ and polymerization techniques with appropriate organic reagents.⁸⁻¹⁰,¹⁶⁻¹⁹ To date, the development of a simple and efficient synthesis technique is important for the potential application of −PO₄-functionalized adsorbents for U(VI) capture.

Herein, a new phosphate-functionalized polyethylene (PO₄/PE) was synthesized by a simple Ar-jet plasma treatment of PE in H₃PO₄ and employed for U(VI) capture. PE was selected just because this is one of the most studied substrate materials in uranium extraction from seawater.

RESULTS AND DISCUSSION

Characterization. Scanning electron microscopy (SEM) photomicrographs of PE, PO₄/PE, and PO₄/PE after
regenerating eight times are shown in Figure 1. The surface of PE (Figure 1A,B) is smooth with many microgrooves along the fiber, whereas that of PO₄/PE (Figure 1C,D) is rough and covered with a thin layer. Meanwhile, no notable differences of PE surface morphology before and after immersion in H₃PO₄ are observed. Thereby, the thin layer is solid evidence for the successful induction of −PO₄ on the surface of PE. Meanwhile, the sizes of PO₄/PE become larger as compared to those of PE (Figure 1G), which can be ascribed to the penetration of water molecules into the PE polymeric structure and hydrolysis of the modified functional groups. After recycling eight times, PO₄/PE (Figure 1E,F) still retained the stacking morphology as that of raw PO₄/PE, indicating excellent stability and good reusability of PO₄/PE. Elemental mapping images (Figure 1H) indicate that O, P, and U are homogeneously dispersed on the PO₄/PE surface, which demonstrates the excellent adsorption capacity of PO₄/PE for U(VI).

Figure 1. SEM images of PE (A, B), PO₄/PE (C, D), and PO₄/PE after regenerating eight times (E, F). Particle size distributions of PE, PO₄/PE, and U(VI)-laden PO₄/PE (G) and elemental mappings of U(VI)-laden PO₄/PE (H).
The mechanical properties of PO₄/PE is an important parameter for its real application in U(VI) extraction from seawater, which was studied at its break point (Figure 2A). The mechanical properties of substrates are usually significantly reduced in traditional synthesis processes (such as the γ-ray irradiation and electron-beam irradiation techniques). Xing et al.²⁰ reported that the tensile strength of a PE fiber diminished significantly from 3.0 × 10⁹ to 1.3 × 10⁹ Pa after γ-irradiation for 50 kGy. In our case, the tensile strengths are 1.64 × 10⁹ and 1.63 × 10⁹ Pa for PE and PO₄/PE, respectively. It means that the side effect of the traditional synthesis method is well-eliminated in this work. To evaluate the effect of the introduction of −PO₄ by plasma treatment on the PE framework, PE and PO₄/PE are characterized by powder X-ray diffraction (XRD) pattern (Figure 2B). The fairly similar XRD patterns of PE and PO₄/PE further demonstrate no significant structure damages of PE after Ar-jet plasma treatment in H₃PO₄.

The effect of the introduction of −PO₄ by plasma treatment on the PE surface was also studied by the Raman spectroscopy technique. As shown in Figure 2C, PE presents “fingerprint” Raman peaks of C−C vibrations at ~1061 (asymmetric vibrations) and ~1128 cm⁻¹ (symmetric vibrations) and of −CH₂− vibrations at ~1293 (twisting vibrations in all crystalline phases) and ~1415 cm⁻¹ (wagging vibrations in the orthorhombic crystalline phase).²¹ According to the relative intensity ratio of the peak at ~1415 cm⁻¹ to that at ~1293 cm⁻¹,²² the degree of orthorhombic crystallinity of PE was decreased after the introduction of −PO₄ by plasma treatment. It suggests that parts of the C−C/C−H bonds were broken, which react with H₃PO₄ to form −PO₄ groups on the PE surface. In the high-frequency region, the peak intensity ratio of −CH₂− (~2846 cm⁻¹) to −CH₃ (~2880 cm⁻¹) decreases after the Ar-jet plasma treatment in H₃PO₄, which indicates that the formation of −PO₄ groups on the PE surface is mainly by breaking C−H bonds in −CH₂−.

Figure 2. Tensile strengths (A); XRD patterns (B); Raman spectra (C); and X-ray photoelectron spectroscopy (XPS) P 2p (D), XPS C 1s (E), and XPS O 1s spectra (F) of PE, PO₄/PE, and U-laden PO₄/PE.
To study the existing form of −PO$_4$ groups on the PO$_4$/PE surface and its U(VI) adsorption mechanism, PE, PO$_4$/PE, and U-laden PO$_4$/PE were studied by the XPS technique. The P 2p spectra (Figure 2D) of PO$_4$/PE and U-laden PO$_4$/PE are deconvoluted into three components (Table 1): −PO$_4$, poly(−PO$_4$), and metaphosphates (P−O−P). The intensities signification of U(VI)-laden PO$_4$/PE can be explained by the complexation of U(VI) on the surface of PO$_4$/PE in the form of P−O−U(VI) bonds. After U(VI) adsorption, U-laden PO$_4$/PE also showed new peaks centered at ~135.1 eV, indicating the effective binding of U(VI) on the surface of PO$_4$/PE in the form of P−O−U(VI) bonds. The decreased XPS P 2p peak intensity of U(VI)-laden PO$_4$/PE can be explained by the broadening of the XPS P 2p spectrum and the coverage of bound U(VI) on the surface of U(VI)-laden PO$_4$/PE. Meanwhile, parts of poly(−PO$_4$) would decompose into soluble −PO$_4$ during the U(VI) adsorption process, which would dissolve in the solution. The C 1s spectral (Figure 2E) intensities significantly decreased after the Ar-jet plasma treatment in H$_3$PO$_4$ and deconvoluted into five components (Table 2) at 283.4 ± 0.1, 284.9 ± 0.1, 286.2 ± 0.1, 287.3 ± 0.1, and 288.8 ± 0.1 eV, which can be assigned to carbide carbon; C−C; and carbon atoms bound to one (C=O and C−O−P), two (C=O), and three (−COOH) oxygen atoms, respectively. The carbide C and graphite carbon (C−C) are the main species of carbon on the PE and PO$_4$/PE surfaces. The contents of carbide carbon and C−OH decreased after the Ar-jet plasma treatment in H$_3$PO$_4$ which indicates that the −PO$_4$ groups on the PE surface are introduced mainly via the cleavage of carbide C and C−OH. The O 1s spectra of PE and PO$_4$/PE (Figure 2F) can be deconvoluted into four components at 531.0 ± 0.1, 532.5 ± 0.1, 535.0 ± 0.1, and 537.5 ± 0.1 eV, which are related to the double-bonded oxygen (C=O), single-bonded oxygen (C−O and −O−P in −PO$_4$), chemisorbed oxygen and water, and −OH, respectively (Table 3).

Table 1. Curve Fitting Results of XPS P 2p Spectra

| peak                  | position (eV) | FWHM (eV) | %    |
|-----------------------|---------------|-----------|------|
| PO$_4$/PE             | −PO$_4$       | 133.20    | 1.59 | 11.0 |
|                       | poly(−PO$_4$)| 134.00    | 2.83 | 89.0 |
| U-laden PO$_4$/PE     | −PO$_4$       | 133.20    | 0.47 | 7.03 |
|                       | poly(−PO$_4$)| 133.85    | 1.43 | 20.5 |

*“Binding energy.” Full width at half-maximum.

Table 2. Curve Fitting Results of XPS C 1s Spectra

| peak                  | position (eV) | FWHM (eV) | %    |
|-----------------------|---------------|-----------|------|
| PE                    | carbide C     | 283.30    | 1.91 | 40.5 |
|                       | C−C           | 284.95    | 1.47 | 56.2 |
|                       | C−O           | 286.30    | 2.72 | 2.88 |
|                       | C=O           | 287.36    | 1.00 | 0.00 |
|                       | COOH          | 288.70    | 1.97 | 0.45 |
| PO$_4$/PE             | carbide C     | 283.30    | 2.08 | 34.2 |
|                       | C−C           | 284.92    | 1.67 | 64.6 |
|                       | C−O           | 286.10    | 1.00 | 0.00 |
|                       | C=O           | 287.40    | 1.21 | 0.81 |
|                       | COOH          | 288.70    | 1.05 | 0.36 |
| U-laden PO$_4$/PE     | carbide C     | 283.30    | 1.03 | 0.74 |
|                       | C−C           | 284.82    | 1.36 | 83.5 |
|                       | C−O           | 286.10    | 1.16 | 11.03|
|                       | C=O           | 287.38    | 1.58 | 4.22 |
|                       | COOH          | 288.70    | 2.04 | 0.54 |

and 288.8 ± 0.1 eV, which can be assigned to carbide carbon; C−C; and carbon atoms bound to one (C−OH and C−O−P), two (C=O), and three (−COOH) oxygen atoms, respectively. carbide C and graphite carbon (C−C) are the main species of carbon on the PE and PO$_4$/PE surfaces. The contents of carbide carbon and C−OH decreased after the Ar-jet plasma treatment in H$_3$PO$_4$ which indicates that the −PO$_4$ groups on the PE surface are introduced mainly via the cleavage of carbide C and C−OH. The O 1s spectra of PE and PO$_4$/PE adsorption U(VI), the enhanced U(VI) adsorption capability of PO$_4$/PE for U(VI).

Adsortion. The prepared PO$_4$/PE shows excellent performance in selective adsorption of U(VI) from seawater. As shown in Figure 3A, the introduced −PO$_4$ groups increase the enrichment of U(VI) on PO$_4$/PE under experimental conditions. Moreover, the adsorption of U(VI) on PE and on H$_3$PO$_4$-dispersed PE (data are not shown) is similar under the experimental uncertainties. Thereby, the enhanced U(VI) adsorption can be attributed to the strong complexation of U(VI) and −PO$_4$ groups on the PE surface. Meanwhile, the adsorption of U(VI) on PO$_4$/PE increases with increasing pH and then slowly decreases with a further increase in pH. It reveals that U(VI) adsorption on the PO$_4$/PE surface is fairly pH-dependent. The charges of the U(VI) species in the solution are affected by solution pH. Meanwhile, the available −PO$_4$ groups gradually deprotonate with increasing solution pH.

It is well known that seawater is a very complex matrix and alkaline metal ions (such as Na(I) and K(I)) and alkaline earth metal ions (such as Ca(II) and Mg(II)) are the predominant cations in seawater. To estimate the effect of ionic strength on PO$_4$/PE adsorption capability for U(VI), the effects of those cations were studied using concentrations of 0.010–1.0 mol/L and are shown in Figure 3B. PO$_4$/PE exhibits high selectivity for U(VI) against Na(I) and K(I) because the adsorption of U(VI) against Na(I) and K(I) because the adsorption of U(VI) just slightly decreases with an increase in their concentrations. Ca(II) and Mg(II) have a more significant negative effect than that of Na(I) and K(I) on U(VI) adsorption, which can be explained by the complexation reaction among U(VI), CO$_3^{2−}$, and alkaline earth metal ions in the solution. Many researchers found that these complexes determine the extraction behavior of U(VI) in seawater because of the high concentrations of Ca(II) and Mg(II). It is well known that many metal ions, such as...
Cu(II), Al(III), Fe(III), and V(IV), coexist with U(VI) in seawater and these metal ions result in serious interference in U(VI) separation. Therefore, the selective extraction capability of PO₄/PE toward U(VI) was compared to that toward the coexisting metal ions. The results in Figure 3C show that U(VI) adsorption on the surface of PO₄/PE is significantly higher than that of other metal ions under the same experimental conditions, which reveals the high selectivity of PO₄/PE toward U(VI) in seawater in comparison to that toward other metal ions, especially V(IV). A number of researchers reported a similar highly selective retention behavior of U(VI) on −PO₄ functionalized adsorbents, and they explained it by the strong complexation of −PO₄ groups with U(VI). Furthermore, the competition between U(VI) and V(IV) is one of the most serious challenges for the commercial application of most adsorbents in U(VI) recovery from seawater. Our results show that PO₄/PE can tolerate V(IV) interference, highlighting the potential application of PO₄/PE in the selective extraction of U(VI) from seawater.

The effect of reaction time on the adsorption of U(VI) by PO₄/PE was studied because seawater temperature varies widely with the position and season. As depicted in Figure 3D, the adsorption percent increases quickly in the first 6 h and then maintains the level with the increasing reaction time. To further investigate the adsorption kinetics, the pseudo-first-order kinetic models ($q_t = q_e \times (1 - \exp(-k_1t))$, where $k_1$ (h⁻¹) is the adsorption rate constant, $q_e$ (mg/g) and $q_t$ (mg/g) are the equilibrium and experimental adsorption capabilities, respectively) and pseudo-second-order kinetic models ($q_t = q_e \times t/((K'q_e) + t)$, where $K'$ (g/(mg·h)) is the adsorption rate constant) are used to simulate the experimental data. The related parameters are shown in Table 4. According to...
Table 4. Parameters for Kinetic Models of U(VI) Adsorption on PE and PO4/PE at pH = 8.2, T = 25 °C, m/V = 0.20 g/L, and C_{U(VI)initial} = 50.0 mg/L

| adsorbent | kinetic model | k_1 (h^{-1}) | q_e (mg/g) | R^2 | K' (g/(mg h)) | q_e (mg/g) | R^2 |
|-----------|---------------|--------------|------------|-----|---------------|------------|-----|
| PE        | pseudo-first-order model | 6.27         | 151.9      | 0.622 | 114           | 153.1      | 0.999 |
| PO4/PE    | pseudo-second-order model | 3.02         | 68.5       | 0.762 | 104           | 70.0       | 0.961 |

Table 5. Parameters Calculated from the Langmuir and Freundlich Models for U(VI) Adsorption on PO4/PE at pH = 8.2

| Langmuir model | Freundlich model |
|---------------|-----------------|
| C_{max} (mg/g) | b (L/mg) | K (mg/g) | n | R^2 |
| 288 K         | 170.2 | 0.296 | 54.3 | 0.313 | 0.945 |
| 298 K         | 173.8 | 0.352 | 59.9 | 0.298 | 0.959 |
| 308 K         | 176.7 | 0.383 | 62.7 | 0.293 | 0.954 |

According to the R^2 values, the adsorption process followed the Langmuir model, which indicates that the adsorption of U(VI) on PO4/PE surfaces is localized in a monolayer. According to the analysis results, the maximum adsorption capacities (C_{max}) of PO4/PE for U(VI) at pH 8.2 are ~170.2, ~173.8, and ~176.7 mg/g at 288, 298, and 308 K, respectively, which are comparable to those of other adsorbents (Table 6), which highlighted the application of our method in preparing PO4/PE for selectively extracting U(VI) from seawater. The related distribution coefficient (K_d) and thermodynamic parameters are also calculated and are shown in Figure 3F and Table 7, respectively. The negative Gibbs free energy change (ΔG°), the positive value of entropy change (ΔS°), and the positive value of standard enthalpy change (ΔH°) reveal that U(VI) adsorption on the PO4/PE surface is an endothermic and spontaneous process.

The regeneration–reusing property of PO4/PE was studied to further estimate its potential application. Na2CO3 is selected as the eluting agent, and the results are depicted in Figure 4A. The elution of U(VI) from the PO4/PE surface quickly increases with increasing Na2CO3 concentrations from 0.002 to 0.1 mol/L and then remains at this level. The result demonstrates the effective regeneration of PO4/PE in a 0.1 mol/L Na2CO3 solution. Thereby, the 0.1 mol/L Na2CO3 solution is selected for the regeneration of PO4/PE. The effect of recycling times on the adsorption capability of PO4/PE for U(VI) was studied and is shown in Figure 4B. The regenerated PO4/PE still exhibits high adsorption capability for U(VI) even after regenerating eight times under experimental conditions, which reveals the excellent reusability property of PO4/PE in the extraction of U(VI) from seawater.

Because of the attractive adsorption capability of PO4/PE for U(VI), the real application of PO4/PE in U(VI) extraction from deionized and real waters was studied, and the results are shown in Table 8. First, we studied the performance of PO4/PE in the restoration of deionized water (m/V = 0.20 g/L, pH = 8.2) in which 5–50 mg/L U(VI) was intentionally added. PO4/PE can quantitatively extract mg/L levels of U(VI) from the aqueous solution. Encouraged by the above results, we then studied the adsorption capability of PO4/PE in extraction of U(VI) from real water. The aqueous solution from Chaohu lake was selected and μg/L level of U(VI) was intentionally added. PO4/PE still showed excellent adsorption efficiency for μg/L levels of U(VI). Thereby, the real application of PO4/PE in the extraction of U(VI) from seawater was performed and the seawater from East China Sea was used. The reaction temperature, contact time, PO4/PE mass, and seawater volume were 25 ± 1 °C, 24 h, 200 mg, and 100 mL, respectively. The adsorption efficiency of ~39% for 3.8 μg/L U(VI) in seawater reveals the excellent adsorption efficiency of PO4/PE in real separation of trace U(VI) from seawater.
CONCLUSIONS

From the results of PO₄/PE characterization and U(VI) adsorption on PO₄/PE under different experimental conditions, the following conclusions can be drawn: (1) A PO₄/PE adsorbent with high adsorption capacity for U(VI) was synthesized by a simple and efficient Ar-jet plasma treatment technique. (2) A plausible reaction mechanism is proposed and confirmed based on microscopic and spectroscopic characterization. (3) PO₄/PE exhibits excellent adsorption capability for U(VI) in seawater (maximum adsorption capacity of 173.8 mg/g at pH 8.2 and 298 K and regeneration–reuse property). The results in this work highlight the application of PO₄/PE as an adsorbent in the extraction of U(VI) from seawater.

EXPERIMENTAL SECTION

Synthesis of PO₄/PE. The PE fiber (TYZ Safetex FT-103) was purchased from Beijing Tongyizhong Advanced Material Company. PO₄/PE was synthesized by the Ar-jet plasma treatment of PE in H₃PO₄. Briefly, 5.0 g of PE and 100 mL of 85% H₃PO₄ were added into a 250 mL round-bottom flask and treated by Ar-jet plasma (high-purity Ar) for 60 min at room temperature and atmospheric pressure under continuous stirring. The Ar-jet plasma conditions were as follows: Ar of 200 sccm, voltage of 5000 V, and electrical current of 1.0 mA. The PE fibers floated on the surface of 85% H₃PO₄ initially and were gradually dispersed in the H₃PO₄ solution with the progress of Ar plasma treatment. The obtained material was washed with Milli-Q water after the Ar-jet plasma treatment and dried at 60 °C for 24 h. To evaluate and compare the effects of Ar-jet plasma treatment, H₃PO₄-dispersed PE was also synthesized by the same method. The diagram of the Ar-jet plasma apparatus used in this work is shown in scheme 1.

Characterization. The physiochemical properties of PO₄/PE were characterized by SEM, element distribution mapping, tensile strength and elongation measurement, powder XRD, Raman spectroscopy, and XPS in detail. SEM images and element distribution mapping were obtained by a field emission-SEM (JSM-6320F; JEOL). The tensile strengths and elongation properties were measured on a tensile tester (MTS criterion model 43-3041E). The powder XRD pattern was collected by XRD (Rigaku D/max 2550) at ambient temperature. Raman spectroscopy analysis was performed by a Raman spectrometer (LabRam HR). XPS spectroscopies were performed by a surface microanalysis system (ESCALab220i-XL; VG Scientific) equipped with an Al Kα (hλ = 1486.6 eV)

Table 8. Selected Results of U(VI) Adsorption on the PO₄/PE Surface

| sample               | m/V (g/L) | initial [µg/L] | final [µg/L] | adsorption (%) |
|----------------------|-----------|----------------|--------------|----------------|
| deionized water      | 8.2       | 0.2            | 50 000       | 61.0           |
|                      | 8.2       | 0.2            | 20 000       | 86.0           |
|                      | 8.2       | 0.2            | 10 000       | 90.0           |
|                      | 8.2       | 0.2            | 1000         | 89.8           |
| Chaohu lake water a  | 8.2       | 0.2            | 1000         | 80.5           |
|                      | 8.2       | 2.0            | 100          | 68.8           |
|                      | 8.2       | 2.0            | 10           | 58.6           |
| contaminated seawater| 8.2       | 2.0            | 100          | 62.2           |
|                      | 8.2       | 2.0            | 10           | 48.0           |
| original seawater b  | 8.2       | 2.0            | 3.8          | 39.4           |
|                      | 8.2       | 2.0            | 2.3          | 39.4           |

The main composition of Chaohu lake water was 6.6 mg/L Na(I), 1.7 mg/L K(I), 34 mg/L Ca(II), 7.6 mg/L Mg(II), 3.1 mg/L Cl⁻, 12 mg/L SO₄²⁻, and 141 mg/L HCO₃⁻. The pH values of the suspensions were kept at the initial value by adding negligible amount of NaOH solutions. The main composition of seawater was 9.8 g/L Na(I), 0.36 g/L K(I), 0.42 g/L Ca(II), 0.93 g/L Mg(II), 17 g/L Cl⁻, 2.1 g/L SO₄²⁻, and 0.12 g/L HCO₃⁻. The pH values of the suspensions were kept at the initial value by adding negligible amount of NaOH solutions.

Scheme 1. Schematic Diagram of the Synthesis of PO₄/PE
source at a chamber pressure of 3 × 10−3 mbar. The surface charging effects were corrected with the C 1s peak at 284.4 eV as a reference.

**U(VI) Adsorption on PO4/PE and on PE.** To evaluate the effect of –PO4 group functionalization by the Ar-jet plasma treatment on the adsorption capability of PE toward U(VI), U(VI) adsorption on PO4/PE and PE was studied by the batch adsorption technique. After the adsorbent and salinity (such as NaCl) were pre-equilibrated for 24 h, the U(VI) solution (UO2CO3) and Milli-Q water were added to achieve the desired components and the pHs of the suspensions were adjusted by the corresponding acid and basic solutions. After shaking for 48 h, the supernatant was filtered by 0.45 μm membrane filters. The final U(VI) concentrations (of mg/L and μg/L levels) in the supernatants were measured on an Optima 2100 DV inductively coupled plasma (ICP) atomic emission spectroscopy system (Perkin Elmer) and on an ICP mass spectrometry (ICP-MS, Thermo Scientific X-Series II) system, respectively.

**Regeneration–Reuse of PO4/PE.** Na2CO3 was selected for the regeneration of PO4/PE. About 25 mg of U-laden PO4/PE was regenerated in 50 mL of Na2CO3 solution for 24 h followed by rinsing with water and drying at 60 °C. Thus, regenerated PO4/PE was obtained and reused in following experiments.

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