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100871

ThO_2
ThO_2 NH_4 4 UO_2 CO_3 3
2 mmol/L NH_4HCO_3 20 mmol/L -HCl
NH_4 4 UO_2 CO_3 3 pH = 8.45 ThO_2
20 mg/L ThO_2 6.52 mg/g ThO_2
Freundlich ThO_2
ThO_2 UO_2^{2+} Ca^{2+} Cu^{2+} Ni^{2+}

ThO_2
O615 A 1000-0518 2017 10-1177-09
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ThO_2 [1-3] [4-6]
[7-8] [9-10]

ThO_2
Wang [7] ThO_2 ThO_2
Misha [8] ThO_2
Ba^{2+} Sr^{2+} Gupta [11]
ThO_2 UO_2^{2+}
ThO_2
UO_2 CO_3 $_3^{4-}$ ThO_2 CO_3^{2-}
ThO_2 NH_4 4 UO_2 CO_3 3 Tris-HCl

1

1.1

Tecnai G2 T20 TEM FEI 200 kV Nova Nano SEM 430
SEM FEI 10 kV Rigaku Dmax-2000 X XRD
Rigaku Cu α λ = 0.15418 nm Prodigy ICP-AES Leeman Dionex model ICS-900 Dionex IonPac AS16

2017-05-11 2017-06-12 2017-07-04
TZ2016004 U1507203 91226112

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250 × 4 mm IonPac AG 1650 × 4 mm 30 mmol/L KOH
0.8 mL/min AMMS-300 75 mA T70 Delta 320
pH Mettler Toledo ASAP-2010 Micrometer
Th NO_3 4 · $4H_2O$
UO_2 NO_3 2 · $6H_2O$ Chemapol
Tris

1.2 NH_4 4 UO_2 CO_3 3

12 UO_2 NO_3 2 · $6H_2O$ 350 ℃ 3 h UO_3
1.8 g UO_3 90 mL NH_4HCO_3 60 ℃
NH_4 4 UO_2 CO_3 3
3% NH_4HCO_3 40 ℃ NH_4 4 UO_2 CO_3 3 71%
/% C 6.87 6.90 H 3.09 3.09 N 10.70 10.73

1.3 ThO_2

7 ThO_2 1.1042 g Th NO_3 4 · $4H_2O$ 60 mg 2 mL
50 mL 100 mL
120 ℃ 4 h
ThO_2 800 ℃ 5 h ThO_2

1.4 ThO_2

30 mg ThO_2 10 mL 20 ~
120 mg/L NH_4 4 UO_2 CO_3 3 2 mmol/L NH_4HCO_3 20 mmol/L Tris-HCl pH =
8.45 25 ℃ ICP-AES
mg/g 1

$$\frac{\rho_0 \quad \rho_e}{} \tag{1}$$

ρ_0 mg/L ρ_e mg/L
L g ThO_2
30 mg ThO_2
Na^+ Mg^{2+} Ca^{2+} Cu^{2+} Ni^{2+} 20 mg/L
UO_2 CO_3 $_3^{4-}$ Cl^- Br^- NO_3^- SO_4^{2-} 20 mg/L 25 ℃
ICP-AES
2

$$_{U/M} \quad \frac{_{d\,U}}{_{d\,M}} \tag{2}$$

$_{d\,U}$ mL/g $_{d\,M}$ mL/g ThO_2 $_d$ 3

$$_d \quad \frac{\rho_0 \quad \rho_e}{\rho_e} \times 1000 \tag{3}$$

1.5 ThO_2

ThO_2 10 mL 0.01 ~ 0.05 mol/L 25 ℃
ICP-AES 4

$$\frac{\rho}{\rho_0 \quad \rho_e} \tag{4}$$

ρ mg/L

1.6 ThO_2

N_2

pH 4.01 6.86 9.18 3 pH pH

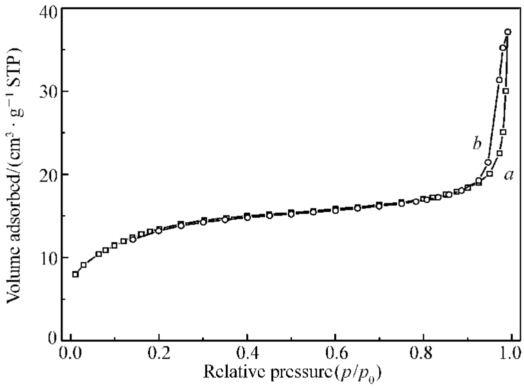
25 ℃ pH 6 ~ 8 −52 mV/pH

ThO_2 ThO_2

N_2 - 2 ThO_2 48.9 m^2/g

[7] 60.3 m^2/g ThO_2 100 ~

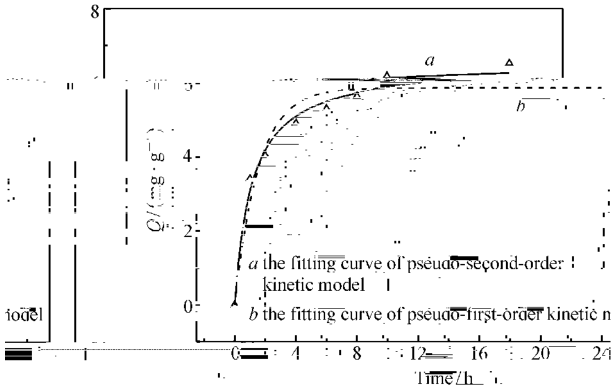
300 nm ~30 nm



2 ThO_2 N_2 -

Fig. 2 N_2 adsorption desorption isotherm of ThO_2 nanospheres

. N_2 adsorption . N_2 desorption



3 ThO_2

Fig. 3 Adsorption kinetics curves of uranium adsorption by ThO_2 nanospheres

ThO_2 = 30 mg volume of solution for adsorption 10 mL

ρ Uranium = 20 mg/L NH_4HCO_3 = 2 mmol/L

Tris-HCl = 20 mmol/L pH = 8.45 temperature = 25 ℃

2.2

3 ThO_2 NH_4 4 UO_2 CO_3 3

3 24 h

[13-14] 5 6

e 1 1 5

$$\frac{\quad 2 \quad e^{2}}{1 \quad 2 \quad e}$$ 6

e mg/g mg/g h 1 h^{-1} 2 mg/ g·h

06857f 0 Tr 10.285714 0 0 10.285714 0 -8.742857 Tm 37.2586541.7 (5 T9.11701)Tj3 1 Tf 0 Tr 21.250286 0 0 21.250286 0 -8.742857 Tm 15.997634 TD415598.06502â
(â)Tj /F3 1 Tf 0 Tr 21.250286 0 0 21.250286 0 -8.742857 Tm 17.362953 181D (98.06502â)Tj 0.305943 0 196(_)Tj 0. 0.502293 0 0 1 Tf 0 Tr 10.525125 0 .741792 TD (06857f 0 Tr 10

6.54 mg/g $_{e\ exp}$ 6.52 mg/g ThO_2 [15]

2.3

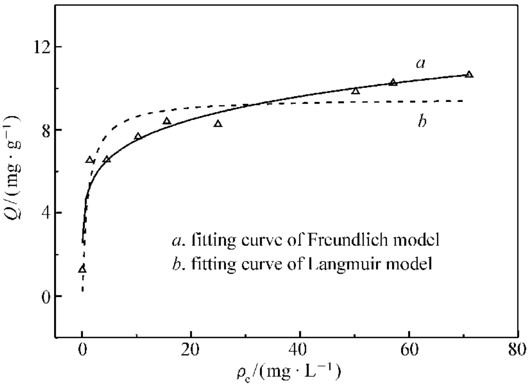
Langmuir (7) Freundlich (8) [16]

$$_e \quad \frac{{}_m \rho_e}{\rho_e \quad 1} \qquad (7)$$

$$_e \quad {}_f \rho_e^{1} \qquad (8)$$

$_m$ mg/g $_e$ mg/g ρ_e mg/L L/mg Langmuir $_f$ mg·g^{-1}· mg^{-1}·L 1/ Freundlich

4 ThO_2 NH_4 4 UO_2 CO_3 3 ρ_e Langmuir Freundlich ^{2} 2 Freundlich ^{2} Langmuir Freundlich ThO_2 Freundlich



4 ThO_2

Fig. 4 Adsorption isotherms of uranium adsorption by ThO_2 nanospheres

ThO_2 = 30 mg $_{solution}$ = 10 mL NH_4HCO_3 = 2 mmol/L Tris-HCl = 20 mmol/L pH = 8.45 adsorption time = 24 h temperature = 25 ℃

2 Langmuir Freundlich ThO_2

Table 2 Fitting results of the Langmuir and Freundlich models

Langmuir parameters			Freundlich parameters		
/ L·mg^{-1}	$_{max}$/ mg·g^{-1}	^{2}	$_f$/ mg·g^{-1}· mg^{-1}·L 1/		^{2}
0.97 ± 0.45	9.53 ± 0.54	0.854	4.97 ± 0.40	5.59 ± 0.74	0.934

2.4

ThO_2

3 ThO_2

Table 3 Desorption parameters of ThO_2 nanospheres after uranium adsorption

HCl / mol·L^{-1}	Time/h	/%	ρ_{Th}/ mg·L^{-1}
0.01	4	73.9	1.05
	24	81.6	0.80
0.02	8	77.6	1.77
	24	77.4	2.50
0.03	8	79.4	1.10
	24	81.0	1.39
0.04	8	78.9	1.03
	24	78.0	1.20
0.05	8	81.3	3.51
	24	81.7	3.07

20 mg/L ThO$_2$ 3
HCl mol/L h ρ_{Th} mg/L

ThO$_2$
1.29 mg/L ThO$_2$
3 0.01 mol/L

0.05 mol/L 8 h
ThO$_2$ Th
0.01 mol/L 24 h

2.5

$UO_2(CO_3)_3^{4-}$ UO_2^{2+}
Na^+ Mg^{2+} Ca^{2+} Sr^{2+} Cu^{2+} Ni^{2+} $UO_2(CO_3)_3^{4-}$
Cl^- Br^- NO_3^- SO_4^{2-} ThO$_2$ $(NH_4)_4UO_2(CO_3)_3$
ThO$_2$ ThO$_2$
d 4

4 ThO$_2$

Table 4 Selective adsorption properties of ThO$_2$ nanospheres

	K_{dU}/(mL·g^{-1})	K_{dM}/(mL·g^{-1})	$S_{U/M}$
Na^+	(3.68 ± 0.58) × 10^3	0	—
Mg^{2+}	(1.29 ± 0.12) × 10^3	(3.56 ± 0.07) × 10^1	36.12 ± 2.73
Ca^{2+}	(4.18 ± 0.06) × 10^2	(2.56 ± 0.15) × 10^2	1.64 ± 0.10
Ni^{2+}	(3.98 ± 0.16) × 10^2	(1.66 ± 0.02) × 10^2	2.40 ± 0.07
Cu^{2+}	(4.05 ± 0.16) × 10^2	(3.81 ± 0.07) × 10^2	1.06 ± 0.03

$(NH_4)_4UO_2(CO_3)_3$ Na^+ Mg^{2+} ThO$_2$
Ca^{2+} Cu^{2+} Ni^{2+} ThO$_2$
1
ThO$_2$
ThO$_2$ UO_2^{2+}

2.6

ThO$_2$ pH
ThO$_2$ 8.5 ~ 11 [17] ThO$_2$
ThO$_2$ pH pH
ThO$_2$ ThO$_2$
ThO$_2$ Wang [7] ThO$_2$
ThO$_2$ Mishra [8] ThO$_2$
pH > 11 Ba^{2+} Sr^{2+}

pH 8.45 ThO$_2$
Park [18]
2 mmol/L NH_4HCO_3 20 mmol/L
Tris-HCl 8.87 × 10^{-3} mol/L
 ThO$_2$

ThO_2

ThO_2 ThO_2 2 mmol/L NH_4HCO_3

20 mmol/L Tris-HCl pH

ThO_2 Na^+ NO_3^- 8.87×10^{-3} mol/L

$NaNO_3$

ThO_2 UO_2^{2+} Ca^{2+} Cu^{2+} Ni^{2+}

材 (Supporting Information) [ThO_2 XRD] 本 (http://yyhx.ciac.jl.cn/) 。

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Adsorption Performance of Thorium Dioxide Nanospheres Towards Uranium in the Aqueous Solution of Ammonium Uranyl Tricarbonate

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100871

Abstract Thorium dioxide ThO_2 a kind of actinide metal oxides has good performance as adsorbent but it is necessary to extend their application scope. In the present work we investigated the adsorption property of ThO_2 nanospheres prepared by hydrothermal method in the aqueous solution of ammonium uranyl tricarbonate. In the presence of 2 mmol/L NH_4HCO_3 and 20 mmol/L tris hydroxymethyl aminomethane -HCl ThO_2 nanospheres had an adsorption capacity of 6.52 mg/g when the initial concentration of uranium was 20 mg/L which followed pseudo-second-order kinetic model well. In isothermal adsorption studies Freundlich model was preferable. The desorption of ThO_2 nanospheres after adsorption was achieved easily by using dilute HCl solution. In the mechanism research it was found that ThO_2 nanospheres have negative charges under the experiment conditions and adsorb UO_2^{2+} cation adsorption mechanism. However the uranium uptake was seriously affected by other cations such as Ca^{2+} Cu^{2+} and Ni^{2+}. This work will contribute to the recovery of uranium from seawater and alkaline/neutral radioactive waste by metal oxides.

Keywords thorium dioxide uranium adsorption ammonium uranyl tricarbonate

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