The Removal of Sr(II) by a Nanoparticles ZnO Prepared by Microwave-Assisted Ignition Reaction

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Abstract

Adsorptive behavior of nanoparticles ZnO was assessed for removal of Sr(II) in aqueous media in the different trial circumstance such as the effect of parameters (time, pH, concentration of Sr(II) and temperature) on sorption process. The capability of nanoparticles ZnO to remove Sr(II) from aqueous media was followed by a series of adsorption isotherms (Langmuir, Freunlinch and Temkin). The sorption percent and distribution coefficient for nanoparticles ZnO in ideal circumstances are % 96.5 ± 1.76; 6959 $mL.g^{-1}$ for Sr(II), respectively. According to the Langmuir adsorption model, the maximum adsorption capacity of nanoparticles ZnO for Sr(II) were found to be 1396 $mg.g^{-1}$ and sorption kinetics was preferable explained by the pseudo-second order equation. Thermodynamic data were identified. The results have been showed that nanoparticles ZnO was appropriate as adsorbent materiel for recovery and sorption of Sr(II) ions from aqueous media. It was found that synthesized ZnO powders had high capacity to recovery Sr(II) of the aqueous media as adsorbent.

Keywords— Microwave-assisted medhod, Nanoparticles ZnO, Sorption, Sr(II), Thermodynamic

1 Introduction

Many activities dealing with radioactive materials produce of radioactive wastes which require advanced treatment [1]. The removal of heavy metals and nuclear wastes in waste waters in terms of environmental protection is an important problem [2]. These heavy metals come in sight from some industrial production such as mining, nuclear and laboratory activities [3]. Strontium is an important nuclear fission product, contained in the radioactive waste of liquid effluents [4].

Sr does not cause a significant health hazards at the stable forms while Sr-90 is not a stable isotope. The presence of Sr-90, even with low concentration in the aqueous effluents represents a serious concern. This is due to its long period of half-life (30 years) and to its great solubility in the aqueous system. Thus, Sr-90 emitting beta particles is referred to as a ''bone seeker'' and exposure to it will increase the risk for several diseases including bone cancer [4,5].

Adsorption is a well-known, a yielding and convenient method to recover metals from waste product [6-8]. Because, it has a cheap, efficient, cheap and no chemical reagents needed for industrial and radioactive waste [9].

Nowadays the use of some natural and synthetic adsorbent such as: modified clays [9,10], montmorillonite–iron oxide composite [11], hexagonal tungsten oxide [12], MnO_2–ZrO_2 composite [13], alumina [14], activated carbons [15], bentonite [16], carbon nanotubes [17], and nanocomposite [18]

have been tested for sorption of Sr(II) from waste waters.

Todays, one of the most significant developments in material science is nanotechnology. The word of nanomaterial is used to accentuate nanoparticles structures in the nanometer size range (1–100 nm) [19]. Nanoparticles are much more active than larger one because of their higher surface area and they show inimitable physical and chemical properties [20].

Nanomaterials have got the features of big surface area, high specificity, extreme reactivity potential for self assembly, high adsorption capacity for water treatment applications [19,21]. Nano metal oxides are immensely active and it is significant that both pollution control and chemical synthesis. Besides, for industrial applications, zinc oxide (ZnO) is one of foremost used materials [22].

In surveys conducted in the last years, nanoparticles ZnO have been used by the sorption works [20,23,24]. Also ZnO as an adsorbent, was commonly used for adsorption of lead [25]. Additionally, as far as we know, Sr(II) sorption properties of the nanoparticles ZnO has not been reported so far.

It is shown in our previous study [26] that uranium (VI) adsorption percent for nanoparticles ZnO powders were 98.65 % ±1.05. The purpose of study was to study the high efficiency of nano- ZnO for remove of Sr(II). Influence of contact time, pH, concentration of Sr (II) and temperature on sorption method were investigated. Balance adsorption isotherms were examined. It was calculated the values of thermodynamics of the process. The solutions of Sr(II) (1 $g.L^{-1}$) were made by dissolving of metal salts in milipore water. The entire chemicals used were of A.R. grade. In the said study, the crystallite structure accuracy and definition of particle size were 52 nm characterizations of the nano-ZnO which prepared with microwave-assisted medhod [26].

2 Experimental

It was necessary to optimize the experimental parameters such as contact time, pH, initial concentrations of strontium (II) and temperature in order to obtain maximum recovery of Sr(II) from aqueous media. Deionized water, was used to prepare solutions and all samples. To evoluate sorption of Sr(II) on nanoparticles ZnO, 0.1 g of sorbent was equilibrated with 20 mL of metal solutions in 50 mL centrifuge tube. Adsorption of metal ions on nanoparticles ZnO was done using by the bacth method on the shaker (GFL 1083). For the Sr(II) concentration analysis, inductively coupled plasma-optic emission spectroscopy (ICP-OES, Perkin-Elmer opt. 2000) was used.

The amount of Sr(II) sorbed on nano-ZnO was calculated at equilibrium (q_e) and at time (q_t) by the formula .(2.1).

$$q_{e,t} = (C_o - C_e)x(V/m)\ (\mathrm{mg.g^{-1}})\ (2.1)$$

C_o is the early Sr(II) concentration ($mg.L^{-1}$), C_e is the amount of metal present in the media at equilibrium time t ($mg.L^{-1}$), m (g) is the dry weight of sample, V (L) is the volume of metal ion concentration.

The sorption isotherms were examined via equilibrating 0.1 g of ZnO nanoparticles by varying the concentrations of metals between 25 $mg.L^{-1}$ and 125 $mg.L^{-1}$.

The sorption activity is defined with the ions distribution coefficient, K_d ($mL.g^{-1}$), determined using with formula (2.2).

$$K_d = \frac{(C_o - C_e)}{C_e} x \frac{V}{m} \quad (2.2)$$

The sorption strontium (II) ions signify in a terms of sorption ratio (Ads %) was found from by the formula (2.3).

$$\%Ads = \frac{C_o - C_e}{C_o} x100 \ (2.3)$$

The influence of temperature on Strontium (II) sorption was investigated between 293 K and 313 K. The thermodynamic parameters (ΔH^o, ΔS^o and ΔG^o) were calculated. This synthesis method of the sorbent and mechanism of sorption was seen in Fig.1.

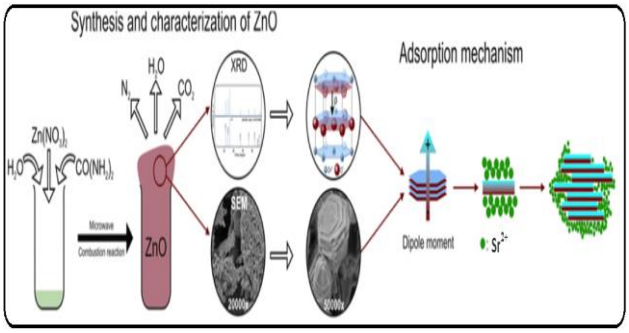

Fig. 1 The synthesis method and mechanism of the sorption.

3 Results and Discussion

3.1 The Effect of Time

Influence of contact time was investigated using a fixed concentration of strontium solutions at 293 K. The99 time dependence of Sr(II) sorption experiments was shown in Fig.2.

The sorption of Sr(II) has been examined onto nano-ZnO as a function of time in the range of (15 to 120) min. The Sr(II) absorption efficiency is 82% at 60 min. After that the absorption efficiencies become constant (Fig. 2).

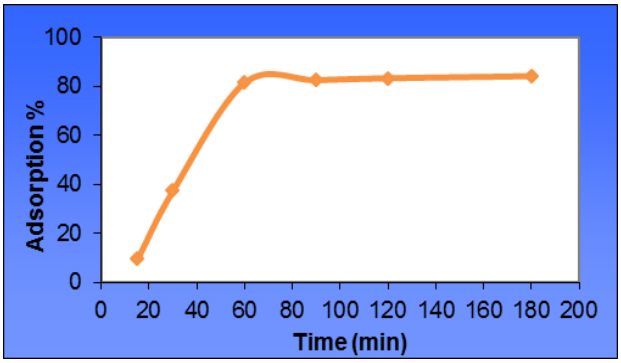

Fig. 2 Effect of contact time (m: 0.1 g, V: 25 mL, pH 3.5, 20°C, Const: 50 mg.L^{-1}

3.2 Effect of Solution pH

Influence of pH on the relative sorption of Sr(II) by the ZnO nanoparticles was shown in Fig.3.

The pH of sorption media has a complex phenomenonand strong influence on the ionic state of metal ions on the sorbent surfaces. The different metal ion species can occur the metallic cations in solution as a function of pH and metal ions [27]. The influence of pH on the sorption of strontium (II) on nano-ZnO was investigated (Fig.2). The sorption of Sr(II) depending on pH value increased. Under the optimum adsorption conditions, pH was found to be 7 for Sr(II). Similar tendency for sorption of Sr(II) has been found by other researchers [17, 28].

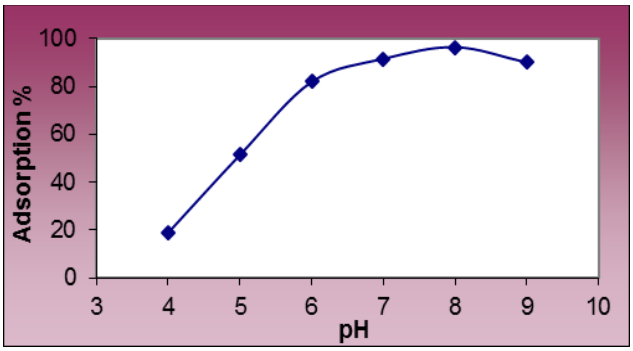

Fig. 3 Effect of solution pH. (m: 0,1 g, V:25 mL, Const: 50 mgL^{-1} , 20 °C, time: 4h)

3.3 Isotherm Studies

The aim of the sorption isotherms (Langmuir, Freundlich and Temkin) is to concern the adsorbate concentration in the media and the sorbent amount at the interface. The isotherm models are commonly used to describe the equilibrium sorption. To assess the applicability of sorption processes, the different initial metal concentrations (25-125 mg.L^{-1}) were used.

In our work, Langmuir, Freundlich and Temkin were used to analyze the data. The Langmuir equation;

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \quad (3.1)$$

Where K_L (Lmg^{-1}) is the Langmuir constant relative to the free energy of sorption, q_m (mgg^{-1}) is the max. monolayer adsorption capacity, plot of C_e/q_e versus C_e yields a straight line with slope $1/q_m$ and intercept $1/q_mK_L$.

Freundlich equivalence was depended on sorption on a heterogeneous surface. The equation is represented by:

$$q_e = k_f C_e^{1/n} \quad (3.2)$$

Eq.(2.6) can be converted into another linear form:

$$\log q_e = \log k_f + 1/n \log C_e \quad (3.3)$$

Where k_f is the constant of sorption capacity and 1/n is the constant of sorption intensity for the Freundlich. Temkin equation stating the isotherm is given as follows:

$$q_e = B \ln A_T + B \ln C_e \quad (3.4)$$

B is the constant of adsorption heat (J.mol^{-1}) and A_T is equilibrium binding coefficient (L.g^{-1}). Binding energy is carried out by plotting the quantity sorbed q_e against lnC_e and the factors were calculated with the slope and intercept.

It is sorption isotherms of Sr(II) from aqueous media on to ZnO nanoparticles at a different temperature were shown in Fig.4.

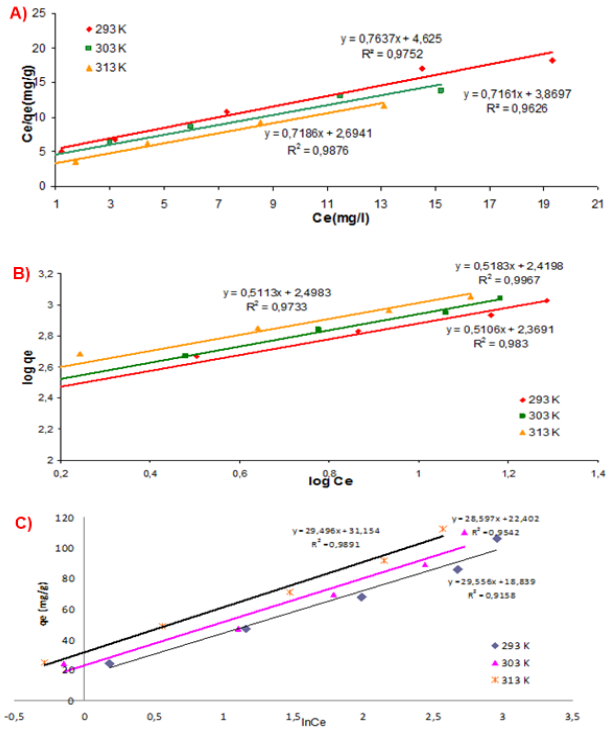

Fig. 4 Langmuir (A), Freundlich (B) and Temkin (C) plots for sorption of Sr (II) at different temperature.

It was shown Langmuir, Freundlich and Temkin constants, R was calculated according to Eqs.(3.1 – 3.4) and were given in the Table 2. Maximum adsorption capacity for Langmuir was found 1396 mgg^{-1}. It is recommended that the ZnO nanoparticles under consideration can be effectively used for Sr(II) sorption from aqueous media.

Essentially, nanoprous ZnO exhibits very sorption performances for metal ions in comparisons with other adsorpbents given in literature and listed in Table 1. Analyzing the data, the adsorbent used in this study is found to give better results.

Table 2 Isotherm parameters for Strontium (II) sorption onto the nano- ZnO at 303 K.

Langmuir		
q_m(mg g^{-1})	K_L(L mg^{-1})	R^2
1,396	0,185	0.9626
Freundlich		
K_F(mg g^{-1})	n(g L^{-1})	R^2
11,244	1,93	0.9967
Temkin		
B(J mol^{-1})	A_T(L g^{-1})	R^2
29.496	85.406	0.9891

Table 1 The comparison of adsorption capacity of nanoporous ZnO with various adsorbent reported in the literature.

Ions	**Adsorbent**	**Max.Ads. Capacity (mg g^{-1})**	**pH**	**Ref.**
U(VI)	Carbonfromusedtires	226.1	3.5	[29]
Sr(II)	Hexagonal tungsten oxide	20.5	4	[12]
Sr(II)	Montmorillonite–ironoxidecomp	55.5	6	[11]
U(VI)	CS/CPL composite	536.55	4	[30]
Th(IV)	CS/CPL composite	438.55	5.5	[30]
Th(IV)	Monazite	1,666	4	[31]
Th(IV)	Titanatenanotubes	232.56	3	[32]
Th(IV)	Grapheneoxide	411	3	[33]
A.Dye	Nano-Adsorbent	1883	3	[34]
U(VI)	Nanoporpus ZnO	1111	5	[26]
Sr(II)	NanoporousZnO	1,396	7	This study

Freundlich isotherm parameters fits for Sr(II) sorption on nanoparticles ZnO yielded isotherms that are in a good conformity of detected conduct (R^2 ≥0.99 for Sr) (Table 2).

Values of K_f and n for Sr(II) were found to be 11.244 $mg.g^{-1}$ and 1.93; respectively. The Freundlich adsorption coefficient, Kf, was raised by temperature, so the sorption process was endothermic [35].

On the Temkin plot seen in Figure 4, values of A_T and B for Sr(II) were found to be: 85.406 Lg^{-1}, 29.496 $Jmol^{-1}$, respectively, It is indicative of the heat of sorption define a physical sorption process.

3.4 Kinetics of Adsorption

To investigate the controlling mechanism of sorption of Sr(II) ions onto the nano-ZnO, pseudo-first and pseudo-second order kinetic equations were studied for different time intervals. The only best-fitted plot was shown in Fig. 5.

The mathematical equations were used and the plots were improved the formula:

$$t/q_t = 1/K_2 q_{e^2} + t/q_e \quad (3.5)$$

Where q_t and q_e ($mg.g^{-1}$) are the amount of Sr(II) adsorbed at time t and equilibrium time, respectively; K_2 ($mg.g^{-1}$.min): reaction rate constant.

The related kinetic parameters have been appraised from the slopes and intercept of kinetic equations.

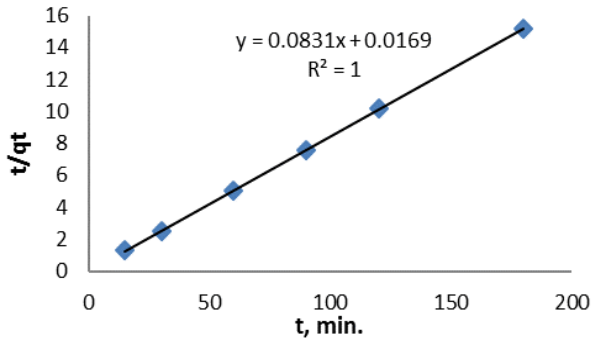

Fig. 5 The pseudo-second order adsorption kinetics of Sr (II) onto the nanoporous ZnO.

The results founded from the analysis of this data have been showed that Sr(II) ion sorptions on ZnO have been defined by kinetic equation (Table 3).

Table 3 Kinetic parameters of Sr(II) sorption onto the by nanoporous-ZnO

Kinetic models	**Parameters**	
Pseudo-second order	R^2	1.0
$t/q_t = 1 / K_2 q_e^2 + t/q_e$	K_2 (mgg^{-1} min^{-1})	0.44
	q_e (mgg^{-1})	12.81

3.5 Adsorption Thermodynamics

The sorption capacity of ZnO nanoparticles spontaneously has increased with the temperature from 293 to 313 K.

In thermodynamics, the enthalpy change (ΔH^o) and entropy change (ΔS^o) were determined according to the Van't Hoff formula:

$$\ln K_d = (\Delta S^o / R) - (\Delta H^o / RT) \quad (3.6)$$

Where lnKd is the distribution coefficient ($mL.g^{-1}$), ''T'' is the temperature (K), "R" is gas constant ($kJmol^{-1}K^{-1}$). The free energy values (ΔG^o) were found by the formula:

$$\Delta G^o = \Delta H^o - T\Delta S^o \quad (3.7)$$

The experiments were carried out at different temperatures (293, 303, 313 K) and constant concentration (25 $mg.L^{-1}$ of Sr(II). The distribution coefficient (K_d) values raised with temperature indicating the endothermic nature of the adsorption. ΔH^o and ΔS^o were obtained from the slopes and intercepts of lnK versus 1/T (R^2: 0.96 for Sr(II)).

Table 4 Thermodynamic parameters for strontium sorption by nanoporous-ZnO

Metal Ions	**ΔH^o ($kjmol^{-1}$)**	**ΔS^o ($jK^{-1}mol^{-1}$)**	**ΔG^o ($kjmol^{-1}$)**		
			293 K	**303 K**	**313 K**
Sr(II)	18.4	139.68	-40.91	-42.30	-43.70

The positive value of ΔH^o and entropy (ΔS^o) is suggestive of an endothermic nature which is favored and higher randomness of sorption in the system and favors the stability of the at higher temperature. [26]. As shown in Table 4, with increasing temperature, temperatures on nanoparticles ZnO occur more efficient sorption of Sr(II) ions at elevated.

4 Conclusion

This works, the adsorption of strontium (II) ions from aqueous media with ZnO nanoparticles as a function of the pH, times, metal ions and temperature were an exemined.

• The monomolecular sorption capacity of nanoparticles at optimum condition was 1,396 mgg^{-1} for Sr (II) ions.

•According to Freundlich, values of K_f and n for Sr(II) were obtained to be 11.244 $mg.g^{-1}$ and 1.93, respectively.

•Sorption for Sr(II) ions on ZnO nanoparticles is physical-chemistry sorption because ΔH^o was 18.4 $kJ.mol^{-1}$. The percent sorption (%) and K_d for this nanoparticles at 313 K were 97.9 % and 8017 $mL.g^{-1}$, respectively.

• Additional, sorption of Sr (II) studies in the mixed Cu^{2+}, Pb^{2+} and Zn^{2+} ions on nanoparticles have been investigated in acidic media. (Consantration of Sr(II): 100 ppm, Time: 60 min.; pH: 4; Mixed ions const: 25 ppm; 303 K). Sorption values in the mixed metal ions for Sr(II) ions was 62 %. (pH 4).

• This Works, prepared ZnO nanoparticles materials are used as powerful adsorbent materials for Sr(II) ions remove for aqueous media. The data obtained from this study will not only contribute to the scientific literature, but also they will be very important in terms of the removal of the dangerous radionuclides from the environment.

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