

SORPTION REMOVAL OF Sr^{2+} AND Y^{3+} IONS FROM AQUEOUS SOLUTIONS BY A TiO_2 -BASED SORBENT

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Abstract. In present investigations, the sorption removal of Sr^{2+} and Y^{3+} ions from aqueous solutions by TiO_2 – La sorbent was researched. The dependence of the sorption process on the time of interaction, solution's acidity, and initial concentrations of the Sr^{2+} and Y^{3+} ions were investigated. Four simplified kinetic models – Lagergren's pseudo-first and pseudo-second order kinetic models, Intra-particle diffusion and Elovich models – were tested to describe the adsorption process. Equilibrium isotherms data were analyzed using Langmuir and Freundlich isotherm models. The kinetic data indicated that the adsorption of Sr^{2+} and Y^{3+} ions by TiO_2 –La fitted well with the pseudo-second order kinetic model with coefficients of linear approximation ($R^2 = 0.99$) for both elements. The application of the Elovich model to the experimental data of the sorption of Sr^{2+} and Y^{3+} ions by TiO_2 –La shows that yttrium is absorbed due to the mechanism of chemisorption. Coefficient of linear approximation $R^2 = 0.98$. Strontium is absorbed via physical sorption or ion exchange mechanism. The maximum adsorption capacity of TiO_2 –La was found to be $0,9 \text{ mmol}\cdot\text{g}^{-1}$ (79 mg g^{-1}) for Sr^{2+} and $1.6 \text{ mmol}\cdot\text{g}^{-1}$ ($134 \text{ mg}\cdot\text{g}^{-1}$) for Y^{3+} .

Key words: Titanium dioxide, strontium, yttrium, sorption, water solutions, kinetic models

1. INTRODUCTION

The investigation of the amount and migration of strontium ions in the environment is very important. Therefore, considerable attention of scientists is devoted to the sorption of strontium ions from aqueous solutions [1-3]. Much less research is devoted to sorption removal of yttrium ions – daughter radionuclide of $^{89-92}\text{Sr}$ in β^- - decay chain (Fig. 1).

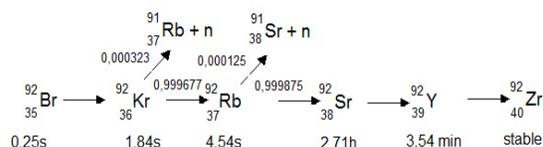


Figure 1. β^- decay chain with $A_r = 92$.
Daughter isotope of ^{92}Y – formed after ^{92}Sr decay.

Natural yttrium consists of one stable isotope – ^{89}Y . Heavy radioactive isotopes of yttrium obtained in fission reactions of uranium nuclei and can be found in the emission of nuclear power plants. Radioactive isotopes of yttrium - ^{90}Y and ^{91}Y are use in medicine for the treatment of cancer neoplasms on the surface of the body, skull or pituitary glands.

In our opinion, the investigations of the sorption of strontium and yttrium when simultaneously present in

water solution is interesting as well as research of the mechanism of interaction between strontium or yttrium and the surface of sorbents. The study of the sorption of Sr^{2+} and Y^{3+} ions will help to divide effectively of $^{89-92}\text{Sr}$ and $^{90,91}\text{Y}$.

As a rule, for sorption removal of yttrium ions calcite, ferrite or some metal hydroxides are offered. On the other hand, titanium dioxide is a unique compound that has sorption ability in relation to strontium ions.

In this work, TiO_2 was used as a sorbent. TiO_2 was synthesized in Vasyl Stefanyk Precarpatian National University (Ukraine) by an original technique. The difference of the sample that was used in our research from the samples of TiO_2 previously described in [4, 5] was that lanthanum was introduced in its structure. It was supposed, that having lanthanum in the structure increases the selectivity of the sorbent to the yttrium ions and probably other rare earth elements. For the synthesis of TiO_2 -La, the reaction of liquid-phase hydrolysis of TiCl_4 was used. The reaction mixture was cooled. At the pH of the reaction mixture 3.0-8.0 ellipsoidal particles with anatase structure are formed. When the concentrated dispersions are dried, aggregates in the size of 10-30 nm are organized. The aqueous solution of $\text{La}(\text{NO}_3)_3$ was introduced in the reaction mixture in the step of hydrolysis so that in the final product its content was 2-4%. With the aid of a fluorescent X-ray analyzer, the composition of the sorbent formed was determined as well as yttrium and

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strontium presence on the surface of the sorbent after the sorption process.

Preliminary studies of the distribution coefficients (K_d) of Sr²⁺ ions from aqueous solutions by TiO₂-La showed satisfactory values of K_d in a neutral medium, a quite high value of K_d from the acidic medium and a very high K_d in an alkali medium (Table 1). Distribution coefficients of Sr²⁺ ions in an alkali medium indicate the perspective of a study of the sorption of Sr²⁺ and Y³⁺ ions onto TiO₂-La - sorbent.

Table 1. Distribution coefficients of Sr²⁺ ions onto TiO₂-La

Acidity of solution	pH=1	pH=4	pH=7	pH≥10	Rate of Sr ²⁺ withdrawn at pH≥10, %
$K_d, \text{ ml} \cdot \text{g}^{-1}$	1300	1300	154	19900	99.5
	$1.3 \cdot 10^3$	$1.3 \cdot 10^3$	$0.15 \cdot 10^3$	$1.99 \cdot 10^4$	

In the present investigations, the sorption removal of Sr²⁺ and Y³⁺ ions from aqueous solutions by TiO₂-La sorbent was researched. The dependence of the sorption process on time of interaction between the solution and the surface of sorbent was investigated. Four simplified kinetic models: Lagergren's pseudo-first and pseudo-second order kinetic models, Intra-particle diffusion and Elovich models were tested to describe the adsorption process. The adsorption of Sr²⁺ and Y³⁺ ions has been investigated as a function of the solutions acidity, and the initial concentrations of the Sr²⁺ and Y³⁺ ions. Equilibrium isotherms data has been analyzed using Langmuir and Freundlich isotherm models. As well, we investigated the sorption of strontium and yttrium with their simultaneous presence in water solution.

2. MATERIALS AND METHODS

Experimental studies of strontium and yttrium removal from aqueous solutions was carried out using the stable isotopes of ⁸⁸Sr and ⁸⁹Y in the form of SrCl₂·6H₂O or YCl₃·6H₂O.

To study the effect of pH, agitation time and equilibrium concentration of Sr²⁺ or Y³⁺ (isotherms of adsorption), we performed batch studies at the room temperature of 18°C. The mass of sorbent was 50 mg, volume of solution – 5 ml, times intervals from 5 to 150 min with performing the intermittent shaking; interval of initial concentrations of Sr²⁺ or Y³⁺ ions was 0.0005 – 0.1 mol·L⁻¹. The mixture was removed at a specified interval of time, and residual concentration of Sr²⁺ and Y³⁺ was analyzed using complexometric titration [6, 7]. For Y³⁺, spectrophotometric analysis was also used [8]. The amount of adsorbed Sr²⁺ or Y³⁺ was calculated using the concentration in solutions before and after the adsorption process.

The value of the distribution coefficients (K_d) was measured in an acidic, neutral and alkali medium in batch studies. The volume of the solution was 20 ml, the mass of sorbent 50 mg⁻¹; the initial concentration of Sr²⁺ - ions was $5 \cdot 10^{-4}$ mol·L⁻¹, the time interval of

sorption no less than 100 min under intermittent shaking.

The effect of the pH of the solutions was controlled by adding a 4 ml 10% HNO₃ or 5ml 10% NH₄OH, using a pH – meter pH-150M «Bilorus' 2003» (Belarus).

The sorption and distribution coefficients were measured using equations (1), (2) from literature [2]

$$A = (C_i - C_e) / m A_r \text{ [mmol} \cdot \text{g}^{-1}] \quad (1)$$

$$K_d = (C_i - C_e) / C_e (V/m), \text{ [ml} \cdot \text{g}^{-1}] \quad (2)$$

C_i - initial ion's concentration, *mg*,

C_e - equilibrium concentration, *mg*; *m* – mass of sorbent, *g*; *V* - solution's volume, *ml*.

For the studies of the sorption of Sr²⁺ and Y³⁺ ions from their mixture, a solution of their chlorides with a concentration of 0.88 mg·ml⁻¹ of Sr²⁺ and 0.89 mg·ml⁻¹ of Y³⁺ was prepared. 5 ml of such mixture interacted with 50 mg TiO₂ – La under intermittent shaking, during 40 minutes. After sorption, the solution was removed from the sorbent. The residual concentration of Sr²⁺ was determined by titration with Na-ed, and that of Y³⁺ by photometric analysis.

3. RESULTS AND DISCUSSION

The dependence of the sorption process on the time of interaction between Sr²⁺ and/or Y³⁺ ions and the surface of TiO₂-La sorbent is shown in Figure 2.

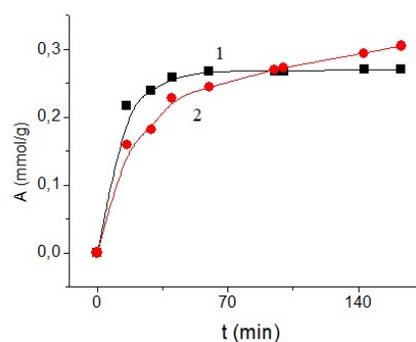


Figure 2. Kinetic of sorption of Sr²⁺ (1) and Y³⁺ (2) ions by TiO₂-La in neutral medium. The initial concentrations of Sr²⁺ and Y³⁺ was 0.01 mol·L⁻¹.

The dependence of the sorption process on time interaction is often used in determining the time at which the sorption equilibrium is established.

By this parameter we can make conclusions about the mechanism of sorption, is it physical or chemisorption. If the sorption process is chemisorption, the experimental results may be described by the Elovich model. In according to this model under a large time of interaction dependence between ability of adsorption ($A, \text{ mg/g}$) and $\ln t$ must be linear [9, 10].

In this work, in order to analyze experimental data, four kinetic adsorption models were applied: Lagergren's pseudo-first and pseudo-second order kinetic models, Intra-particle diffusion and Elovich

models. Correspond equations are described in literature [11-16].

The kinetics of sorption of Sr²⁺ and Y³⁺ ions in the Elovich and Lagergren coordinates are shown in Figures 3, 4.

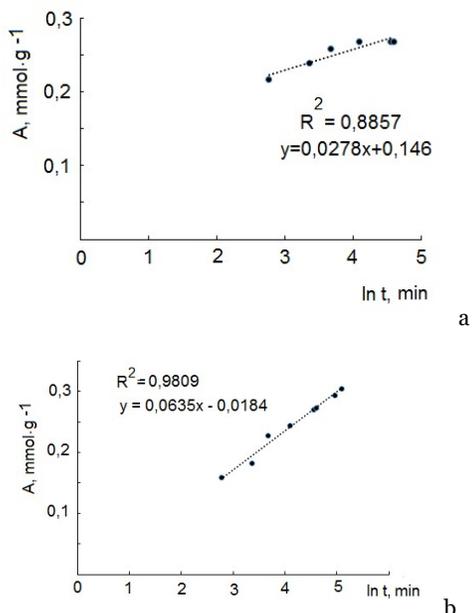


Figure 3. Using the Elovich model to experimental data of sorption of Sr²⁺ (a) and Y³⁺ (b) ions by TiO₂–La in neutral medium. The initial concentrations of Sr²⁺ and Y³⁺ is 0.01 mol·L⁻¹.

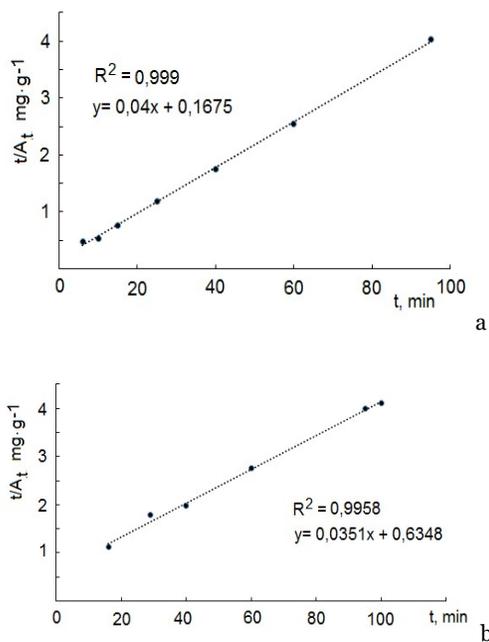


Figure 4. Using the Lagergren pseudo-second kinetic model on the experimental data of the sorption of Sr²⁺ (a) and Y³⁺ (b) ions by TiO₂–La in neutral medium. The initial concentrations of Sr²⁺ and Y³⁺ was 0.01 mol·L⁻¹

Table 2. The application of the kinetics models on the experimental data of the sorption of Sr²⁺ and Y³⁺ ions by TiO₂–La

Kinetic model	Y ³⁺	R ²	Sr ²⁺	R ²
Elovich	$A_t = 0.064 \ln t - 0.018$	0.98	$A_t = 0.0278 \ln t + 0.15$	0.88
Diffusion	$A_t = 0.019 \sqrt{t} + 0.09$	0.94	$A_t = 0.0078 \sqrt{t} + 0.19$	0.79
Pseudo first	$\log(A_e - A_t) = -0.014 t + 1.25$	0.95	$\log(A_e - A_t) = -0.031 t + 1.1$	0.95
Pseudo second	$t/A_t = 0.035 t + 0.64$	0.996	$t/A_t = 0.04 t + 0.17$	0.999

Lagergren’s coefficients for Sr²⁺ and Y³⁺ ions sorption onto TiO₂ –La were calculated. The values of Lagergren’s coefficients pseudo-first and pseudo-second order kinetics are given in the Table 3. They are measured as the arithmetic means [17] of the coefficients calculated for different values of At and t, prior to the establishment of the sorption equilibrium. However, for the sorption of yttrium onto TiO₂ –La, the dependence of k₂ on t is well described by the exponent with the correlation coefficient R² = 0.95 (Figure 5(b)). This fact indicates a smooth change of k₂ and hence the dependence of the sorption of yttrium ions on the degree of TiO₂ –La’s surface filling [18]. The sorption of Sr²⁺ ions within 0–60 minutes of interaction is independent on the degree of filling of the surface.

Table 3. Lagergren coefficients measured according to the experimental data of the sorption of yttrium and strontium by TiO₂ – La.

Y ³⁺		Sr ²⁺	
K ₁ , min ⁻¹	K ₂ , g·mg ⁻¹ ·min ⁻¹	K ₁ , min ⁻¹	K ₂ , g·mg ⁻¹ ·min ⁻¹
0.11±0.017	0.02±0.009	0.31±0.04	0.078±0.023

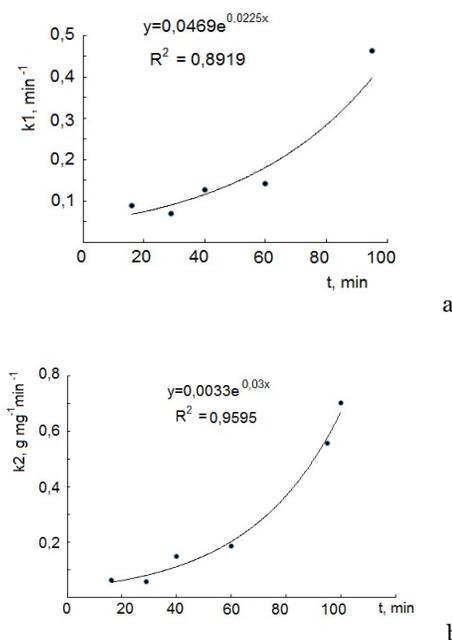


Figure 5. Lagergren’s coefficients k₁ (a) and k₂ (b) for sorption of Y³⁺ by TiO₂ –La.

The kinetic data indicated that the adsorption of Sr²⁺ and Y³⁺ ions onto TiO₂ –La fitted well with the pseudo-second order kinetic model.

The application of the Elovich model to the experimental data of the sorption of Sr²⁺ and Y³⁺ ions onto TiO₂ –La shows that yttrium is absorbed rather due to the mechanism of chemisorption (precipitation or surface complexation). Strontium is absorbed via physical sorption or an ion exchange mechanism. This information is important in terms of the desorption process. If Sr²⁺ was adsorbed by the physical sorption, then it will be easily desorbed as well. In order to desorb yttrium from the surface of the sorbent, it is necessary to use excess energy equal to the energy of desorption. So, yttrium will be strongly bound to the matrix of the TiO₂ –La. The results described in [19-21] confirm our conclusions.

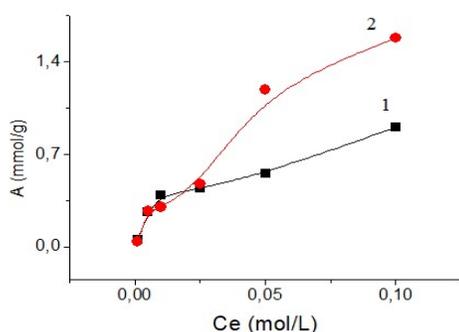


Figure 5. Adsorption isotherm of Sr²⁺ (1) and Y³⁺ (2) ions by TiO₂ –La

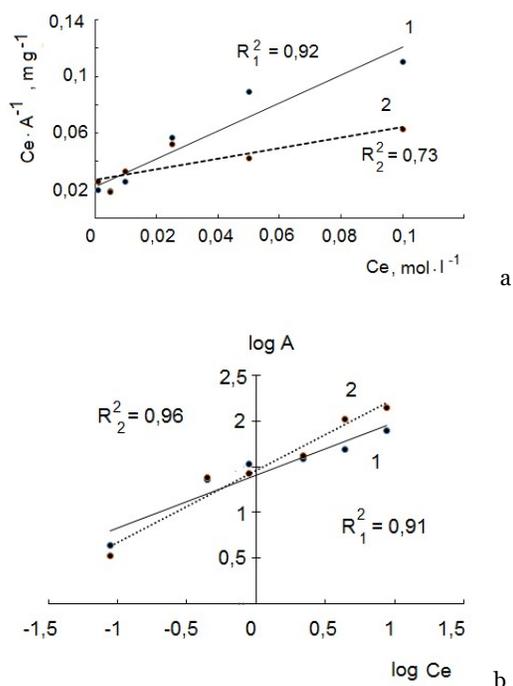


Figure 6. Adsorption isotherm of Sr²⁺ (1) and Y³⁺ (2) ions onto TiO₂ –La in Langmuir coordinate (a), according to [18]; (b) Freundlich's coordinate.

Equilibrium isotherm data for the sorption of Sr²⁺ and Y³⁺ ions are shown in Figures 5, 6. The maximum adsorption capacity of TiO₂ –La was found to be 0.9 mmol·g⁻¹ (79 mg·g⁻¹) for Sr²⁺ and 1.6 mmol·g⁻¹ (134 mg·g⁻¹) for Y³⁺ (we can compare with the results in publication [1] 1.14 mg·g⁻¹ for Sr²⁺).

Equilibrium isotherm data were analysed using Langmuir and Freundlich's isotherm models. The adsorption of Sr²⁺ ions well described by the Langmuir isotherm model. The adsorption of Y³⁺ ions – by Freundlich's isotherm model.

Table 4. Experimental data of the sorption of Sr²⁺ and Y³⁺ ions onto TiO₂ –La from their mixture in neutral medium

Initial concentration of the element mol·L ⁻¹	A, mmol·g ⁻¹ of Y ³⁺ from individual solution	A, mmol·g ⁻¹ of Sr ²⁺ from individual solution
0.01	0.294	0.268
Sr ²⁺ + Y ³⁺ (0.01)	A, mmol·g ⁻¹ from their mixture 1.034	

The results presented in Table 4 and Figures 5, 6 allow us to assume that the sorptions of Sr²⁺ and Y³⁺ ions occur by different mechanisms. However, the value of the total adsorption from their mixture is greater than the sum of the adsorption of these elements from their individual solutions. According to [19-21], the adsorption of yttrium is well correlate with the pattern for yttrium hydrolysis. Because, after pH = 9 Y³⁺ precipitates in the form of an insoluble hydroxide, at pH= 7 it will be predisposed to the reactions of complexations, colloid formation or precipitations at the interface of the phases. Herewith, it can interact with Sr²⁺, which exists in the individual solution as a cation at pH = 7, and form compounds Y²⁺ - O - Sr⁺ on the surface of the sorbent. In this case, although these two elements are adsorbed by the TiO₂ – La by different mechanisms, they will in the mixture increase the sorption of each other. However, this needs more investigate.

Such studies are very important, due to the determination of the content of Sr²⁺ in effluents or the water cooler of a nuclear power plant, there is a needlessness of its separation from yttrium and rare earth elements (REE).

The dependence of the adsorption value of Sr²⁺ and Y³⁺ ions as a function of pH is shown in Figure 7.

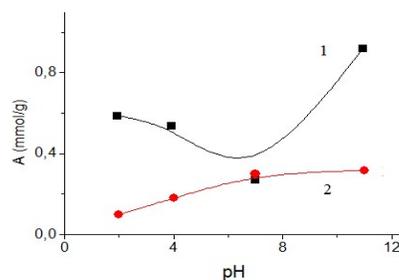


Figure 7. pH dependence of the sorption of Sr²⁺ (1) and Y³⁺ (2) ions onto TiO₂ –La. The initial concentration of the ions is 0.01 mol·L⁻¹.

The study of the dependence of Y³⁺ ions adsorption from pH requires special attention. As mentioned, yttrium exists in the form of a trivalent cation in the range of pH = 1-9. Then, according to literature data, they exist as Y(OH)^[2+] (pH=9-10.5); YO₃H₃s (pH ≥10.5). (Under conditions $\sum \gamma 10^{-10}$; 298.15K; 10⁵ Pa). According to another data in the range of pH = 7.4-10.5 – yttrium exists as Y(OH)^[2+] (pH=7.4-8.8); YO^[+] (pH=8.8-9.7); HYO₂ (aq) (pH=9.7-10.5); YO₂^[-] (pH=10.5-13) [7].

Taking into account all of the above, we can conclude that the studied sorbent best absorbs yttrium in a neutral medium in the form of cation.

5. CONCLUSIONS

1. Adsorptions of Sr²⁺ and Y³⁺ ions occur by different mechanisms onto TiO₂ –La. The kinetic data indicated that the adsorption of Sr²⁺ and Y³⁺ ions onto TiO₂ –La is well describes with the pseudo-second order kinetic model. The application of Elovich model to experimental data of sorption of Sr²⁺ and Y³⁺ ions onto TiO₂ –La shows that yttrium is absorbed rather due to the mechanism of chemisorption. Strontium is absorbed via physical sorption. The adsorption of Sr²⁺ ions is well describes by Langmuir isotherm model. The adsorption of Y³⁺ ions - by Freundlich's isotherm model. The maximum adsorption capacity of TiO₂ –La was found to be 0.9 mmol·g⁻¹ (79 mg g⁻¹) for Sr²⁺ and 1.6 mmol·g⁻¹ (134 mg·g⁻¹) for Y³⁺.

2. For the effective separation of Sr²⁺ and Y³⁺ ions, the differences in their sorption at neutral and alkali medium should be used, as well as differences in their dependence of time interaction.

3. Sorbent TiO₂ –La can be used for the sorption of Sr²⁺ and Y³⁺ ions from individual solutions and from their mixture. In this case, these two elements will increase the sorption of each other.

4. The results of this work can be used to create sorbents that can absorb both divalent Sr²⁺ and trivalent Y³⁺ and, probably, other rare earth elements. The study of yttrium removal is essential and performed. Difference in Sr²⁺ and Y³⁺ sorption behavior are interesting from the aspect of their separation in the environmental protection, and analytical competitions.

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