

## SORPTION CHARACTERISTICS OF PHOSPHATE AND THIOCYANATE ANIONS ON MULTICOMPONENT Mg/Ni-Al -TYPE HYDROTALCITES

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This paper investigated the sorption equilibrium of phosphate and thiocyanate anions from aqueous solutions on hydrotalcites with various Ni/Mg isomorphous substitutions using the memory effect property of the resulting materials. The XRD results showed that the reconstruction process is inhibited by the presence of the Ni<sup>2+</sup> cations in the HT matrix. The sorption results on the substituted Ni/Mg-HT could be associated with the external surface area developed when the samples were thermally activated at 500°C. Langmuir and Freundlich isotherms were used to interpret equilibrium data for phosphate. The investigation of Henry's domain on Langmuir isotherms indicated the decrease of phosphate sorption for hydrotalcites within Mg/Ni-Al series (from Mg<sub>3</sub>Al as a reference to Ni<sub>3</sub>Al sample) as Mg was substituted by Ni. The following selectivity order resulted: Mg<sub>3</sub>Al > Mg<sub>2</sub>NiAl > Mg<sub>1.5</sub>Ni<sub>1.5</sub>Al > MgNi<sub>2</sub>Al > Ni<sub>3</sub>Al. This sequence corresponded to that attributed for the maximum adsorption capacities, which decreased from 111 to 37 mg · g<sup>-1</sup> for Mg<sub>3</sub>Al and Ni<sub>3</sub>Al, respectively. Freundlich isotherm was used for low and medium equilibrium concentrations, where the deviation of experimental points from the Langmuir isotherm was noticeable. Compared to the reference adsorbent, Mg<sub>3</sub>Al, which was characterized by a maximum sorption capacity of 38 mg · g<sup>-1</sup> for thiocyanate, Mg/Ni-Al-type hydrotalcites rendered decreased sorption capacities (7.8–4.8 mg · g<sup>-1</sup>) as Mg was substituted by Ni. These findings underlined the negative influence of nickel within brucite interlayer on retention capacity for anions and they did not justify using Mg/Ni-Al-type hydrotalcites for thiocyanate sorption.

### INTRODUCTION

Layered Double Hydroxides (LDHs) are natural or synthetic mixed metal hydroxides with interlayer spaces containing exchangeable anions, relatively simple and cheap to synthesize on both laboratory and industrial scales. Their characteristics such as: wide chemical compositions due to variable isomorphous substitutions of metallic cations, variable layers charge density, anion adsorption/exchange properties, “memory effect”, makes them an important “intelligent” family of porous materials.<sup>1,2</sup> The most representative mineral within LDHs series is Mg<sub>6</sub>Al<sub>2</sub>(OH)<sub>16</sub>CO<sub>3</sub> · 4H<sub>2</sub>O hydrotalcite. Other minerals with similar structure are known as hydrotalcite like compounds (HTlc).

The structure of hydrotalcite-type anionic clays can be described as [M<sup>II</sup><sub>1-x</sub>M<sup>III</sup><sub>x</sub>(OH)<sub>2</sub>]<sup>x+</sup> A<sup>n-</sup><sub>x/n</sub>

· mH<sub>2</sub>O, where M<sup>II</sup> = Mg<sup>2+</sup>, Zn<sup>2+</sup>, Ni<sup>2+</sup>, etc., M<sup>III</sup> = Al<sup>3+</sup>, Fe<sup>3+</sup>, Ga<sup>3+</sup>, etc., and A<sup>n-</sup> = CO<sub>3</sub><sup>2-</sup>, Cl<sup>-</sup>, HO<sup>-</sup>, etc. They consist structurally of stacked brucite-like (M<sup>2+</sup>(OH)<sub>2</sub>) layers in which M<sup>2+</sup> ions are partially substituted by M<sup>3+</sup> ions. The substitution of M<sup>3+</sup> for M<sup>2+</sup> on the layers demands the incorporation of interlayer anions to balance the resulting positive charge. In addition to divalent and trivalent cations, a wide range of cations in monovalence or higher valence such as Li<sup>+</sup>, Sn<sup>4+</sup>, Zr<sup>4+</sup>, Ti<sup>4+</sup>, etc., may be also accommodated in the octahedral sites of the layers.<sup>3</sup> There are numerous studies on synthesis, structure and physical-chemical characteristics of these compounds.<sup>4-8</sup>

The presence of phosphate anion in surface water leads to serious problems of eutrophication, which requires the decrease of phosphate concentration to stipulated limits. A wide range of processes has addressed phosphate removal from water.<sup>9-12</sup> Thiocyanate containing wastewaters

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result from a variety of industrial processes such as: herbicide and insecticide production, acrylic fibre production, manufacturing of thiourea, electroplating etc. The removal of thiocyanates from wastewater is a must since this anion is well-known as a dangerous pollutant.

The removal of phosphates and thiocyanates by sorption on hydrotalcites might be an alternative to the conventional processes that have been applied and investigated so far.<sup>13,14</sup>

The aim of this work was to investigate the influence of Ni/Mg substitution ratio on the sorption characteristics of phosphate and thiocyanate anions on Mg/Ni-Al-type hydrotalcites.

## EXPERIMENTAL

### Synthesis and characterization of Mg/Ni-Al -type hydrotalcites

A series of hydrotalcite-type solids with various compositions were synthesized in order to obtain different Ni/Mg substitution ratios. The synthesis was carried out by following a classic procedure - the co-precipitation method.<sup>6</sup>

A solution of  $1 \text{ mol} \cdot \text{L}^{-1}$   $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  and  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  (Sigma) in distilled water was slowly added to a  $2 \times 10^{-3} \text{ M}$   $\text{Na}_2\text{CO}_3$  solution under magnetic stirring. During the synthesis the pH was kept constant between 8 and 8.5 by adding suitable amounts of 2M NaOH solution. The measurements were carried out by means of a Hanna HI 991003 pH-meter. The mixing step was carried out under stirring for 1 h at 25°C and then it continued on an oil bath at 60°C under magnetic stirring and reflux for 18 h. The hydrotalcite powders were recuperated by centrifugation and washed several times with demineralized water until pH = 7. This step was followed by drying at 60°C overnight. Then the dried samples were crushed and sieved. The fraction between 0.060 and 0.125 mm was used as adsorbent. The sample activation was performed in an oven at temperatures of 500°C in air and a rate of  $1^\circ \cdot \text{min}^{-1}$  for 4 h.

The chemical composition of the as-synthesized materials was determined using a JEOL JCXA 733 superprobe EPMA (electron probe microanalysis).

Differential scanning calorimetry (DSC) analysis was performed using an automatic thermal analyzer (Mettler Toledo DSC821e, Novate Milanese, Italy). Temperature calibrations were performed with indium as a standard. Sealed and holed aluminium pans were used in the experiments for all

the samples, and an empty pan prepared in the same way was used as a reference. Samples of 3 and 6 mg were weighted directly into the aluminium pans, and the thermal analyses were conducted at a scanning rate of 10°C/min from 20 to 1000°C.

XRD powder patterns were collected on a BRUKER D8 Advance instrument by using  $\text{CuK}\alpha$  radiation (Ni filter;  $\lambda = 0.15401 \text{ nm}$ , 40 kV and 50 mA).

### Sorption equilibrium of phosphate and thiocyanate on Mg/Ni-Al -type hydrotalcites

All sorption experiments were performed on activated Ni-Mg/Al hydrotalcite samples. To study the retention capacity at equilibrium, identical quantities of synthesized solids were contacted with identical volumes of aqueous solutions containing phosphate and thiocyanate anions. The solution pH was  $7 \pm 0.2$  and the solid/liquid ratio was  $1 \text{ g} \cdot \text{L}^{-1}$ . The samples were maintained at constant temperature ( $25 \pm 1^\circ\text{C}$ ) in a Shaker Bath thermostat to reach the sorption equilibrium for 12 h. The kinetic studies preceded the determination of the equilibrium isotherm in order to determine the time demand for setting up thermodynamic equilibrium. At equilibrium, the solid was separated by centrifugation.

The phosphate and thiocyanate concentration in solution was analysed by using a UV-VIS Varian Cary 50 spectrophotometer. Phosphate concentration in the aqueous solution was determined at 450 nm in line with the vanado-molybdate method.<sup>17</sup> The concentration of thiocyanate was also determined spectrophotometrically at 475 nm.<sup>18</sup>

## RESULTS AND DISCUSSION

### Samples characterization

The cationic ratio in the as-synthesized hydrotalcite-like samples was in good agreement with the initial one, as the EPMA results showed no major changing in the final composition. The corresponding chemical formulas (calculated from the Mg, Ni and Al content and considering the  $\text{CO}_3^{2-}$  as the interlayer anion) are shown in Table 1.

The analysis by X-ray diffraction allowed the computation of the parameters of cell unit, *a* and *c*, which highlighted the rhomboedric symmetry of the synthesized hydrotalcite-like compounds (Table 2).

Table 1

The chemical formula for the as-synthesized samples based on the EPMA results

Sample	%wt Mg	%wt Ni	%wt Al	x	Chemical formula
Mg <sub>3</sub> Al	39.0	-	17.8	0.29	$[\text{Mg}_{0.708}\text{Al}_{0.291}(\text{OH})_2] \cdot (\text{CO}_3^{2-})_{0.145} \cdot 0.563\text{H}_2\text{O}$
Mg <sub>2</sub> NiAl	24.7	26.0	13.2	0.25	$[\text{Mg}_{0.499}\text{Ni}_{0.249}\text{Al}_{0.251}(\text{OH})_2] \cdot (\text{CO}_3^{2-})_{0.125} \cdot 0.623\text{H}_2\text{O}$
Mg <sub>1.5</sub> Ni <sub>1.5</sub> Al	15.9	40.5	11.4	0.23	$[\text{Mg}_{0.380}\text{Ni}_{0.380}\text{Al}_{0.239}(\text{OH})_2] \cdot (\text{CO}_3^{2-})_{0.119} \cdot 0.641\text{H}_2\text{O}$
MgNi <sub>2</sub> Al	11.4	46.7	11.0	0.24	$[\text{Mg}_{0.252}\text{Ni}_{0.504}\text{Al}_{0.243}(\text{OH})_2] \cdot (\text{CO}_3^{2-})_{0.121} \cdot 0.623\text{H}_2\text{O}$
Ni <sub>3</sub> Al	-	64.8	9.1	0.23	$[\text{Ni}_{0.765}\text{Al}_{0.234}(\text{OH})_2] \cdot (\text{CO}_3^{2-})_{0.117} \cdot 0.649\text{H}_2\text{O}$

$$x = \text{M}^{\text{III}} / (\text{M}^{\text{II}} + \text{M}^{\text{III}})^{15}$$

Table 2

Parameters of cell unit, *a* and *c*, for the synthesized samples

Ni/Mg	Sample	<i>a</i> , Å	<i>c</i> , Å	Symmetry
0	Mg <sub>3</sub> Al	3.04	23.37	3R
0,5	NiMg <sub>2</sub> Al	3.03	23.25	3R
1	Ni <sub>1,5</sub> Mg <sub>1,5</sub> Al	3.02	23.18	3R
2	Ni <sub>2</sub> MgAl	3.00	23.13	3R
3	Ni <sub>3</sub> Al	3.01	23.00	3R

The *a* parameter corresponded to cation-cation distance within brucite layer and *c* = 3*d*<sub>003</sub> parameter is the thickness of the layer made of a brucite-like film and an interlayer.

The decrease of the *a* and *c* parameter as the isomorphous substitution degree increase can be attributed to the changes in the brucite-like network (the shorter band length of Ni-O (2.084Å) than Mg-O (2.11Å)).<sup>1</sup>

In addition, the thermal behaviour of samples was influenced by the isomorphous substitution (Table 3).

The thermal stability of the as-synthesized hydrotalcite-like samples is lowered as the substitution degree increase. Therefore, the reconstruction of the ternary Mg/Ni-Al-type hydrotalcites is inhibited by the presence of the Ni<sup>2+</sup> cations and the adsorption results could be associated with the increased surface area after calcinations (Figure 1).

Table 3

Thermal analysis results

Stage	Samples Mg <sub>x</sub> Ni <sub>y</sub> Al									
	Mg/Ni = 3 : 0		Mg/Ni = 2 : 1		Mg/Ni = 1.5 : 1.5		Mg/Ni = 1 : 2		Mg/Ni = 0 : 3	
	T (°C)	%	T (°C)	%	T (°C)	%	T (°C)	%	T (°C)	%
1	100	2.35	100	3.99	88.6	2.53	100	4.47	88.8	2.68
2	212.2	12.67	138.5	7.30	215.1	12.91	163.1	10.21	207.5	12.23
3	379.0	31.85	371.8	29.39	372.2	29.35	322.2	26.18	337.6	26.43

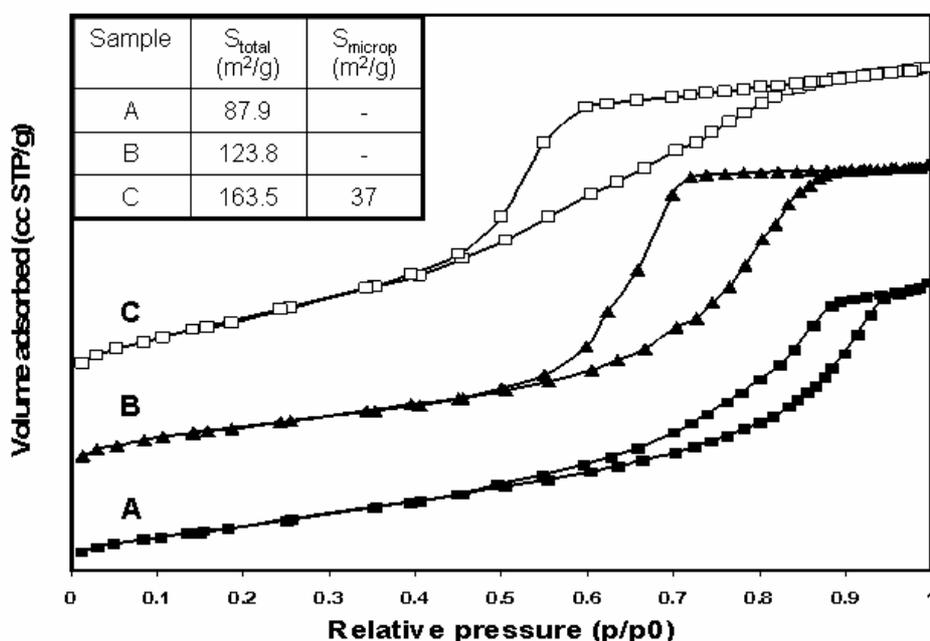


Fig. 1 – N<sub>2</sub> adsorption/desorption isotherms at 77K of calcined (A) Mg<sub>3</sub>Al, (B) Mg<sub>1,5</sub>Ni<sub>1,5</sub>Al and (C) Ni<sub>3</sub>Al samples.

All the isotherms are of the Type IV but the hysteresis loops are different. The calcined Mg<sub>3</sub>Al shows the H<sub>1</sub> type hysteresis loop while the presence of the H<sub>2</sub> type in the Ni-containing

samples is observed. Therefore, the pore shapes are different when the Ni<sup>2+</sup> cations are introduced. In the first case the H<sub>1</sub> hysteresis loop is relatively narrow with the adsorption and desorption

branches nearly parallel showing the presence of the tubular pores. In the former case, the  $H_2$  hysteresis loop is broad, the desorption branch being much steeper than the adsorption branch, typical for many inorganic oxides, where the pore structures are complex and tend to be made up of interconnected networks of pores at different size and shapes.<sup>16</sup>

### Sorption equilibrium of phosphate on Mg/Ni-Al -type hydrotalcites

The experimental results showed that the sorption capacity of phosphate remained constant for all studied samples at the upper limit of the equilibrium concentration range. That was the reason for using, in a first approach, the Langmuir isotherm (Eq. 1 and Eq. 2) to interpret equilibrium data:

$$q_e = q_{\max} \frac{K_L \cdot C_e}{1 + K_L \cdot C_e} \quad (1)$$

$$\frac{C_e}{q_e} = \frac{C_e}{q_{\max}} + \frac{1}{K_L \cdot q_{\max}} \quad (2)$$

where:  $q_e$  – anion quantity sorbed at equilibrium,  $\text{mg} \cdot \text{g}^{-1}$ ;

$q_{\max}$  – maximum sorbed quantity,  $\text{mg} \cdot \text{g}^{-1}$ ;

$C_e$  – equilibrium concentration,  $\text{mg} \cdot \text{L}^{-1}$ ;

$K_L$  – equilibrium constant,  $\text{L} \cdot \text{mg}^{-1}$ .

The corresponding sorption isotherms are shown in Figures 2 – 6.

By analysing the results, it resulted that the best sorption behaviour was for  $\text{Mg}_3\text{Al}$  sample. The maximum sorption capacity of phosphate on this adsorbent was  $111 \text{ mg} \cdot \text{g}^{-1}$ . This sorption behaviour of the  $\text{Mg}_3\text{Al}$  sample can be attributed to the memory effect property of this type of materials. On the other hand, as  $\text{Mg}^{2+}$  substitution by  $\text{Ni}^{2+}$  increased the sorption capacity decreased (Table 4) and the sorption results could be attributed only to the external surface area developed when the samples are thermally activated at  $500^\circ\text{C}$ .

There was a sharp decrease from  $\text{Mg}_3\text{Al}$  to  $\text{Mg}_2\text{NiAl}$  followed by a gradual decrease within the rest of Mg/Ni-Al hydrotalcite series. The sharp decrease of the sorption capacity between the  $\text{Mg}_3\text{Al}$  and  $\text{Mg}_2\text{NiAl}$  sample can be attributed to the presence of the  $\text{Ni}^{2+}$  cations in the brucite-like network. These cations are acting as inhibitors for the reconstruction process; therefore, the reduced sorption capacity can be attributed to an impeded degree of reconstruction in the presence of nickel. The sorption experiments of the phosphate anions on three representative samples were monitored using the X-ray diffraction technique (Figure 7).

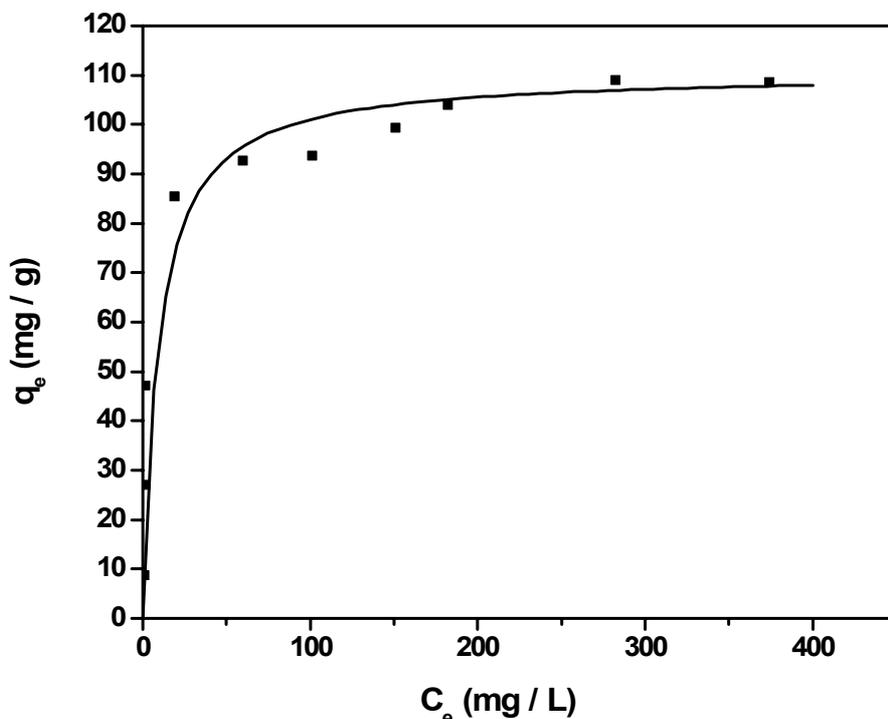


Fig. 2 – Sorption isotherm of phosphate on  $\text{Mg}_3\text{Al}$  sample.

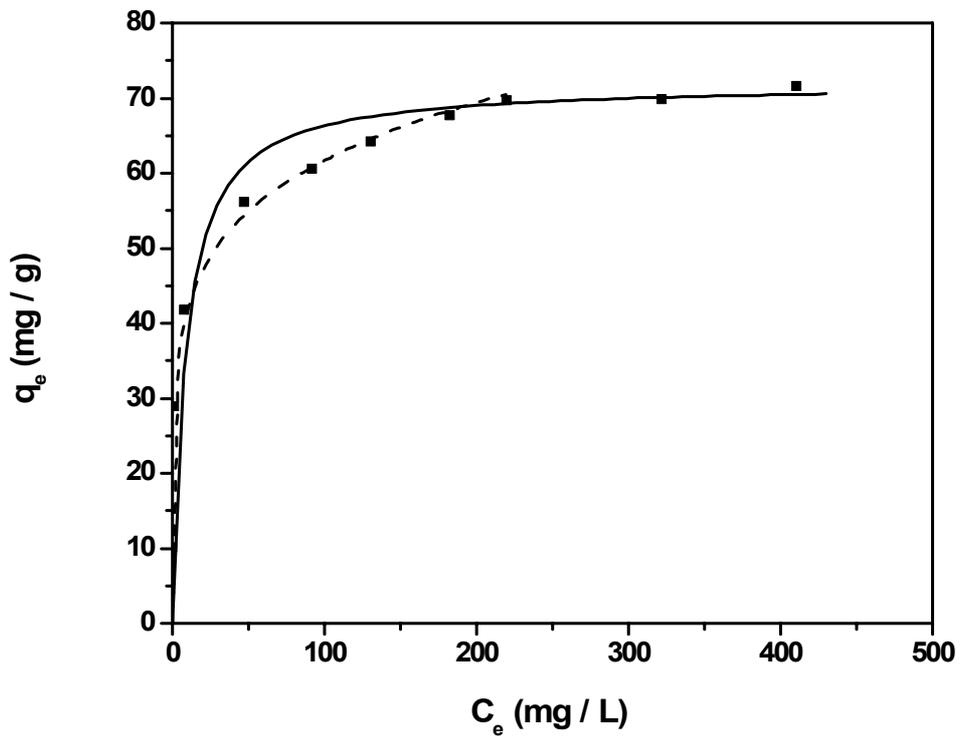


Fig. 3 – Sorption isotherms of phosphate on Mg<sub>2</sub>NiAl sample; Full line – Langmuir; dotted line – Freundlich.

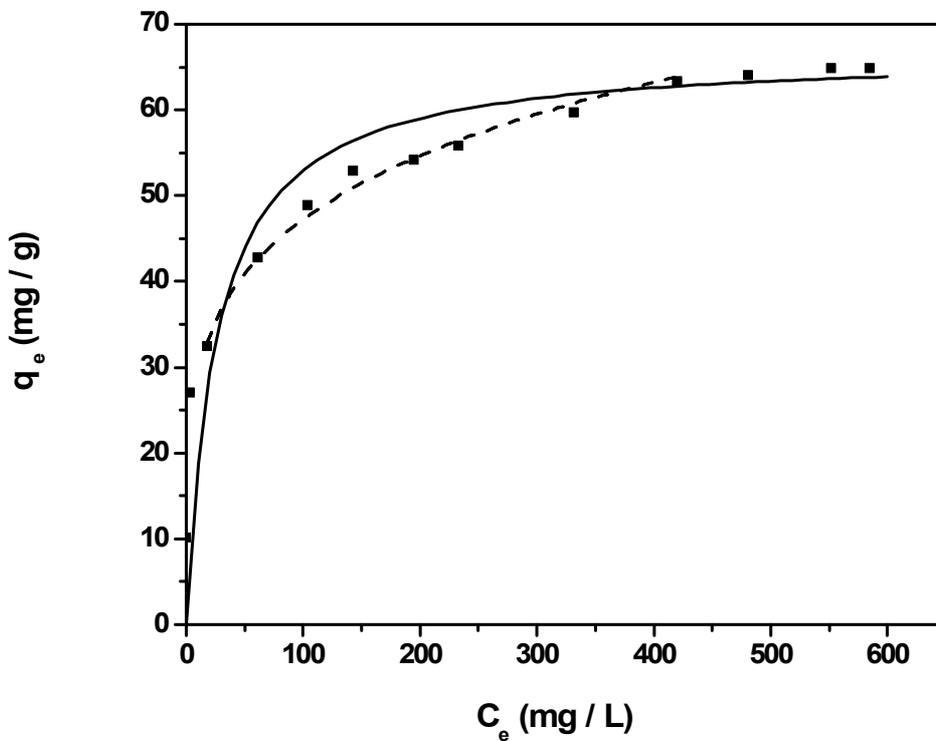


Fig. 4 – Sorption isotherms of phosphate on Mg<sub>1.5</sub>Ni<sub>1.5</sub>Al sample.

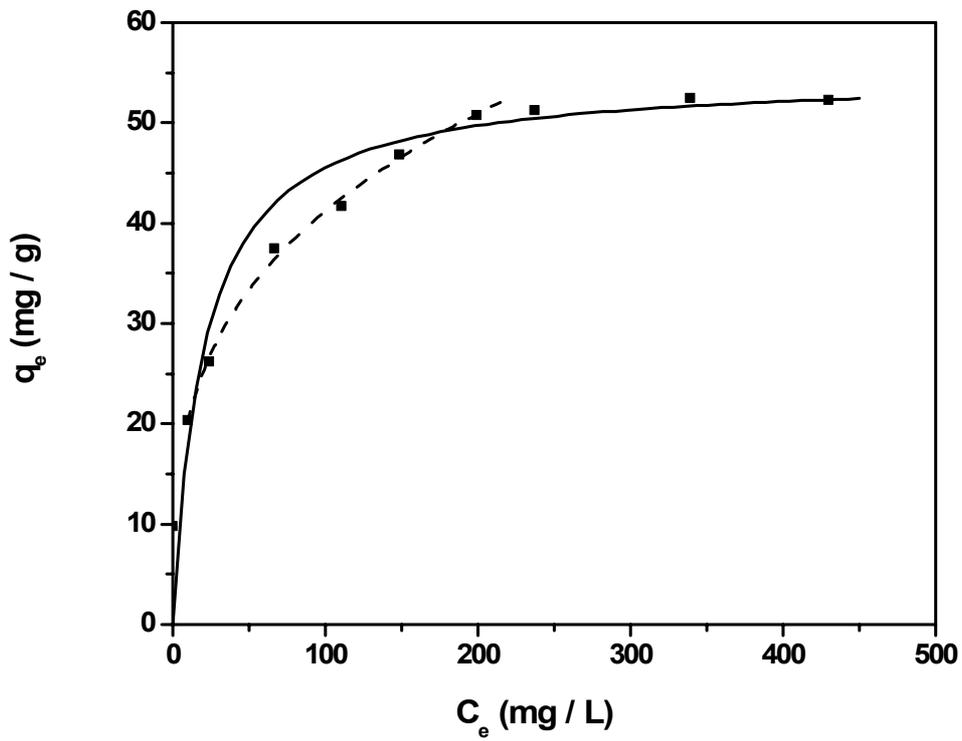
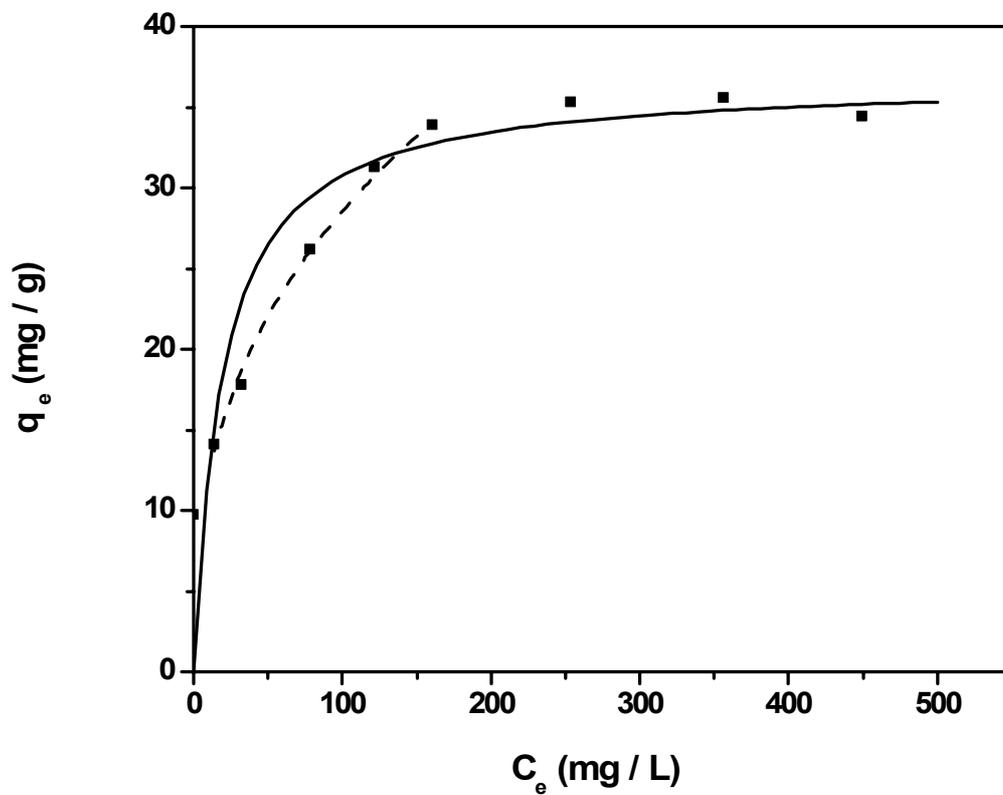
Fig. 5 – Sorption isotherms of phosphate on MgNi<sub>2</sub>Al samples.Fig. 6 – Sorption isotherms of phosphate on Ni<sub>3</sub>Al sample.

Table 4

Sorption equilibrium characteristics of phosphate on Mg/Ni-Al –type hydrotalcites

Sample	Langmuir isotherm					Freundlich isotherm		
	$\frac{C_e}{q_e} = \frac{C_e}{q_{\max}} + \frac{1}{K_L \cdot q_{\max}}$					$\ln q_e = \ln \alpha + \beta \cdot \ln C_e$		
	Concentration range (mg · L <sup>-1</sup> )	q <sub>max</sub> (mg · g <sup>-1</sup> )	Equilibrium constant, K <sub>L</sub>		Δ <sub>ads</sub> G <sup>0</sup> <sub>298</sub> (kJ · mol <sup>-1</sup> )	Concentration range (mg · L <sup>-1</sup> )	α	β
(L · mg <sup>-1</sup> )			(L · mol <sup>-1</sup> )					
Mg <sub>3</sub> Al	0-375	111	0.107	1.01 · 10 <sup>4</sup>	-22.9	-	-	-
Mg <sub>2</sub> NiAl	0-410	72	0.118	1.12 · 10 <sup>4</sup>	-23.1	0-220	28.4	0.169
Mg <sub>1.5</sub> Ni <sub>1.5</sub> Al	0-580	67	0.039	0.37 · 10 <sup>4</sup>	-20.4	20-420	18.0	0.210
MgNi <sub>2</sub> Al	0-430	55	0.050	0.47 · 10 <sup>4</sup>	-21.0	10-200	10.1	0.306
Ni <sub>3</sub> Al	0-450	37	0.051	0.49 · 10 <sup>4</sup>	-21.0	15-160	5.1	0.375

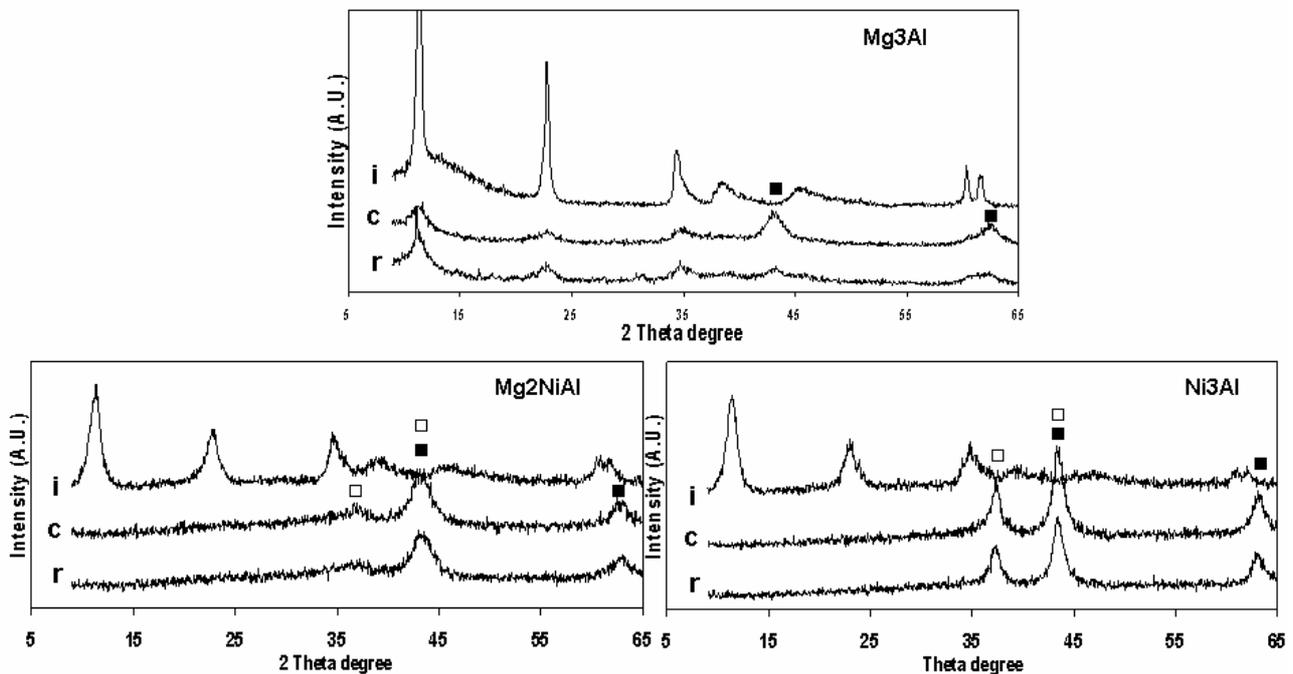


Fig. 7 – The XRD patterns of the as-synthesized, calcined and rehydrated Mg<sub>3</sub>Al, Mg<sub>2</sub>NiAl and Ni<sub>3</sub>Al samples (where i – uncalcined, c - calcined and r – rehydrated); ■ MgO and □ NiO.

The reconstruction of the layered structure of the Mg/Al mixed oxides obtained from the hydrotalcite precursor is reversible at room temperature and atmospheric pressure due to the presence of the memory effect property. On the other hand, the XRD patterns showed that if small amounts of Ni<sup>2+</sup> are added, the reconstruction process is inhibited.

Figures 2-6 also show that the slope of linear part of Langmuir isotherm (Henry's domain) decreased as the substitution degree of Mg by Ni increased following the order: Mg<sub>3</sub>Al > Mg<sub>2</sub>NiAl > Mg<sub>1.5</sub>Ni<sub>1.5</sub>Al > MgNi<sub>2</sub>Al > Ni<sub>3</sub>Al.

The Langmuir model allowed the computation of the sorption equilibrium constant, K<sub>L</sub>, and standard Gibbs energy, Δ<sub>ads</sub>G<sup>0</sup><sub>298</sub>:

$$\Delta_{ads}G_{298}^0 = -R \cdot T \cdot \ln K_L \quad (3)$$

where: R – universal constant of gases, J · mol<sup>-1</sup> · K<sup>-1</sup>;  
T – absolute temperature, K.

In accordance with the calculated standard Gibbs energies, about -20 kJ · mol<sup>-1</sup>, the process could be considered as a physical adsorption.<sup>3</sup>

The examination of Langmuir isotherms also showed that, at low/medium equilibrium concentrations, the experimental data did not follow the model closely. The deviations from Langmuir isotherm might be explained by the lack of homogeneity of adsorption surfaces (active sites are not uniformly spread on the surface; active sites are not equivalent etc.). That was the reason

for using Freundlich isotherm as exponential and linear forms (Eqs. 4 and 5) (Figures 3-6).

$$q_e = \alpha \cdot C_e^\beta \quad (4)$$

$$\ln q_e = \ln \alpha + \beta \cdot \ln C_e \quad (5)$$

where:  $q_e$  and  $C_e$  have the same meaning as previously mentioned;

$\alpha, \beta$  – constants.

In addition, the interpretation of experimental results by Freundlich isotherm can be justified by the fact that the advanced wastewater treatment does not assume operation at the quite high concentrations used for this study.

For all situations, both the subunit  $\beta$  (Table 3) and  $\Delta_{\text{ads}}G_{298}^0$  values underlined that the interaction forces between phosphate and the studied adsorbents were weak and thus the process was a physical adsorption.<sup>3</sup>

### Sorption equilibrium of thiocyanate on Mg/Ni-Al -type hydrotalcites

Figure 8 shows experimental sorption isotherms for thiocyanate. By analysing the sorption isotherms, two aspects are noticeable. Firstly, the shape of the initial part of isotherms (the absence of Henry's domain) is an indication of low sorption of thiocyanate for Mg/Ni-Al hydrotalcites. The sorption decreased in the following order:  $\text{Mg}_3\text{Al} > \text{Mg}_2\text{NiAl} > \text{Mg}_{1.5}\text{Ni}_{1.5}\text{Al} > \text{MgNi}_2\text{Al} > \text{Ni}_3\text{Al}$ .

The second aspect referred to the low values of sorption capacities that decreased in the same order as affinity did. The lower sorption capacities of thiocyanate anions on the studied samples may be due to the affinity of the mixed oxides, obtained from hydrotalcite-type precursors, greater for anions with higher charge density.

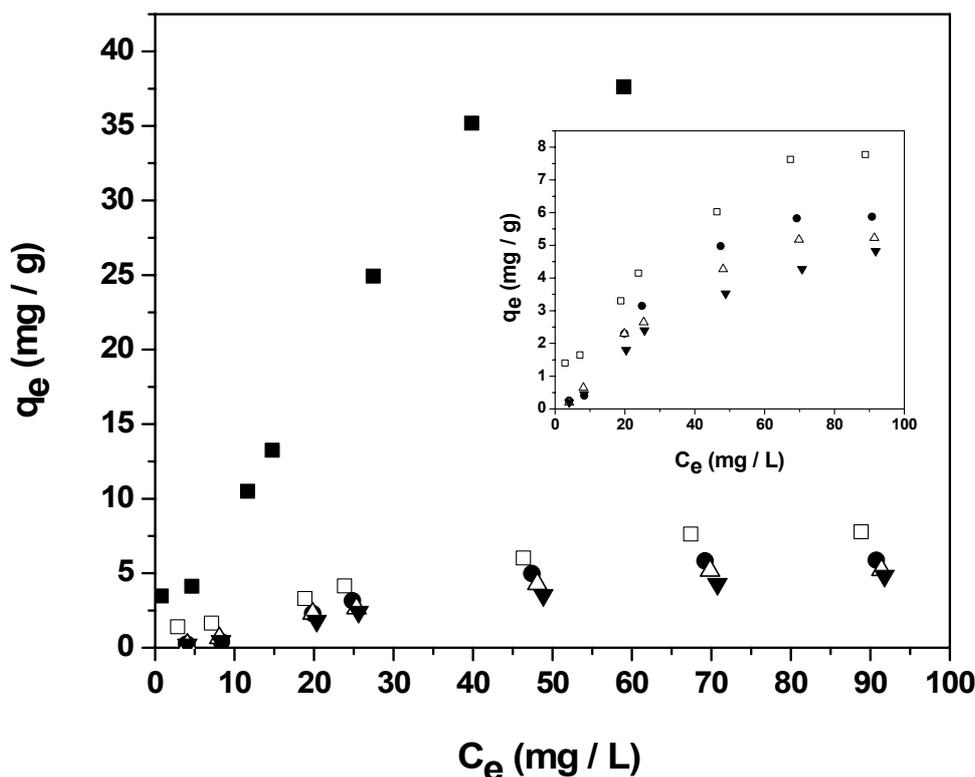


Fig. 8 – Sorption isotherms for thiocyanate on:  $\text{Mg}_2\text{NiAl}$  (□),  $\text{Mg}_{1.5}\text{Ni}_{1.5}\text{Al}$  (●),  $\text{MgNi}_2\text{Al}$  (△),  $\text{Ni}_3\text{Al}$  (▲) as compared to  $\text{Mg}_3\text{Al}$  (■). Inset: more detail on sorption isotherms of thiocyanate on Mg/Ni-Al adsorbent series.

For a given equilibrium concentration, there was a sharp decrease of sorption capacity by five times from  $\text{Mg}_3\text{Al}$  to  $\text{Mg}_2\text{NiAl}$  followed by a gradual decrease within the rest of Mg/Ni-Al hydrotalcite series. These sorption results can be also attributed to the memory effect property directly influenced by the presence of the  $\text{Ni}^{2+}$

cations in the brucite-like framework of the studied products.

The results did not justify the practical use of Mg/Ni-Al hydrotalcites for the sorption of thiocyanate.

By comparing the sorption of the two anions on Mg/Ni-Al -type hydrotalcites, it followed that

phosphate sorption occurred with good results, highlighted by process characteristics (values of the maximum sorption capacities and the slope of linear part of Langmuir isotherm), while thiocyanate sorption was ineffective; the affinity of the studied adsorbents for this anion was scanty. The results were in line with literature data,<sup>13</sup> which reported that hydrotalcites affinity for monovalent anions was modest.

## CONCLUSIONS

The equilibrium of phosphate and thiocyanate sorption on Mg/Ni-Al – type multicomponent hydrotalcites was studied under various Ni/Mg substitution ratios, using the memory effect property of the resulting materials. The thermal analysis indicated that the thermal stability of the as-synthesized hydrotalcite-like samples was lower as the substitution degree increase. The XRD results showed that the reconstruction process is inhibited by the presence of the Ni<sup>2+</sup> cations in the HT matrix.

Langmuir and Freundlich isotherms were used to interpret the equilibrium data for phosphate. The sorption behaviour of the Mg<sub>3</sub>Al sample is due to the memory effect property of this type of materials. On the other hand, as Mg<sup>2+</sup> substitution by Ni<sup>2+</sup> increase, a decrease of the sorption capacity was noticed, and the sorption results could be attributed to the external surface area enhancement when the samples are thermally activated at 500°C.

The investigation of Henry's domain on isotherms allowed assessing phosphate affinity for hydrotalcites within Mg/Ni-Al series. The following selectivity order resulted: Mg<sub>3</sub>Al > Mg<sub>2</sub>NiAl > Mg<sub>1.5</sub>Ni<sub>1.5</sub>Al > MgNi<sub>2</sub>Al > Ni<sub>3</sub>Al. This sequence corresponded with that for the maximum adsorption capacities, which decreased from 111 mg · g<sup>-1</sup> to 37 mg · g<sup>-1</sup> for Mg<sub>3</sub>Al and Ni<sub>3</sub>Al, respectively.

The work out of equilibrium data by using the Langmuir model allowed the computation of the sorption equilibrium constant, K<sub>L</sub>, and standard Gibbs energy, Δ<sub>ads</sub>G<sup>0</sup><sub>298</sub>. Based on results, the process was considered as a physical adsorption.

The lower sorption capacities of thiocyanate anions on the studied samples may be attributed to the affinity of the mixed oxides, obtained from hydrotalcite-type precursors, grater for anions with higher charge density.

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