Separation of Pt(IV) and Rh(III) from Chloride Solution by Solvent Extraction with Amine and Neutral Extractants

P. P. Sun¹, J. Y. Lee² and M. S. Lee^{1,*}

¹Department of Advanced Materials Science & Engineering, Mokpo National University, Chonnam 534-729, Korea

²Metals Recovery Department, Minerals Resources Research Division, Korea Institute of Geoscience & Mineral Resources, Daejeon 305-350, Korea

Solvent extraction experiments were performed to separate platinum and rhodium from mixed chloride solution by using amine and neutral extractants. Effects of extraction conditions on the separation of the two metals were investigated as a function of extractant concentration in the HCl concentration range from 1 to 9 mol/dm³. Alamine304-1, Alamine308, Alamine336, Aliquat336, TBP, TOPO and DOS were tested in our study. The concentration of Pt(IV) and Rh(III) in the mixed solution was fixed at 0.001 mol/dm³ and 0.0001 mol/dm³, respectively. In the extraction with amine, Pt was completely extracted irrespective of HCl concentration, while the extraction percentage of Rh depended on extraction condition. In the extraction with neutral extractants, the extraction percentage of Rh was nearly zero whereas 1 mol/dm³ TOPO completely extracted most of Pt in our experimental range. Separation of Pt from Rh by amines except Alamine336 was possible only from 9 mol/dm³ HCl solution, while complete separation of the two metals by TOPO was possible in the HCl concentration range from 1 to 9 mol/dm³. From the consideration of the economics of the separation process, our results indicate that TBP is better than TOPO in separating Pt and Rh from chloride solution. [doi:10.2320/matertrans.M2011159]

(Received May 30, 2011; Accepted August 22, 2011; Published October 25, 2011)

Keywords: platinum(IV), rhodium(III), amine, trioctylphosphine oxide (TOPO), hydrogen chloride (HCl), solvent extraction

1. Introduction

Platinum group metals (PGMs) are extensively used in automobile, chemical and electronics industry owing to their specific physical and chemical properties. Since ores containing PGMs are rare and the processes for the production of PGMs with high purity from these ores are very complicated, recovery of PGMs from secondary sources such as spent catalysts has been of importance. Platinum and rhodium are used in automobile catalysts to reduce the emission level from the exhaust gas and the automobile industry consumes PGMs in a great measure.¹⁾ The weight ratio of platinum to rhodium in the spent catalyst is about 10 : 1.

The most prominent feature of PGMs in chloride solution is that PGMs readily forms complexes with chloride ion. Among PGMs, the extraction of Rh has been known to be the most difficult owing to its intricate chemical properties in chloride solution.²⁾ Rhodium has seven existence forms of aqua-chloro complexes from Rh(H₂O)₆³⁺ to RhCl₆³⁻.²⁾ The highly charged octahedral complexes are difficult to extract owing to steric effect.²⁾ It has been reported that the tendency for the PGMs-chloro complexes to form ion pairs with anion exchangers is: MCl₆²⁻ > MCl₄²⁻ >> MCl₆³⁻.³⁾

Many commercial amines, such as Alamine304-1, Alamine308, Alamine336, and Aliquat336, have been used in extracting and separating diverse metals.⁴⁻¹³⁾

TBP, TOPO, and DOS have been widely used in the separation of base metals together with PGMs^{13,14)} and in the refining of precious metals.^{15,16)}

The existence form of PGMs and availability of certain type of metal species are highly dependent on the acidity and chloride concentration in the aqueous solution.¹⁷⁾ According to the literature, Aliquat336 is more suitable in separating

PGMs at low concentration of HCl, while tertiary amine or solvating extractants are more suitable at high concentration of hydrogen ion.¹⁷⁾

In this study, an optimum condition to separate Pt(IV) and Rh(III) from mixed chloride solution has been investigated by using several extractants, such as, Alamine304-1, Alamine308, Alamine336, Aliquat336, TBP, TOPO and DOS. By employing these extractants, the concentrations of HCl and extractant were varied and the extraction percentage of the two metals at each experimental condition was obtained. Considering the molar ratio of Pt to Rh in the spent catalysts, the molar ratio of the two metals in the mixed chloride solution was kept at 10. Moreover, an optimum stripping condition was reported to recover pure Pt(IV) from the loaded organic phase.

2. Experimental

Stock solutions of platinum and rhodium were prepared by dissolving the necessary amount of PtCl₄ (Aldrich, 98%) and RhCl₃·4H₂O (Aldrich, 99.99%) in doubly distilled water. Throughout our experiments, the concentration of Pt(IV) and Rh(III) was controlled to 1 × 10⁻³ mol/dm³ and 1 × 10⁻⁴ mol/dm³, respectively. The acidity of the mixed solution was adjusted by adding reagent HCl solution (Junsei Chem., 35%). Thiourea solution in the stripping experiments was prepared by dissolving certain amount of thiourea powder (Daejung chemicals & metals co., 96%) in doubly distilled water.

Alamine304-1, Alamine308, Alamine336, and Aliquat336 were purchased from Cognis corporation and were used without further purification. Chemical formula of each amine is represented in Table 1. Except Alamine304-1, amine extractants were pretreated with HCl to form salt (R₃NHCl) which takes part in the solvent extraction of metals.

*Corresponding author, E-mail: mslee@mokpo.ac.kr

Table 1 Chemical formula of extractants used in this study.

Extractant	Reagent class	Structure
Alamine304-1	tertiary amine	$[\text{CH}_3(\text{CH}_2)_{11}]_3\text{N}$
Alamine308	tertiary amine	$[(\text{CH}_3)_2\text{CH}(\text{CH}_2)_5]_3\text{N}$
Alamine336	tertiary amine	$[\text{C}_8\text{-C}_{10} \text{ mixture}]_3\text{N}$
Aliquat336	Aliphatic quaternary ammonium salt	$[(\text{C}_8\text{H}_{17})_3\text{N}(\text{CH}_3)]^+\text{Cl}^-$
TBP	Tri-n-butyl phosphate	$[\text{CH}_3(\text{CH}_2)_3\text{O}]_3\text{PO}$
TOPO	Trioctylphosphine oxide	$[\text{CH}_3(\text{CH}_2)_7]_3\text{PO}$
DOS	Di-n-octyl sulfide	$[\text{CH}_3(\text{CH}_2)_7]_2\text{S}$

Alamine304-1 was used as received for its solidification after the pretreatment with HCl solution. The concentration of extractant was adjusted by diluting with toluene.

TBP and DOS solutions were also prepared by diluting certain amounts of TBP (Yakuri pure chemicals co., 99%) and DOS (Aldrich, 97%) in toluene. TOPO solution was prepared by dissolving the necessary amount TOPO powder (Aldrich, 95%) in toluene. Chemical formula of each agent is also represented in Table 1.

Some studies have reported that aging effect exists in handling rhodium chloride solution.^{2,14,15,18)} Therefore, throughout our solvent extraction experiments, freshly prepared aqueous solutions were used. Equal volumes (20 cm^3) of aqueous and organic phases were mixed in a 100 cm^3 screwed cap bottle and shaken for 30 min with a wrist action shaker. The aqueous phase was separated after settling the mixture for 24 h. The organic phase was stripped by certain reagent at equal volume (20 cm^3). All the extraction and stripping experiments were carried out at ambient temperature. The concentration of metal in the aqueous phase was measured by ICP-OES (Spectro arcs). The concentration of metal in the organic phase was obtained by mass balance.

3. Results and Discussion

3.1 Separation of Pt(IV) and Rh(III) by amines

Effect of the concentration of amine extractant and HCl on the separation of Pt(IV) and Rh(III) was investigated. For this purpose, the concentration of amine extractant was varied from 0.1 to 0.7 mol/dm^3 , and HCl concentration was controlled from 1 to 9 mol/dm^3 . The composition of the mixed chloride solution was $1 \times 10^{-3}\text{ mol/dm}^3$ Pt and $1 \times 10^{-4}\text{ mol/dm}^3$ Rh.

Figure 1 is the results obtained by Alamine304-1. In our experimental range, most of Pt(IV) was extracted irrespective of Alamine304-1 concentration in the HCl concentration range from 1 to 9 mol/dm^3 . However, the extraction percentage of Rh(III) was very low when Alamine304-1 concentration was 0.1 mol/dm^3 in the whole HCl concentration range. The extraction percentage of Rh was more than 50% when Alamine304-1 concentration was higher than 0.5 mol/dm^3 in 1 and 3 mol/dm^3 HCl solution. Extraction percentage of Rh decreased with the increase of HCl concentration at the same Alamine304-1 concentration and the extraction percentage of Rh was nearly zero at 9 mol/dm^3

Fig. 1 Effect of Alamine304-1 and HCl concentration on the extraction of Pt(IV) and Rh(III).

HCl solution. Therefore, it might be concluded that Pt and Rh can be completely separated from mixed chloride solution by solvent extraction with Alamine304-1 by adjusting the concentration of HCl to 9 mol/dm^3 .

The extraction reaction of Pt(IV) by amine can be represented as^{2,10)}

The above equation implies that the extraction of platinum decreases at higher concentration of hydrochloric acid owing to the mass action effect of chloride ions.^{19,20)} However, the effect of HCl concentration on the extraction of platinum was not obvious in our system.

There are seven kinds of aqua-chloro complexes of Rh in chloride solution, and RhCl_6^{3-} is predominant at high concentration of HCl (9 mol/dm^3).²⁾ Extraction of RhCl_6^{3-} by Alamine304-1 from strong HCl solution could be represented as:

When HCl concentration was low, extensive aquation reaction took place and Rh exists as a hydrophilic complex, such as $\text{RhCl}_5(\text{H}_2\text{O})^{2-}$. Extraction reaction of this species by Alamine304-1 is represented below^{2,21,22)}

Anion exchange reaction is responsible for the decrease in the extraction percentage of Rh with the increase of HCl concentration from 1 to 9 mol/dm^3 . Mass action effect of chloride ion suppressed the extraction of Rh by Alamine304-1 at high HCl concentration.

Figure 2 shows extraction percentage of Pt and Rh by Alamine308. Extraction percentage of Pt was higher than 99% in our experiment conditions, while that of Rh depended on the concentration of HCl and Alamine308. When HCl concentration was 1 and 3 mol/dm^3 , extraction percentage of Rh increased linearly with the increase of Alamine308 concentration. However, in strong HCl solution from 5 to 9 mol/dm^3 , extraction percentage of Rh was nearly zero.

Fig. 2 Effect of Alamine308 and HCl concentration on the extraction of Pt(IV) and Rh(III).

Fig. 4 Effect of Aliquat336 and HCl concentration on the extraction of Pt(IV) and Rh(III).

Fig. 3 Effect of Alamine336 and HCl concentration on the extraction of Pt(IV) and Rh(III).

Hence it is possible to separate Pt and Rh when HCl concentration is higher than 5 mol/dm³.

Figure 3 shows the experimental results obtained with Alamine336. Extraction behavior of Pt by Alamine336 was similar to those results obtained by Alamine304-1 and Alamine308. Most of Pt was extracted into Alamine336 in our experimental range. However, the concentration of HCl and Alamine336 had a great effect on the extraction of Rh by Alamine336. Unlike Alamine304-1 and Alamine308, about 10% of Rh was extracted from 7 and 9 mol/dm³ HCl solution by Alamine336. Extraction percentage of Rh was similar to each other when the concentration of HCl was 1 and 3 mol/dm³. When HCl concentration was 5 mol/dm³, extraction percentage of Rh by Alamine336 was around 60%.

Figure 4 shows the experimental results obtained by Aliquat336. As is shown in Fig. 4, extraction percentage of Pt was higher than 99% in our experimental range. The highest extraction percentage of Rh was 30% at 1 mol/dm³ HCl. Although extraction percentage of Rh at 7 mol/dm³ HCl was slightly higher than that at 9 mol/dm³, extraction percentage of Rh was nearly zero when HCl concentration

was higher than 5 mol/dm³. Extraction percentage of Rh decreased with increasing HCl concentration from 1 to 9 mol/dm³.

In the extraction with Alamine308 and Aliquat336, separation factor between Pt and Rh by Aliquat336 was higher than that by Alamine308. Therefore, Aliquat336 was found to be the best amine extractant for the separation of Pt and Rh from mixed chloride solution in our experimental range.

Solvent extraction reaction of Pt and Rh from chloride solution by amines follows anion exchange mechanism. Amines belong to strong-base extractant and are readily protonated in inorganic acid solution. The difference in the extraction percentage by amine between Pt and Rh might be related to the tendency to form chloro-complexes with anion exchanger, $\text{MCl}_6^{2-} \gg \text{MCl}_6^{3-}$. Therefore, extraction percentage of Pt was much higher than that of Rh in our experimental range.

3.2 Separation of Pt(IV) and Rh(III) by TBP, TOPO, and DOS

Separation experiments of Pt and Rh from mixed chloride solution were also carried out by using neutral extractants, such as TBP, TOPO, and DOS. The concentration of HCl of the mixed solution was varied from 1 to 9 mol/dm³. TBP concentration was varied from 0.5 to 2 mol/dm³, and the results are shown in Fig. 5. Extraction percentage of Rh was zero at all the HCl concentration in our experimental range. In the case of Pt, extraction percentage depended on the concentration of HCl and TBP. When HCl concentration was 1 mol/dm³, extraction percentage of Pt was low. However, extraction percentage of Pt increased with the increase of HCl concentration up to 7 mol/dm³ and then decreased at 9 mol/dm³ HCl. Low extraction percentage of Rh by TBP has been pointed out in the literature.^{15,21)} The maximum value of Pt extraction percentage was 81% at 7 mol/dm³ HCl by 2.0 mol/dm³ TBP. The extraction reaction of Pt by TBP might be expressed as:²¹⁾

Fig. 5 Effect of TBP and HCl concentration on the extraction of Pt(IV) and Rh(III).

Fig. 6 Effect of HCl concentration on the extraction of Pt(IV) and Rh(III) by DOS, TBP and TOPO (Concentration of extractants were 1.0 mol/dm³).

In order to compare the extraction behavior of the two metals by TBP, TOPO, and DOS, solvent extraction experiments with TOPO and DOS were done by varying HCl concentration from 1 to 9 mol/dm³. In these experiments, the concentration of TOPO, and DOS was fixed at 1 mol/dm³, and the results are shown in Fig. 6. Extraction percentage of Rh by the above three extractants was nearly zero at all the HCl concentration investigated in this study. At the same time, less than 10% of Pt was extracted by DOS while most of Pt was extracted by TOPO irrespective of HCl concentration. Therefore, TOPO was found to be superior to TBP and DOS in extracting and separating Pt and Rh from the mixed chloride solution.

Solvent extraction reaction of Pt and Rh by TBP and TOPO may be same and follows solvation mechanism.²³⁾ In terms of their structure as shown in Table 1, the difference in the extraction abilities between TBP and TOPO might be correlated with their donor ability of phosphoryl oxygen.²⁴⁾ They are separately affected by three electron donation groups $[\text{CH}_3(\text{CH}_2)_7-]$ in TOPO and electron withdrawing groups $[\text{CH}_3(\text{CH}_2)_3\text{O}-]$ in TBP, so donor ability of phosphoryl oxygen follows the order: TOPO > TBP.

Table 2 Effect of HCl concentration on the stripping of Pt from loaded TOPO.

HCl concentration, $C/\text{mol}\cdot\text{dm}^{-3}$	Stripping (%)
0.001	0.08
0.1	0.02
0.2	0.02
0.3	0.02
0.5	0.02

Our experimental results show that amine extractants except Alamine336 can separate Pt from Rh from 9 mol/dm³ HCl solution. In the extraction by neutral extractants, Rh was not extracted by TBP, TOPO, and DOS, and the extraction percentage of Pt depended on HCl concentration and on the nature of extractants. Among the neutral extractants, extraction with TOPO led to complete separation of Pt from Rh from not only weak but also strong HCl solution in our experimental range.

3.3 Stripping

In the solvent extraction of Pt(IV) and Rh(III) by 1 mol/dm³ TOPO, most of Pt was extracted while Rh remained in the aqueous solution. In order to find an optimum condition for the stripping of Pt from the loaded TOPO phase, stripping experiments have been done. At first, HCl solution was chosen as a stripping agent and Table 2 shows the results on the variation of stripping percentage of Pt with HCl concentration. In the HCl concentration range from 0.01 to 0.5 mol/dm³, stripping percentage of Pt was lower than 0.1%.

It has been reported that mixture of HCl and thiourea can strip Pt from the loaded Alamine336, Alamine308, Aliquat336 organic phase after solvent extraction from chloride solution.^{7,25,26)} Therefore, mixture of HCl and thiourea was tried to strip Pt from the loaded TOPO phase. In these experiments, the concentration of thiourea was changed from 0.1 to 1 mol/dm³, while HCl concentration was kept at 0.5 mol/dm³, and the results are shown in Fig. 7. The stripping percentage of Pt increased with increasing the concentration of thiourea, and 80% of Pt was stripped by a mixture of 0.5 mol/dm³ HCl and 1 mol/dm³ thiourea. The concentration of Rh in the stripped solution was negligible. In the absence of thiourea, the stripping percentage of Pt by 0.5 mol/dm³ HCl was below 0.1%. Addition of thiourea to HCl solution has a dramatic effect in increasing the stripping of Pt from the loaded TOPO. Increase in the stripping percentage of Pt in the presence of thiourea might be related with the fact that thiourea is a soft ligand. Therefore, thiourea is effective in stripping Pt(IV) from the loaded TOPO through the formation of $\text{Pt}(\text{thiourea})_4^{+4}$, which is water soluble.²⁷⁾

In the extraction with TBP, the highest extraction percentage was 81% at 7 mol/dm³ HCl by 2.0 mol/dm³ TBP. However, stripping of metal from the loaded TBP phase is very easy. It has been reported that Pt can be easily stripped from the loaded TBP phase by using distilled water.²⁸⁾ Although TOPO can extract most of the Pt from the mixed

Fig. 7 Effect of thiourea concentration on the stripping of Pt from TOPO in the presence of 0.5 mol/dm³ HCl.

Table 3 Comparison of the separation Pt and Rh by using TBP and TOPO.

Material Needed	Separation of Pt and rh by using TOPO	Separation of Pt and rh by using TBP
Extractant	1 mol·dm ⁻³	2 mol·dm ⁻³
HCl	1 mol·dm ⁻³	7 mol·dm ⁻³
Extraction percentage of Pt	99%	81%
Strip agent	0.5 mol·dm ⁻³ HCl + 1 mol·dm ⁻³ thiourea	Distilled water
Stripping efficient	80%	Nearly 100%

solution in our experimental range, stripping of Pt from the loaded TOPO phase can be possible by using a mixture of thiourea and HCl. In terms of the ease of the stripping from the loaded phase, TBP is better than TOPO.

Comparison of the separation of Pt and Rh from the mixed chloride solution by using TBP and TOPO is shown in Table 3. Considering that the feed solutions of both conditions are acid solution, we omit the difference in equipment requirement in practice. The costs to obtain same amount of platinum aqueous solution can be calculated on the basis of the prices shown in Table 4. By calculation, the cost of separation of Pt and Rh by using TOPO is much higher than that of TBP. Besides, after regeneration of TBP, platinum chloride solution will be obtained, while in the case of TOPO, the solution containing Pt(thiourea)₄⁺⁴ will be obtained. Obviously, it is much easier to deal with platinum chloride than Pt(thiourea)₄⁺⁴.

Therefore, if this study could be put into practice, we recommend separate Pt and Rh from mixed chloride solution by using TBP.

Table 4 Comparison of the TBP and TOPO processes on the basis of price.

Agent name	Price (USD/dollar)	Pack size	Company
TBP	174.3	500 cm ³	Fluka
TOPO	376.95	100 g	Aldrich
HCl	304.5	500 cm ³	Sigma
Thiourea	168	500 g	Sigma-Aldrich

4. Conclusion

Solvent extraction separation experiments of Pt(IV) and Rh(III) were performed by using Alamine304-1, Alamine308, Alamine336, Aliquat336, TBP, TOPO, and DOS from mixed chloride solution. In our experimental range, most of Pt was extracted from the mixed solution by using amines and TOPO. In the extraction with amine, extraction percentage of Rh depended on HCl concentration and the nature of extractant. In the case of Alamine304-1, Alamine308, and Aliquat336, extraction percentage of Rh was nearly zero at 9 mol/dm³ HCl concentration, while about 10% Rh was extracted into Alamine336 from 9 mol/dm³ HCl solution. In terms of separation factor, Aliquat336 was found to be better than other amines. In the extraction with TBP, TOPO, and DOS, extraction percentage of Rh was nearly zero irrespective of HCl concentration. While extraction percentage of Pt by TBP and DOS was low and depended on HCl concentration, extraction percentage of Pt was 99% by 1 mol/dm³ TOPO in the HCl concentration range from 1 to 9 mol/dm³.

In the case of amine extractants except Alamine336, separation of Pt and Rh was possible only from 9 mol/dm³ HCl solution. However, it was possible to completely separate Pt and Rh from weak HCl solution as well as strong HCl solution by using TOPO. In terms of economics of the separation process and the ease of handling the stripping solution, TBP is better than TOPO in separating both metals from chloride solution.

Acknowledgments

This work was supported by a grant from the fundamental R&D Program for Core Technology of Materials funded by the Ministry of Knowledge Economy, Republic of Korea.

REFERENCES

- 1) C. R. M. Rao and G. S. Reddi: Trends Anal. Chem. **19** (2000) 565–586.
- 2) E. Benguerel, G. P. Demopoulos and G. B. Harris: Hydrometallurgy **40** (1996) 135–152.
- 3) F. R. Bernardis, R. A. Grant and D. C. Sherrington: React. Funct. Polym. **65** (2005) 205–217.
- 4) N. A. Sayar, M. Filiz and A. A. Sayar: Hydrometallurgy **96** (2009) 148–153.
- 5) N. A. Sayar, A. A. Sayar and M. Filiz: Hydrometallurgy **95** (2009) 280–284.
- 6) M. Filiz: Hydrometallurgy **87** (2007) 58–62.
- 7) J. Y. Lee, J. R. Kumar, J. S. Kim, D. J. Kim and H. S. Yoon: J. Ind. Eng. Chem. **15** (2009) 359–364.
- 8) J. Y. Lee, B. Raju, B. N. Kumar, J. R. Kumar, H. K. Park and B. R. Reddy: Sep. Purif. Technol. **73** (2010) 213–218.

- 9) J. W. An, B. H. Jung, Y. H. Lee, T. Tran, S. J. Kim and M. J. Kim: *Miner. Eng.* **22** (2009) 1026–1031.
- 10) M. S. Lee, J. Y. Lee, R. J. Kumar, J. S. Kim and J. S. Sohn: *Mater. Trans.* **49** (2008) 2823–2828.
- 11) M. S. Lee, K. J. Lee and Y. J. Oh: *Mater. Trans.* **45** (2004) 2364–2368.
- 12) H. F. Gai, J. L. Shen and M. A. Hughes: *Hydrometallurgy* **25** (1990) 293–304.
- 13) M. Moiski: *Talanta* **25** (1978) 163–165.
- 14) E. Benguerel, G. P. Demopoulos, G. Cote and D. Bauer: *Solvent Extr. Ion Exch.* **12** (1994) 497–516.
- 15) L. H. Zou, J. Chen and X. J. Pan: *Hydrometallurgy* **50** (1998) 193–203.
- 16) P. A. Lewis, D. F. C. Morris, E. L. Short and D. N. Waters: *J. less-common Met.* **45** (1976) 193–214.
- 17) C. S. Kedari, M. T. Coll, A. Fortuny, E. Goralska and A. M. Sastre: *Sep. Sci. Technol.* **40** (2005) 1927–1946.
- 18) E. Benguerel, G. Cote, A. Lautie, G. Demopoulos and D. Bauer: *J. Chem. Technol. Biotechnol.* **62** (1995) 380–384.
- 19) A. A. Mhaske and P. M. Dhadke: *Hydrometallurgy* **61** (2001) 143–150.
- 20) M. Shafiqul Alam and K. Inoue: *Hydrometallurgy* **46** (1997) 373–382.
- 21) S. N. Ashrafizadeh and G. P. Demopoulos: *J. Colloid Interf. Sci.* **173** (1995) 448–459.
- 22) G. Levitin and G. Schmuckler: *React. Funct. Polym.* **54** (2003) 149–154.
- 23) P. Charlesworth: *Platinum Metals Rev.* **25** (1981) 106–112.
- 24) K. J. Reddy, A. V. Reddy, B. S. Shaibu and M. L. P. Reddy: *Radiochim. Acta* **95** (2007) 289–295.
- 25) B. R. Reddy, B. Raju, J. Y. Lee and H. K. Park: *J. Hazard. Mater.* **180** (2010) 253–258.
- 26) J. Y. Lee, J. R. Kumar, J. S. Kim, H. K. Park and H. S. Yoon: *J. Hazard. Mater.* **168** (2009) 424–429.
- 27) M. Rovira, J. L. Cortina and A. M. Sastre: *Solvent Extr. Ion Exch.* **17** (1999) 333–349.
- 28) S. S. Dragulović, V. K. Trujić, Z. S. Stanojević Šimšić, V. B. Cvetkovski, Z. S. Ljubomirović, S. B. Dimitrijević and D. B. Simonović: *Trends in the Development of Machinery and Associated Technology*, 14th Int. Research/Expert Conference, ed. by S. Ekinović, Y. Uctug, J. V. Calvet, (2010) pp. 169–171.