

# The Sered' Leached Nickel Ores Residues – A Magnetic Sorbent for Fission Radionuclides and Lead Contaminants of Solids

<sup>a</sup>F. MACÁŠEK, <sup>a</sup>J. KUFCÁKOVÁ, <sup>a</sup>P. RAJEC, <sup>a</sup>R. KOPUNEC, <sup>b</sup>Š. JAKABSKÝ, <sup>b</sup>M. LOVÁS, and <sup>b</sup>S. HREDZÁK

<sup>a</sup>Department of Nuclear Chemistry, Faculty of Natural Sciences, Comenius University, SK-842 15 Bratislava  
e-mail: macasek@fns.uniba.sk

<sup>b</sup>Institute of Geotechnics of the Slovak Academy of Sciences, SK-043 53 Košice

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Black powdered residues from ammoniacal leaching of iron-nickel laterite ores at Sered' plant (further: SOR), the raw and mechanochemically activated by the grinding in potassium ferrocyanide solution in vibrating, ball or attrition mills, were used as a seminatural magnetic sorbent. The sorption properties for Cs<sup>+</sup>, Sr<sup>2+</sup>, Pb<sup>2+</sup>, and Eu<sup>3+</sup> ions were evaluated using the Toth sorption isotherm. Activation of sorbent occurs on a small fraction of surface. The vibrating mill activation appears as the most efficient in respect of its activation, though the biggest increase of specific surface was achieved by attrition grinding. Speciation by the Tessier and BCR standard leaching procedures indicates differences between the ions. Raw and activated SOR have convenient sorption properties for caesium, strontium, lead and partially also for trivalent lanthanides. That, together with their magnetic properties makes them promising for treatment of radioactive contaminated soils and sediments. Sorption of anions was demonstrated by TcO<sub>4</sub><sup>-</sup> sorption as a result of secondary ion-exchange mechanism. The feasible desorption process and recycling of used SOR remains an open task.

The utility of magnetic sorbents is their potential application in heterogeneous systems like suspensions and body liquids, where they can be specifically removed in a magnetic field. This advantage was early recognized in life sciences [1, 2] but also in waste treatment and technology [3]. Ferrites possess the property of spontaneous magnetization and are crystalline materials soluble only in strong acid. Iron atoms in iron ferrite (FeO·Fe<sub>2</sub>O<sub>3</sub>) can be replaced by many other metal ions without seriously altering its spinel structure. Sorption of heavy metals on recyclable magnetite has been applied for wastewater treatment [4–7]. Low-temperature formation of magnetite *in situ* and metal scavenging from aqueous solutions was also reported as an efficient method [8].

The formulations of various ferromagnetic ion exchangers and ferritized sorbents were patented [9, 10]. Last but not least, also the residues of leached iron-nickel laterite ores at the former Sered' (Slovakia) hydrometallurgical plant were found to be a suitable magnetic sorbent for the removal of radionuclides and toxic elements from suspensions [11–14]. Thus, the promising field of utilizing of a stock of six million tons, which represents a local environmental problem because of the resulting spread of the so-called black dusts, and the chromium contamination of underground waters, is the application in the decontamina-

tion of soils. When using the rough physical and chemical methods of decontamination, such as attrition scrubbing and hot dilute nitric acid leaching, the biological function of soil is damaged and remediation becomes impossible [15].

The soil in vicinity of nuclear facilities is generally contaminated by caesium-137, cobalt-60, and strontium-90 and should be decontaminated down the level of radiocaesium of about 1 kBq/kg. The aim of the present work was to present a complex view on general sorption properties of the seminatural material that might be of interest for removal the fission radionuclides and heavy metals contaminants of soil. Such application would convert the residuals into value-added separation materials, which can be applied toward decontamination of local industrial and nuclear pollution sites.

## EXPERIMENTAL

The Sered' hydrometallurgical plant ores residues (SOR) are finely-powdered, the fraction below 20 μm represents about 70 % of the material and 16 % is less than 2 μm with a favourable microporous structure. Specific surface (*s. s.*) was established by means of the laser analyzer SYMPATEC HELOS with the additive of 0.1 M-Na<sub>4</sub>P<sub>2</sub>O<sub>7</sub>. The principal components of SOR consist of the synthetic magnetite, which was formed

**Table 1.** Chemical Composition of SOR

| Component           | $w_i/\text{mass } \%$ | Component | $w_i/\text{mass } \%$ |
|---------------------|-----------------------|-----------|-----------------------|
| Fe <sub>total</sub> | 45.9                  | Mn        | 0.27                  |
| Fe <sub>metal</sub> | 1.32                  | Cr        | 1.44                  |
| Fe <sup>2+</sup>    | 17.3                  | Ti        | 0.22                  |
| Fe <sup>3+</sup>    | 26.9                  | K         | 0.11                  |
| SiO <sub>2</sub>    | 15.0                  | Na        | 0.23                  |
| Al                  | 2.54                  | Ni        | 0.27                  |
| Mg                  | 1.33                  | Co        | 0.06                  |
| Ca                  | 2.53                  |           |                       |

**Table 2.** Magnetic and Nonmagnetic Fractions of SOR

| Fraction    | $w/\text{mass } \%$ | Volume magnetic susceptibility $\kappa$ |
|-------------|---------------------|---|
| Feed        | 100.00              | 0.146                                   |
| Magnetic    | 95.91               | 0.147                                   |
| Nonmagnetic | 4.09                | 0.150                                   |

by the annealing of goethite or hematite of the original ore, and the silicate phase of dried silicate aggregates.

Chemical composition of SOR samples established by the atomic absorption spectrometer Spectr AA-30 Varian, and Fe being determined by titrimetry, is given in Table 1.

Volume magnetic susceptibility  $\kappa$  was measured before and after particle separation in the field of magnetic flux density 0.1 T by a KAPPABRIDGE KLY-2 (Geofyzika Brno) device at the magnetic field strength 300 A m<sup>-1</sup> of homogeneity 0.2 % and at the frequency 920 Hz (Table 2).

Mineralogical composition was derived from the diffractometric data (DRON 20 and a goniometer GUR-5 (Techsnabexport, Russia)) at the FeK $\alpha$  line ( $\lambda = 0.1937$  nm),  $U = 24$  kV and 10 mA current. The main isoline of the magnetite sample under investigation was 0.2516 nm (theoretical 0.2532 nm), *i.e.* closer to maghemite ( $\gamma\text{-Fe}_2\text{O}_3$ , 0.252 nm), however SOR exhibited an excess of Fe<sup>2+</sup> in form of wüstite and subsequent substitution of Fe<sup>2+</sup> by the cations Mg<sup>2+</sup>, Mn<sup>2+</sup>, Ni<sup>2+</sup>, and Co<sup>2+</sup> (Table 3).

### Material Activation

50 g of sorbent was preconditioned by 1 week contact with 50 cm<sup>3</sup> of acetate buffer of pH 4.6 (0.1 M acetic acid and 0.1 M sodium acetate,  $\varphi_r = 1:1$ ). Before sorption experiments a necessary amount of sorbent was washed by demineralized water and dried by air stream on a glass filter for 15 min. This material is indicated as SOR-0.

Grinding was performed in thick suspensions of SOR in water or 5 % K<sub>4</sub>Fe(CN)<sub>6</sub> · 3H<sub>2</sub>O solution by:

1. Vibrating mill (VM-1 KSMH-Hranice) – SOR-I.
2. Ball mill (Fritsch Pulverisette) – SOR-II.
3. Attrition mill (Attritor Netzsch LME075) – SOR-III.

The grain size distribution was obtained by a laser analyzer SYMPATEC HELOS for the samples dispersed in water.

### Sorption

Anal. grade chemicals and deionized water were used for solution preparations. Caesium-137 (Polatom, Otwock Swierck, Poland), strontium-85 (Czech Metrological Institute Prague), and europium-152 stock solutions were used to prepare 10<sup>-6</sup>–5 × 10<sup>-2</sup> mol dm<sup>-3</sup> solutions of [<sup>137</sup>Cs]CsCl, [<sup>85</sup>Sr]SrCl<sub>2</sub> or [<sup>152</sup>Eu](NO<sub>3</sub>)<sub>3</sub>. The Pb-212 radionuclide in the form of nitrate salt was used as a tracer for lead. Pb-212 was received from emanation source Th-228, which was separated from mother radionuclide U-232 by liquid extraction using 3-(2-thenoyl)-1,1,1-trifluoroacetone. Tc-99m was obtained from Mo-99–Tc-99m generator (Amersham, England).

Gamma activity of 0.1–1 cm<sup>3</sup> samples was measured by a NaI(Tl) scintillation detector in connection with a single-channel amplitude analyzer NP 420 (MEEV, Hungary). The activity of Pb-212 was determined by radiometry using a NaI(Tl) well-type detector (gamma spectrometric system, Packard 5330) after reaching equilibrium with the daughter radionuclides for Pb-212.

Sorbed amount  $R$  and distribution ratios  $D$  of the elements under investigation were measured in 7 cm<sup>3</sup> polypropylene tubes. Samples of 40–60 mg of SOR were contacted with 5 cm<sup>3</sup> of solution of initial volume activity  $a_0$  and after 2 h the sorbent was separated by sedimentation or magnetically.

From the volume activity  $a$  of resulting solution the percentage of sorption was calculated as

$$R = \frac{a_0 - a}{a_0} 100 = \left(1 - \frac{A}{A_0}\right) 100 \quad (1)$$

together with its combined standard uncertainty

$$\delta_R^2 = s_R^2 / R^2 = s_0^2 / A_0^2 + s_1^2 / A^2 + 2\delta_v^2 \quad (2)$$

Then, the distribution ratio is

$$K_d = \frac{a_0 - a}{a} \frac{V}{m} = \left(\frac{A_0}{A} - 1\right) \frac{V}{m} = \frac{R}{100 - R} \frac{V}{m} \quad (3)$$

Kinetics of sorption at the batch conditions was followed at continuous mixing in 100 cm<sup>3</sup> Erlenmeyer flask at 22 ± 2 °C, starting with 20 cm<sup>3</sup> of solution and 200 mg of SOR. 1 cm<sup>3</sup> aliquots were taken at time ( $t/h$ ) 0, 0.25, 0.5, 1, 1.5, 2, 4, 6, 8, 24, 48, and 72. Considering the balance of activity

$$A_0 = a_0 V_0 = a_i V_i + \sum a_i v + a_{si} m \quad (4)$$

the fraction of sorption at respective  $i$ -th ( $i > 1$ ) point was calculated as

$$R_i = \frac{a_0 V_0 - a_i V_i - \sum_1^{i-1} a_i v}{a_0 V_0} \quad (5)$$

or

$$R_i = \left[ 1 - \frac{1}{a_0 V_0} \left( a_i V_i + v \sum_1^{i-1} a_i \right) \right] 100 \quad (6)$$

Sorption isotherms were established by measurement of fraction of sorption at different initial concentration of carriers,  $c_0/(\text{mol dm}^{-3}) = 10^{-6}, 10^{-5}, 10^{-4}, 3 \times 10^{-4}, 6 \times 10^{-4}, 10^{-3}, 3 \times 10^{-3}, 6 \times 10^{-3}, 10^{-2}$ , and  $5 \times 10^{-2}$  on the background of 0.001 M-KCl.

Speciation of the radionuclides sorbed on SOR was performed according to the leaching procedures by *Tessier* [16, 14] and BCR (Community Bureau of Reference of the Commission of the European Communities) scheme [14, 17, 18].

The SigmaPlot<sup>®</sup> 4.01 (SPSS Inc., USA) nonlinear regression (fitting) program based on the Newton—Levenberg algorithm was used for data treatment and evaluation. Activity coefficients were calculated by the Ionic Strength Corrections Specific Interaction Theory (SIT) Version 1.2<sup>®</sup> 1.2 program (Academic Software, United Kingdom).

## RESULTS AND DISCUSSION

The kinetics of sorption is complex, having slow and fast steps on the corresponding sorption sites of SOR, but only the fast component of sorption (half-time 0.03—0.5 h [14]) was investigated from the practical point of view. The long-term component of equilibrium (half-times of sorption 17—70 h) which provides maximal sorption is established only after several days and therefore is purposeless for practical applications in a feasible sorption-desorption cycle. Nevertheless, it should be remarked that in such cases about 5 % of the gross sorption capacity for caesium, 90 % of that of strontium, and 55 % of europium is available [14].

The equilibrium data for sorption isotherms were obtained as

$$c = a c_0 / a_0 \quad (7)$$

and

$$\Gamma = (c_0 - c)V/m = c_0(1 - a/a_0)V/m \quad (8)$$

Basically, the Freundlich type isotherm

$$\Gamma = kc^n \quad (9)$$

is applied for sorption on heterogeneous surfaces like SOR and soils [14, 19]. However, the isotherm does not respect limited distribution ratios of sorbate at infinitesimal concentration, and therefore a related Toth isotherm [20]

$$\Gamma = \frac{k_1 c}{(k_2 + c^n)^{1/n}} \quad (10)$$

was applied. The substantial advantage of the isotherm is that it combines both Henry's and Langmuir isotherms at the limits of available concentration interval, and Freundlich isotherm in-between. Therefore it enables not only to consider the distribution constant at zero concentration of sorbate as

$$K_d = \lim_{c \rightarrow 0} \frac{\Gamma}{c} = \frac{k_1}{k_2^{1/n}} \quad (11)$$

but also evaluate the sorption capacity of sorbent

$$\Gamma_\infty = \lim_{c \rightarrow \infty} \Gamma = k_1 \quad (12)$$

which is an important feature for the explanatory evaluation of sorbent activation.  $K_d$  data obtained experimentally at infinitesimal concentration of sorbate (non-carrier-added radionuclide) were used to replace  $k_1$  in eqn (10) by

$$k_1 = K_d k_2^{1/n} \quad (13)$$

issuing from eqn (11). Then the isotherm parameters  $k_2$  and  $n$  were calculated by the nonlinear regression. The obtained parameters are in Tables 4 and 5 with the standard uncertainty at 68 % confidence level. The fitted data (Figs. 1 and 2) passed both the ANOVA and Durbin—Watson statistics tests.

It was supposed that during the grinding of SOR in ferrocyanide solutions some mechanochemical activation [21] occurs in respect of caesium due to the forma-

**Table 3.** Mineralogical Composition of SOR

| Phase   | $w_i/\text{mass } \%$ | Phase                            | $w_i/\text{mass } \%$ |
|---|-----------------------|----------------------------------|-----------------------|
| Magnetite ( $\text{Fe}_{0.75}\text{Mg}_{0.20}\text{Mn}_{0.02}\text{Ni}_{0.02}\text{Co}_{0.005}$ ) · $\text{Fe}_2\text{O}_3$ | 54.09                 | Quartz $\alpha\text{-SiO}_2$     | 13.15                 |
| Wüstite FeO   | 8.02                  | Calcite $\text{CaCO}_3$          | 6.32                  |
| Metallic iron $\alpha\text{-Fe}$  | 1.32                  | Corundum $\text{Al}_2\text{O}_3$ | 2.72                  |
| Ferrochrompicotite $\text{FeO} \cdot (\text{Cr}, \text{Al})_2\text{O}_3$  | 5.51                  | Na-Feldspar                      | 2.62                  |
|   |                       | K-Feldspar                       | 0.78                  |

**Table 4.** The Parameters of Toth Sorption Isotherm (Fig. 1) of Various Ions from 0.001 M-KCl

| Ion              | $K_d$      | $k_1$                        | $k_2$         | $n$         |
|------------------|------------|------------------------------|---------------|-------------|
| Cs <sup>+</sup>  | 49.5 ± 2.5 | 1.4 ± 1.0                    | 0.40 ± 0.07   | 0.26 ± 0.02 |
| Sr <sup>2+</sup> | 155 ± 7.8  | (5.7 ± 15) × 10 <sup>4</sup> | 2.09 ± 0.71   | 0.13 ± 0.03 |
| Pb <sup>2+</sup> | 1680 ± 80  | 0.06 ± 0.10                  | 0.011 ± 0.008 | 0.44 ± 0.07 |
| Eu <sup>3+</sup> | 32.5 ± 1.6 | 0.03 ± 0.14                  | 0.51 ± 0.26   | 0.10 ± 0.03 |

**Table 5.** The Parameters of Toth Sorption Isotherm (Fig. 2) of Caesium from 0.001 M-KCl on Original and Activated Sorbent

| Sorbent | $K_d$      | $k_1$         | $k_2$         | $n$         | $K_d$ relative |
|---------|------------|---------------|---------------|-------------|----------------|
| SOR-0   | 49.5 ± 2.5 | 1.4 ± 1.0     | 0.40 ± 0.07   | 0.26 ± 0.02 | 1.0            |
| SOR-I   | 1670 ± 870 | 0.057 ± 0.082 | 0.094 ± 0.029 | 0.23 ± 0.02 | 34             |
| SOR-II  | 77.3 ± 3.9 | 0.19 ± 0.24   | 0.16 ± 0.06   | 0.30 ± 0.03 | 1.6            |
| SOR-III | 813 ± 41   | 0.028 ± 0.044 | 0.061 ± 0.026 | 0.27 ± 0.03 | 16             |

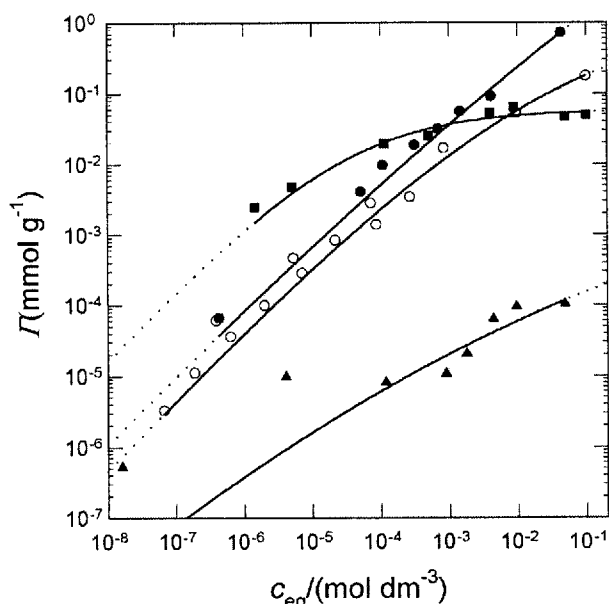
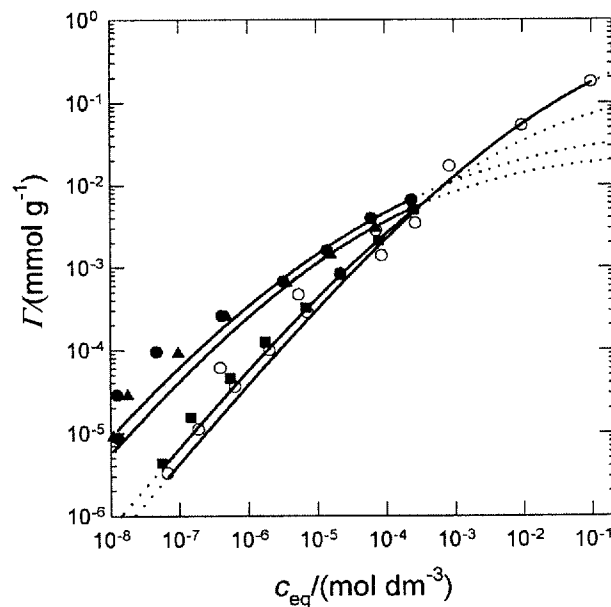
tion of the surface film of the insoluble nickel ferrocyanide, which is the specific sorbent for caesium [22—25].

From the data it issues that the change of SOR sorption properties appears during grinding procedure due to the nickel and other heavy ions ferrocyanide formation on the surface and not due to the specific surface increase (Fig. 3, Table 5). The modelled values of sorption capacity ( $k_1$ ) are determined with a high degree of uncertainty, due to the extrapolation character of this parameter fitting.

As expected, dispersion of SOR increased during the disintegrating procedures, distinctively at attrition mill-

ing when the specific surface is practically tripled (Fig. 3, Table 6).

The increase of dispersion is not reflected in the sorption capacity but the grinding results in qualitative change of surface identified with distribution ratios of caesium (Table 5). The activation exhibits on a small fraction (5—15 %) of available surface as prompted by the observed low  $k_1$  and high  $K_d$ . However, the virtually extrapolated  $k_1$  value is evidently of the high degree of uncertainty. The most efficient seems the activation of surface by grinding in vibrating mill (SOR-I) which gives specific surface increase for only about 25 % (Table 6). Reduction of heterogeneity of the surface, ex-

**Fig. 1.** Sorption isotherms of Cs<sup>+</sup> (○), Sr<sup>2+</sup> (●), Pb<sup>2+</sup> (■), and Eu<sup>3+</sup> (▲) ions on SOR.**Fig. 2.** The sorption isotherms of Cs<sup>+</sup> on SOR-0 (○), SOR-I (●), SOR-II (■), and SOR-III (▲) sorbent.

**Table 6.** Dispersion of SOR Samples

| Material | Grinder        | Time/min | Mass yield of undersizes/% |         |         | Specific surface                             |                |
|----------|----------------|----------|----------------------------|---------|---------|--|----------------|
|          |                |          | < 10 μm                    | < 40 μm | < 63 μm | $\frac{10^{-4} \text{ s.s.}}{\text{m}^{-1}}$ | Relative s. s. |
| SOR-0    | None           | 0        | 42.34                      | 79.47   | 91.90   | 1.31   | 1.00           |
| SOR-I    | Vibrating mill | 2        | 57.50                      | 86.71   | 94.85   | 1.65   | 1.26           |
| SOR-II   | Ball mill      | 15       | 63.55                      | 99.24   | 100.00  | 2.19   | 1.67           |
| SOR-III  | Attrition mill | 15       | 99.79                      | 100.00  | 100.00  | 3.86   | 2.95           |

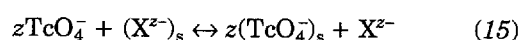
pressed in low exponent  $n$  value, seems also unimportant in the process (Table 5). Thus, the vibrating mill activation appears as the most efficient when the caesium adsorption is concerned.

The specific behaviour of strontium and lead, *i.e.* the extreme sorption capacity  $k_1$  for strontium and also very high distribution ratio  $K_d$  of lead in infinite diluted solutions (Table 2) may be caused by their dominating sorption on carbonate fraction of the SOR. This was proved to be consistent with the data on chemical speciation of the ions.

Sorption of pertechnetate anion,  $\text{TcO}_4^-$ , was investigated as a model for anion sorption and the data were treated according to the possible ion-exchange mechanism [26]. Activity coefficients were calculated in 1 : 1 electrolytes by the specific interaction theory (SIT), considering that the parameters of  $\text{TcO}_4^-$  were the same as those for  $\text{ClO}_4^-$ . For the 1 : 2 or 2 : 1 electrolyte single-ion (of charge  $z$ ) activity coefficients were calculated from the Davies equation [27]

$$\log\{\gamma\} = -0.5z^2 \left( \frac{\sqrt{I}}{1 + \sqrt{I}} - 0.3\{I\} \right) \quad (14)$$

For ion exchange of  $\text{TcO}_4^-$  with an anion  $\text{X}^{z-}$ , thermodynamic constant  $K$  of the equilibrium

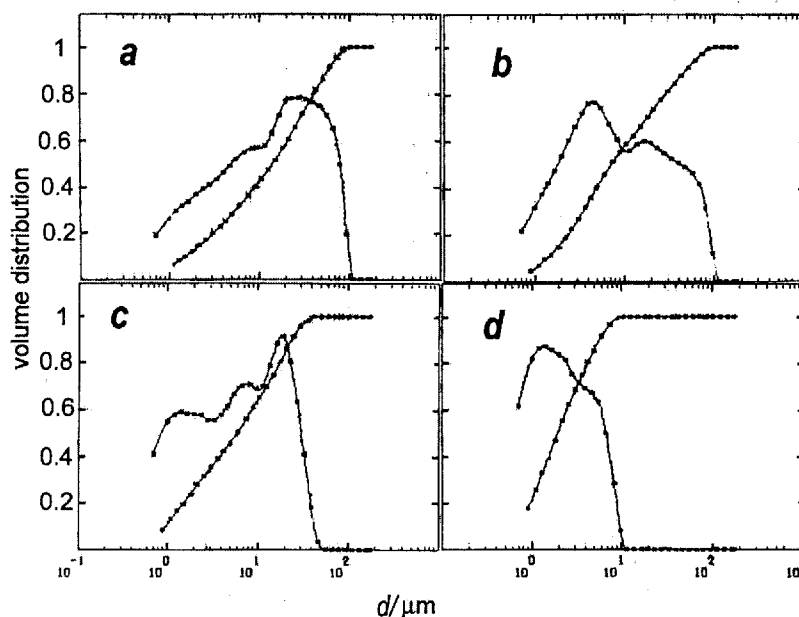


(subscript indicates solid phase) is

$$K = \frac{a_s^z a_x}{a_{xs} a^z} \quad (16)$$

or, when the ratio of the solid phase activity coefficients is close to one,

$$K = D^z \frac{a_x}{\gamma^z} \quad (17)$$



**Fig. 3.** Grain size distribution of SOR-0 (a), SOR-I (b), SOR-II (c), and SOR-III (d).

**Table 7.** The Ion Exchange Characteristics of  $\text{TcO}_4^-$  Ion on SOR-0 from 0.1 M Electrolyte

| Electrolyte                | Experim. | $a_X$  | $a_M$  | $\gamma$ |          | $\gamma^2/a_X$ | $K$      |
|----------------------------|----------|--------|--------|----------|----------|----------------|----------|
|                            | $D$      |        |        | SIT      | Eqn (14) |                | Eqn (17) |
| HCl                        | 0.52     | 0.080  | 0.080  | 0.803    | 0.785    | 9.83           | 0.053    |
| NaCl                       | 0.17     | 0.078  | 0.078  | 0.794    | 0.785    | 10.00          | 0.017    |
| $\text{NaNO}_3$            | 0.14     | 0.077  | 0.070  | 0.774    | 0.785    | 10.20          | 0.014    |
| $\text{Ca}(\text{NO}_3)_2$ | 0.16     | 0.120* | 0.013* | —        | 0.738    | 6.15           | 0.026    |
| $\text{Na}_2\text{CO}_3$   | 0.15     | 0.129* | 0.120* | —        | 0.738    | (42)           | (0.0005) |

\*According to the Davies equation.

Results (Table 7) support the ion-exchange mechanism of pertechnetate sorption only partially: Higher value for HCl solutions may be caused by partial reduction of Tc(VII), which occurs during precipitation of magnetite [28]. Also, the behaviour in  $\text{Na}_2\text{CO}_3$  is clearly different, which may be caused by a rough approximation of activity coefficients or due to a specific carbonate leaching action on the sorbent. The thermodynamic constant of exchange for 1 : 1 electrolytes is  $\log K = -1.62 \pm 0.010$ . The adsorption should be interpreted as the secondary ion exchange in the outer electrostatic double layer of sorbent [29].

Investigation of specific leaching of the radionuclides after sorption on magnetic sorbent indicates a striking difference in the radionuclides behaviour. While major fraction of strontium is in an exchangeable form, both caesium and europium are predominantly bound by the oxide component of SOR (Table 8). The unusually high  $K_d$  of lead may be caused by a noninvestigated sulfide fraction of SOR.

Generally, SOR as a sorbent of moderate specificity towards caesium can be used, but its recycling is still an open question, considering the secondary waste (contaminated sorbent, leaching solution, etc.) production. *E.g.*, solvent extraction of radiocaesium by cobalt dicarbonyl can be considered from acid desorption solutions [15], but still an economically feasible desorption agent for utilized magnetic sorbent should be found on the basis of creating a regeneration flowsheet.

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## SYMBOLS

|              |   |                                  |
|--------------|---|----------------------------------|
| $A$          | activity (or count rate)  | $\text{s}^{-1}$                  |
| $A_0$        | gross activity (or count rate) of indicator                               | $\text{s}^{-1}$                  |
| $A_m$        | activity (or count rate) of contacted sorbent                             | $\text{s}^{-1}$                  |
| $a_M$        | thermodynamic activity of cation M  |                                  |
| $a_X$        | thermodynamic activity of anion X   |                                  |
| $a$          | volume activity (or count rate) of solution<br>( $a = A/V$ )              | $\text{cm}^{-3} \text{s}^{-1}$   |
| $a_0$        | volume count rate of initial solution                                     | $\text{cm}^{-3} \text{s}^{-1}$   |
| $a_i$        | volume count rate of the $i$ -th sample<br>of solution                    | $\text{cm}^{-3} \text{s}^{-1}$   |
| $a_{si}$     | specific (mass) count rate of sorbent at<br>the $i$ -th sample withdrawal | $\text{g}^{-1} \text{s}^{-1}$    |
| $c$          | concentration   | $\text{mol dm}^{-3}$             |
| $D$          | distribution ratio at sorption  | $\text{dm}^3 \text{kg}^{-1}$     |
| $I$          | ionic strength  | $\text{mol dm}^{-3}$             |
| $k$ or $k_2$ | constant of the Freundlich or Toth<br>isotherm, respectively              | $\text{mol}^{-n} \text{dm}^{3n}$ |
| $k_1$        | constant of Toth isotherm   | $\text{mol kg}^{-1}$             |

**Table 8.** Sequential Leaching Speciation of the Radionuclides Sorbed on SOR-0

| Fraction                    | Leached fraction/% |               |                   |               |                   |               |
|-----------------------------|--------------------|---------------|-------------------|---------------|-------------------|---------------|
|                             | Cs                 |               | Sr                |               | Eu                |               |
|                             | Tessier<br>scheme  | BCR<br>scheme | Tessier<br>scheme | BCR<br>scheme | Tessier<br>scheme | BCR<br>scheme |
| Exchangeable and carbonates | 53 ± 5             | 14 ± 4        | —                 | 90 ± 4        | 14 ± 3            | 18 ± 2        |
| Reducible oxides            | 29 ± 3             | 78 ± 9        | —                 | 9 ± 1         | 79 ± 1            | 71 ± 1        |
| Organic and sulfides        | 15 ± 2             | 5.5 ± 0.2     | —                 | 0.6 ± 0.2     | 5 ± 1             | 8 ± 1         |

|          |  |                               |
|----------|--|-------------------------------|
| $K$      | thermodynamic constant of ion exchange   |                               |
| $K_d$    | distribution ratio   | $\text{dm}^3 \text{kg}^{-1}$  |
| $m$      | sorbent mass   | $\text{g}$                    |
| $n$      | exponent of the Freundlich or Toth isotherm  |                               |
| $R$      | equilibrium percentage ( $0 \leq R \leq 100$ ) or fraction ( $0 \leq R \leq 1$ ) of sorption |                               |
| $R_i$    | sorption fraction at the withdrawal of the $i$ -th sample                                    |                               |
| $r$      | solid to liquid phase ratio  | $\text{kg dm}^{-3}$           |
| $r_i$    | solid to liquid phase ratio at the withdrawal of the $i$ -th sample                          | $\text{kg dm}^{-3}$           |
| SOR      | "Sered' Ore Residuals"   |                               |
| $s$      | standard uncertainty (the same as the base unit)   |                               |
| $V$      | aqueous phase volume   | $\text{cm}^3$                 |
| $V_i$    | aqueous phase volume at the withdrawal of the $i$ -th sample                                 | $\text{cm}^3$                 |
| $v$      | aqueous phase sample volume (aliquots)   | $\text{cm}^3$                 |
| $t_i$    | contact time   | $\text{h}$                    |
| $X$      | competing ion  |                               |
| $x_r$    | fraction of surface with a rapid sorption kinetics   |                               |
| $x_s$    | fraction of surface with a slow sorption kinetics  |                               |
| $z$      | charge of competing ion $X$  |                               |
| $\Gamma$ | concentration of element in sorbent  | $\text{mol kg}^{-1}$          |
| $\gamma$ | thermodynamic ionic activity coefficient of $\text{TcO}_4^-$ anion                           | $\text{dm}^3 \text{mol}^{-1}$ |
| $\alpha$ | fraction of a specific form of element in solid phase  |                               |
| $\delta$ | relative uncertainty   |                               |
| $\kappa$ | volume magnetic susceptibility   |                               |

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