Electronic Supplementary Information

Magnetic Nanocomposites based on Zn,Al-LDH intercalated with Citric and EDTA groups for the removal of U(VI) from environmental and wastewater: Synergistic Effect and Adsorption Mechanism Study

Natalia Kobylinska*, Liubov Puzyrnaya, Galina Pshinko
Dumansky Institute of Colloid and Water Chemistry,
National Academy of Sciences of Ukraine,
42 Akad. Vernads'koho Blvd., Kyiv, 03142, Ukraine

Corresponding author: Dr. Natalia Kobylinska

e-mail: kobilinskaya@univ.kiev.ua

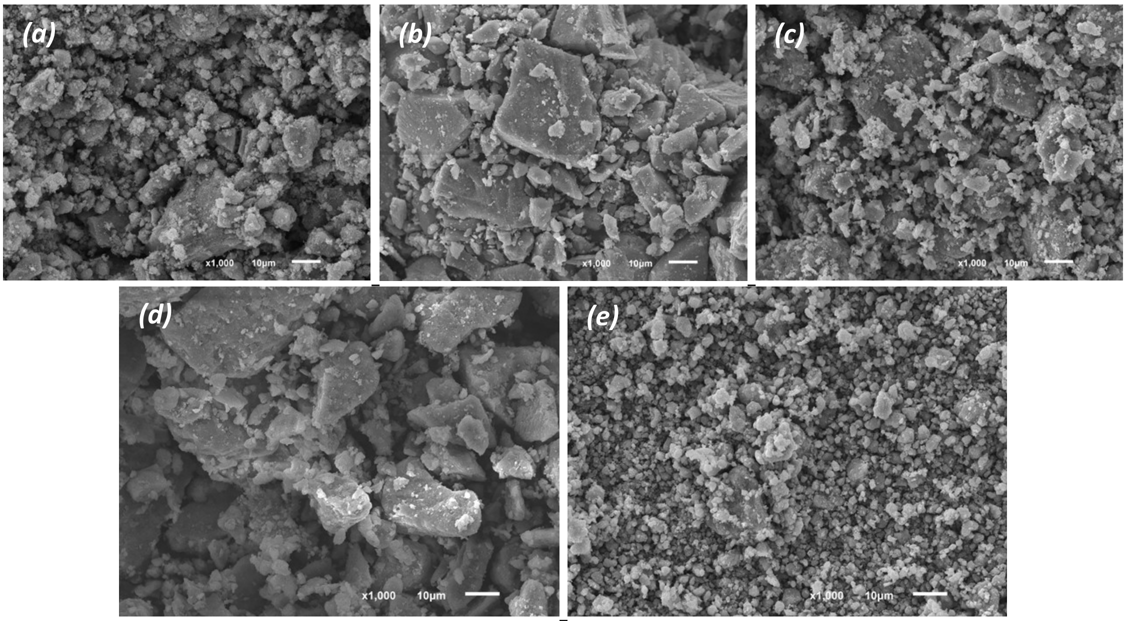

Fig. S1. SEM images of bare *Fe_3O_4* (a), *Zn,Al-LDH/Cit* (b), *Fe_3O_4/Zn,Al-LDH/Cit* (c), *Zn,Al-LDH/EDTA* (d) and *Fe_3O_4/Zn,Al-LDH/EDTA* (e) samples.

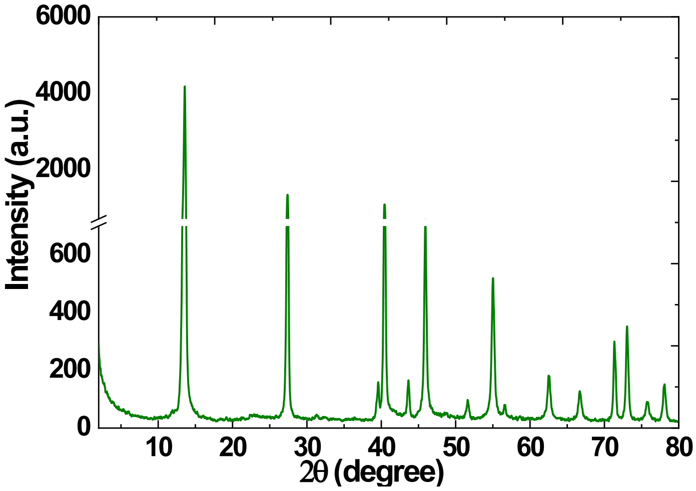

Fig. S2. XRD pattern of initial Zn,Al-LDH/CO_3 sample.

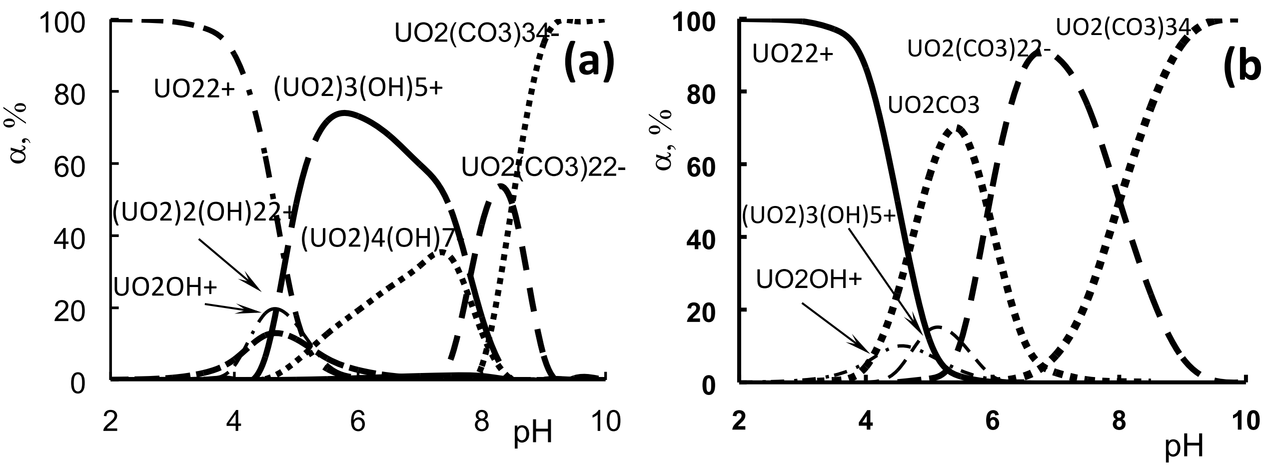

Fig. S3. The effect pH on the distribution of U(VI) forms in aqueous solution containing various concentrations of CO_3^{2-}ions: 0.01 mmol/L (a) i 5.0 mmol/L (b). (*Conditions*: C(U(VI)) = 0.1 mmol/L, calculated with MEDUZA).

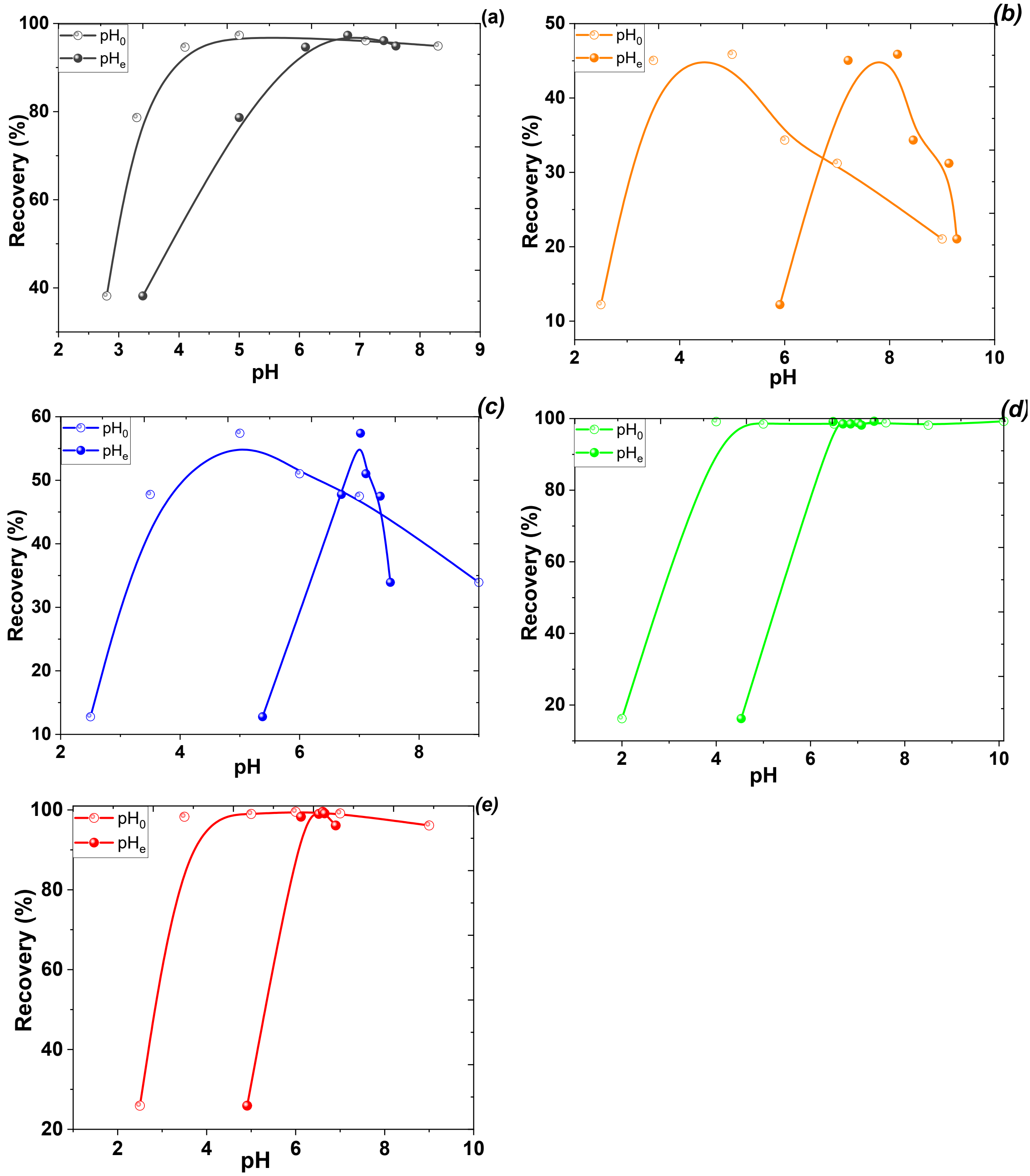

Fig. S4. Effect of initial *(*pH_0*, a)* and equilibrium (pH_e, b) pH on U(VI) removal by obtained samples: prictine *Fe_3O_4* (a), *Zn,Al-LDH/Cit* (b), *Fe_3O_4/Zn,Al-LDH/Cit* (c), *Zn,Al-LDH/EDTA* (d) and *Fe_3O_4/Zn,Al-LDH/EDTA* (*Conditions*: C_0(U(VI)) = 0.1 mol/L, V/m = 500 cm^3/g, time 1 h).

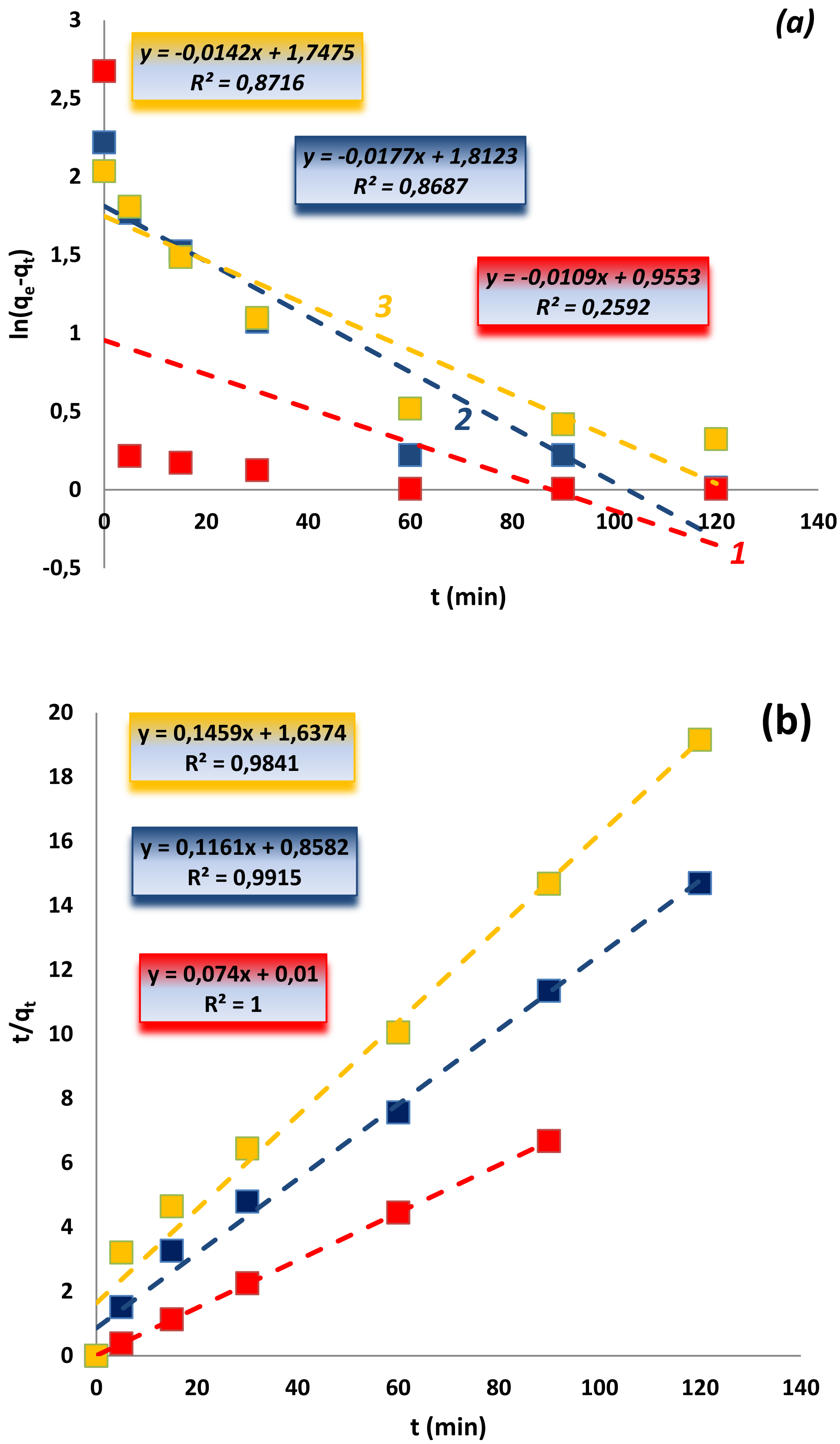

Fig. S5. Pseuvdo-first-order (a) and pseuvdo-second-order (b) kinetic plots for Uu(VI) adsorption onto *Fe_3O_4/Zn,Al-LDH/EDTA* (1), *Fe_3O_4/Zn,Al-LDH/Cit* (2) and *Zn,Al-LDH/Cit* (3).

Table. S1. Equilibrium constants of U(VI) with Citrate and EDTA ligands in aqueous solution [1].

Equilibrium reactions	$\log_{10}\beta$	Remarks/Conversations
$(U(VI)O_2)^{2+} + (EDTA)^{4-} \leftrightarrow U(VI)O_2EDTA^{2-}$	10.9028	Original data for β: $\log_{10}\beta$ =9.28, at I=1.0 M
$H^+ + (U(VI)O_2)^{2+} + (EDTA)^{4-} \leftrightarrow$ $(U(VI)O_2)H(EDTA)^-$	19.62945	$(UO_2) + HL \leftrightarrow (UO_2)HL$ $\log_{10}\beta$ = 7.40, I=1.0 M $H + L \leftrightarrow HL$ $\log_{10}\beta$ = 10.09370, I=1.0 M $(UO_2) + H + L \leftrightarrow (UO_2)HL$ $\log_{10}\beta$ = 17.49370, I=1.0 M I = 0 M: $\log_{10}\beta$ = 19.62945
$2(U(VI)O_2)^{2+} + (EDTA)^{4-} \leftrightarrow (U(VI)O_2)_2EDTA$ *(aq)*	20.43290	Original data for β: $\log_{10}\beta$ =17.87, at I=1.0 M
$2(U(VI)O_2)^{2+} + (OH)^- + (EDTA)^{4-} \leftrightarrow$ $(U(VI)O_2)_2EDTA(OH)^-$	29.41674	$(UO_2)_2L \leftrightarrow (UO_2)_2(OH)L + H$ $\log_{10}\beta$ = -4.81, I=1.0 M $2(UO_2) + L \leftrightarrow (UO_2)_2L$ $\log_{10}\beta$ =17.99498, I=1.0 M $OH + H \leftrightarrow H_2O$ $\log_{10}\beta$ =13.79384, I=1.0 M $2(UO_2) + L \leftrightarrow (UO_2)_2L_2$ $\log_{10}\beta$ =26.97882, I=1.0 M I = 0 M: $\log_{10}\beta$ = 29.41674
$2(U(VI)O_2)^{2+} + 2(EDTA)^{4-} \leftrightarrow$ $(U(VI)O_2)_2(EDTA)_2^{4-}$	29.33290	$(UO_2)_2L + L \leftrightarrow (UO_2)_2L_2 + H$ $\log_{10}\beta$ = 8.90, I=1.0 M $2(UO_2) + L \leftrightarrow (UO_2)_2L$ $\log_{10}\beta$ =17.99498, I=1.0 M $2(UO_2) + 2L \leftrightarrow (UO_2)_2L_2$ $\log_{10}\beta$ =26.89498, I=1.0 M I = 0 M: $\log_{10}\beta$ = 29.33290
$4(U(VI)O_2)^{2+} + 4OH^- + 2(EDTA)^{4-} \leftrightarrow$ $(U(VI)O_2)_6(OH)_4(EDTA)_2^{4-}$	74.17224	$4(UO_2) + 2L \leftrightarrow (UO_2)_2(OH)_4L_2 + 4H$ $\log_{10}\beta$ = 15.34, I=1.0 M $4OH + 4H \leftrightarrow 4H_2O$ $\log_{10}\beta$ =55.17536, I=1.0 M $4(UO_2) + 2L + 4OH \leftrightarrow (UO_2)_4L_2$ $\log_{10}\beta$ =70.5153, I=1.0M I = 0 M: $\log_{10}\beta$ = 74.17224
$6(U(VI)O_2)^{2+} + 4OH^- + 3(EDTA)^{4-} \leftrightarrow$ $(U(VI)O_2)_6(OH)_4(EDTA)_3^{4-}$	95.57016	$6(UO_2) + 3L \leftrightarrow (UO_2)_6(OH)_4L_3$+4H $\log_{10}\beta$ = 34.3, I=1.0M $4OH + 4H \leftrightarrow 4H_2O$ $\log_{10}\beta$ =55.17536, I=1.0 M $6(UO_2) + 3L + 4OH \leftrightarrow (UO_2)_6(OH)_4L_3$ $\log_{10}\beta$ =89.5, I=1.0M I = 0 M: $\log_{10}\beta$ = 95.57016
$(U(VI)O_2)^{2+} + (Citrate)^{3-} \leftrightarrow (U(VI)O_2)(Citrate)^-$	8.68145	Original data for β: $\log_{10}\beta$ =7.4, at I=0.1 M
$2(U(VI)O_2)^{2+} + 2(Citrate)^{3-} \leftrightarrow$ $(U(VI)O_2)_2(Citrate)_2^{2-}$	21.21933	Original data for β: $\log_{10}\beta$ = 18.87, at I=0.1 M

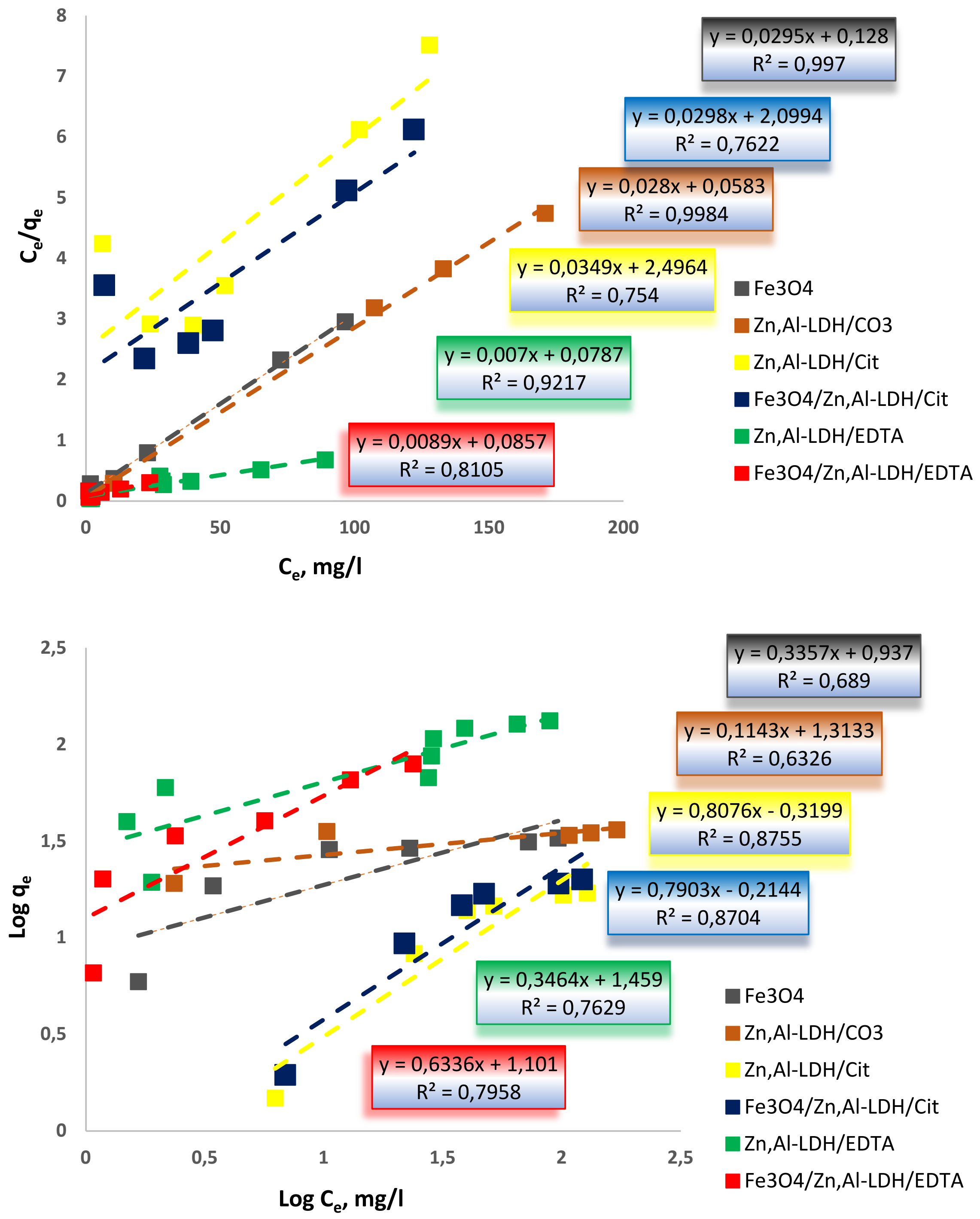

Fig. S6. Linear fitting of adsorption isotherms with Langmuir *(a)* and Freundlish *(b)* equations for U(VI) on obtained samples.

Reference

1. Wilko Verweij, Jean-Pierre Simonin. Implementing the Mean Spherical Approximation Model in the Speciation Code CHEAQS Next at High Salt Concentrations. Journal of Solution Chemistry **2020,** *49* (11), 1319-1327. https://doi.org/10.1007/s10953-020-01008-9.