

SUPPORTING INFORMATION

Enhanced Arsenic Removal from Aqueous Solutions Via Magnesium Hydroxide Coated Iron Nanoparticles

Ibrahim Maamoun^{1,*} | Omar Falyouna² | Islam Mir Shariful^{3,4} |
 Ramadan Eljamal⁵ | Khaoula Bensaida² | Kazuya Tanaka¹ |
 Kohei Tokunaga^{1,6} | Osama Eljamal^{2,*}

¹Advanced Science Research Center, Japan Atomic Energy Agency, Ibaraki, 319-1195, Japan

²Water and Environmental Engineering Laboratory, Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Fukuoka, 816-8580, Japan

³International Institute for Carbon-Neutral Energy Research (I2CNER), Kyushu University, Fukuoka, 819-0395, Japan

⁴Department of Oceanography, University of Dhaka, Dhaka-1000, Bangladesh

⁵Research Center for Negative Emission Technology, International Science Innovation Center, Kyushu University, Fukuoka, 819-0395, Japan

⁶Ningyo-toge Environmental Engineering Center, Japan Atomic Energy Agency, Tomata, Okayama, 708-0698, Japan

*Corresponding authors

E-mail: maamoun.ibrahim@jaea.go.jp (Ibrahim Maamoun); osama-eljamal@kyudai.jp (Osama Eljamal)

Tel.: +81-80-4278-8910; +81-80-3900-8521

Web of Science Researcher ID: Ibrahim Maamoun (AAA-7992-2021)

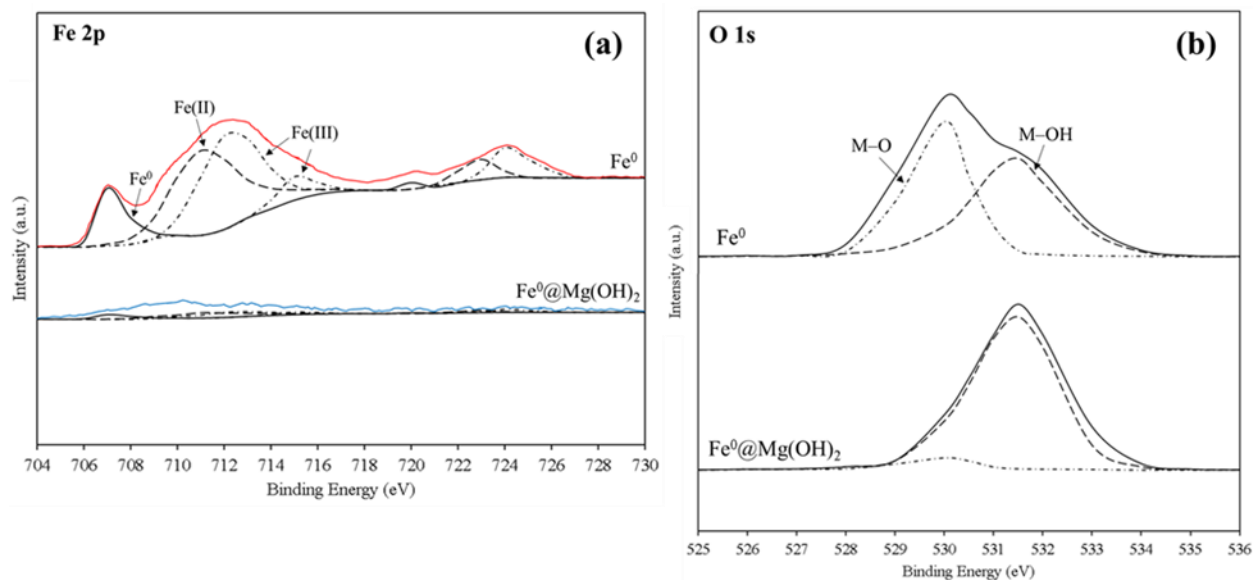


Fig. S1. XPS analysis of fresh (a) Fe 2p and (b) O 1s regions for the surface of nFe⁰ and nFe⁰@Mg(OH)₂ (100% coating ratio), adopted from [1].

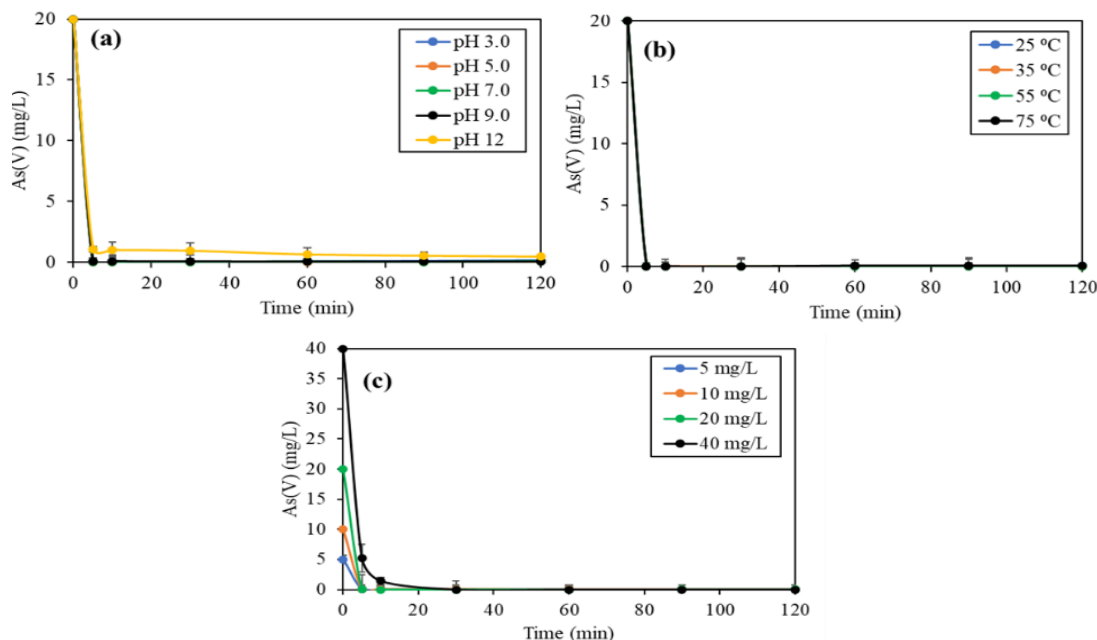


Fig. S2. Effect of initial pH (a), reaction temperature (b), and initial As(V) concentration (c) on As(V) removal by $n\text{Fe}^0/\text{Mg}(\text{OH})_2\text{-100\%}$ (1.0 g/L dosage).

Table S1. Calculated kinetics parameters for As(V) removal by $n\text{Fe}^0$ and $n\text{Fe}^0/\text{Mg}(\text{OH})_2$.

Parameter	Exp.	Pseudo-first-order model					Pseudo-second-order model				Intraparticle diffusion model				Elovich model			
		q_e (mg/g)	k_1 (1/min)	q_e (mg/g)	R^2	AIC	k_2 (g/mg min)	q_e (mg/g)	R^2	AIC	k_{int} (mg/g min ^{0.5})	C_{int} (mg/g)	R^2	AIC	β (mg/g min)	α (g/mg)	R^2	AIC
$n\text{Fe}^0$	39.891	0.270	37.642	0.993	13.161	0.013	39.442	0.998	2.071	2.912	14.99	0.778	33.71	0.304	89.452	0.988	38.34	
Mg/Fe (25%)	33.801	0.278	30.061	0.969	20.112	0.015	31.951	0.984	16.341	2.507	11.17	0.821	30.15	0.310	967.643	0.995	9.711	
Mg/Fe (50%)	38.400	0.338	35.384	0.993	12.824	0.022	36.623	0.995	11.575	2.636	15.204	0.742	33.882	0.400	743.524	0.989	36.293	
Mg/Fe (100%)	39.894	0.305	38.740	0.999	4.008	0.017	40.164	1.000	-3.644	2.833	16.620	0.736	34.991	0.383	131.876	0.981	41.626	
pH 3.0	39.961	0.508	39.492	0.999	3.093	0.048	40.218	1.000	-9.433	2.646	19.309	0.662	36.648	0.626	215.472	0.967	45.923	
pH 5.0	39.894	0.305	38.740	0.999	4.008	0.017	40.164	1.000	-3.644	2.833	16.620	0.736	34.991	0.359	549.725	0.990	37.672	
pH 7.0	38.780	0.334	36.146	0.994	12.007	0.020	37.509	0.997	7.828	2.706	15.440	0.748	34.003	0.394	783.245	0.993	36.512	
pH 9.0	39.882	0.198	35.037	0.980	19.114	0.008	37.685	0.990	14.933	3.024	11.340	0.860	30.475	0.215	159.078	0.996	9.480	
pH 12.0	37.601	0.133	33.045	0.976	19.685	0.005	36.835	0.990	14.238	3.083	8.029	0.918	26.890	0.165	23.420	0.996	9.177	
25 °C	39.894	0.305	38.740	0.999	4.008	0.017	40.164	1.000	-3.644	2.833	16.620	0.736	34.991	0.370	81.925	0.976	42.090	
35 °C	40.0	0.325	39.026	0.999	2.620	0.020	40.321	1.000	-4.301	2.805	17.176	0.722	35.361	0.413	435.180	0.986	40.773	
55 °C	40.0	0.466	39.400	0.999	1.644	0.040	40.200	1.000	-35.397	2.670	18.973	0.672	36.443	0.640	303.146	0.974	45.665	
75 °C	40.0	0.745	39.957	1.000	-25.260	0.188	40.134	1.000	-19.842	2.528	20.673	0.620	37.447	0.720	136.428	0.947	47.283	
Dosage 0.25 g/L	39.201	0.143	37.767	0.996	11.348	0.005	41.190	0.990	15.839	0.165	46.969	0.975	21.419	3.254	10.694	0.851	31.870	
Dosage 0.5 g/L	39.894	0.305	38.740	0.999	4.008	0.017	40.164	1.000	-3.644	0.383	131.92	0.981	41.625	2.833	16.620	0.736	34.991	
Dosage 1.0 g/L	40.0	1.304	40.000	1.000	-113.21	3.407	40.009	1.000	-47.674	2.590	137.42	0.955	50.065	2.491	21.043	0.607	37.657	
Dosage 2.0 g/L	40.0	2.918	40.000	1.000	-134.92	763.94	40.000	1.000	-103.04	2.576	819.32	0.968	49.833	2.489	21.063	0.607	37.668	
C_0 (5 mg/L)	9.859	0.358	9.733	0.999	-16.444	0.096	10.011	1.000	-23.734	0.682	4.434	0.702	19.080	1.715	201.457	0.986	23.748	
C_0 (10 mg/L)	19.847	0.299	19.404	0.997	1.563	0.033	20.162	1.000	-11.990	1.427	8.255	0.740	26.601	0.734	374.188	0.992	29.001	
C_0 (20 mg/L)	39.894	0.305	38.740	0.999	4.008	0.017	40.164	1.000	-3.644	2.833	16.620	0.736	34.991	0.359	554.238	0.990	37.656	
C_0 (40 mg/L)	70.200	0.187	64.571	0.992	20.863	0.004	69.281	0.995	18.591	5.424	21.165	0.842	38.451	0.118	292.593	0.991	21.507	

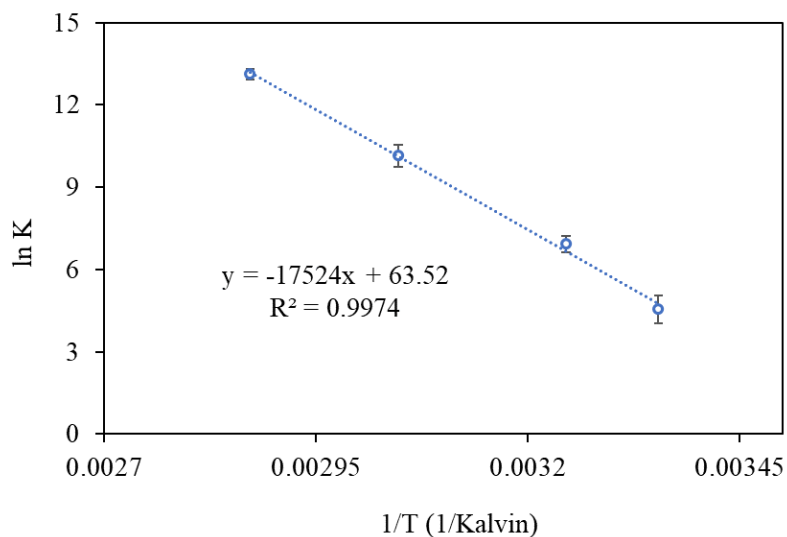


Fig. S3. Van 't Hoff plot of thermodynamic analysis for As(V) removal by nFe⁰@Mg(OH)₂-100%.

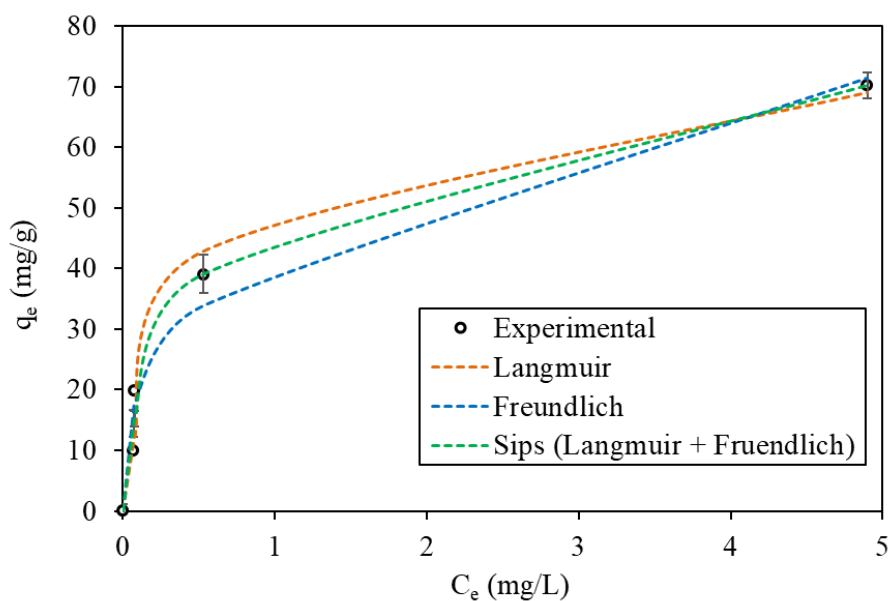


Fig. S4. Non-linear adsorption isotherms for As(V) removal by nFe⁰@Mg(OH)₂-100%.

Table S2. Separation factor calculations for Langmuir isotherm.

Initial As(V) concentration (mg/L)	R _L
0	1
5	0.072692
10	0.037717
20	0.019221
40	0.009704

Table S3. Comparison on the maximum sorption capacity of the reported iron-based materials to As(V).

Reference*	Material	Maximum sorption capacity (mg/g)
[2]	nFe ⁰	50.29**
	S- nFe ⁰	95.76**
[3]	F- nFe ⁰	90.4
[4]	N-S- nFe ⁰	125**
[5]	Fe ₃ O ₄ -reduced graphite oxide-MnO ₂	12.22
[6]	Ultrafine δ-FeOOH	37.3
[7]	Zr-doped β-FeOOH	60
[8]	Fe ₃ O ₄ nanoparticles	46.06
[9]	Fe-Cu LDH	82.7
[10]	Fe-Al LDH	37.9
[11]	Fe ₃ O ₄ coated wheat straw	8.1
[12]	Fe ₃ O ₄ /graphene	180.3**
This study	nFe ⁰ @Mg(OH) ₂	89.97

*The chosen references for the comparison were selected according to the best fitting to the reaction conditions in the present work, and **higher As(V) concentration (up to 600 mg/L).

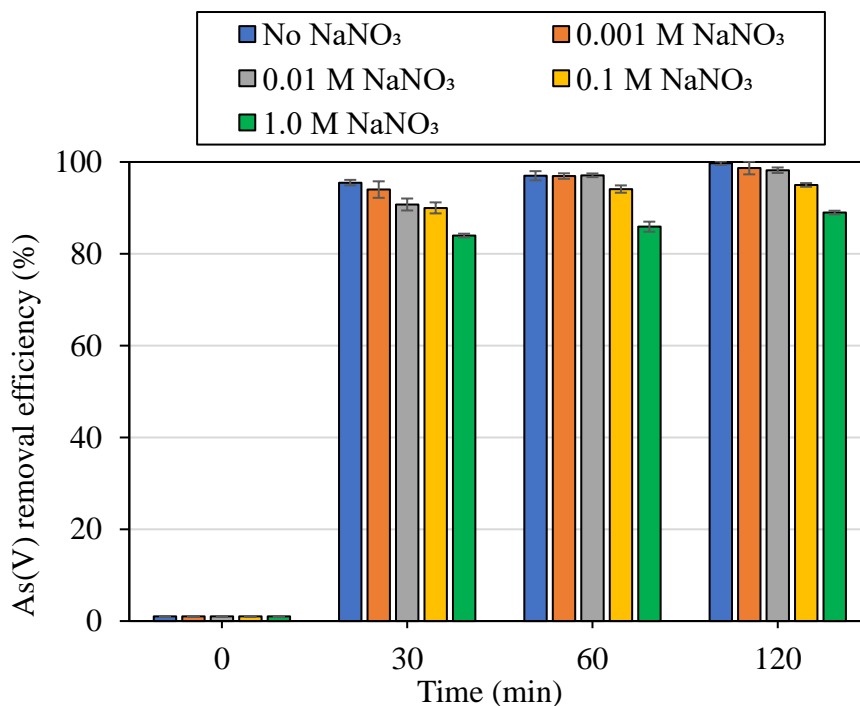


Fig. S5. As(V) removal by nFe⁰@Mg(OH)₂-100% at different ionic strengths (0.001 – 1.0 M NaNO₃) (0.5 g/L dosage, and initial As(V) concentration 20 mg/L).

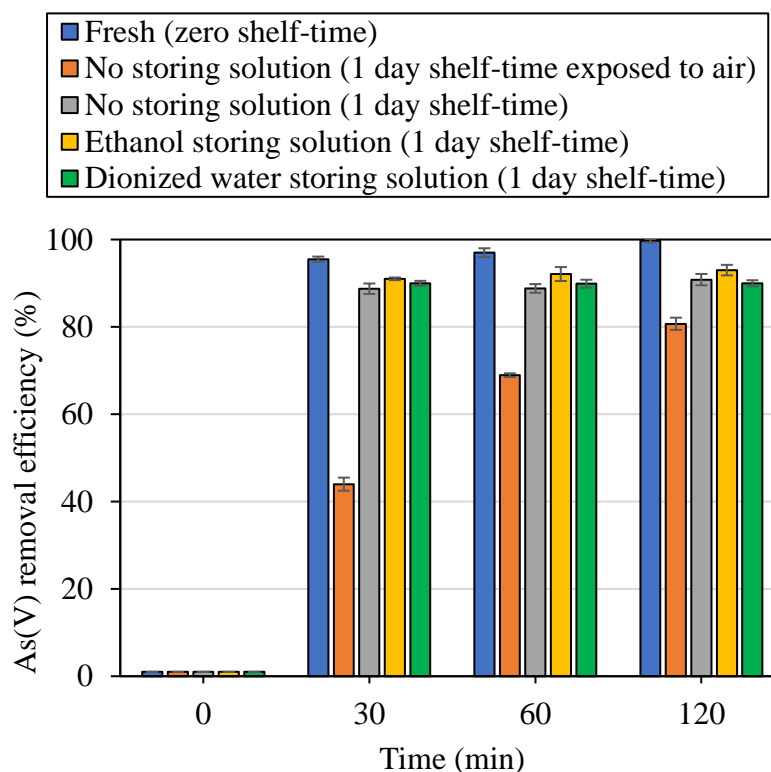


Fig. S6. As(V) removal by $n\text{Fe}^0/\text{Mg}(\text{OH})_2$ -100% considering different storing conditions (0.5 g/L dosage, and initial As(V) concentration 20 mg/L).

REFERENCES

- I. Maamoun, O. Falyouna, R. Eljamal, K. Bensaida, K. Tanaka, T. Tosco, Y. Sugihara, O. Eljamal, Multi-functional magnesium hydroxide coating for iron nanoparticles towards prolonged reactivity in Cr(VI) removal from aqueous solutions, *J. Environ. Chem. Eng.*, **2022**, 10, 107431. doi.org/10.1016/j.jece.2022.107431.
- M.-Z. Ainiwaer XibaiAU - Yin, XianqiangAU - Wen, JiongAU - Su, ShimingAU - Wang, YananAU - Zhang, YangAU - Zhang, TuoAU - Zhang, NanTI - Thermodynamics, Kinetics, and Mechanisms of the Co-Removal of Arsenate and Arsenite by Sepiolite-Supported Nanoscale Zero-Va, Thermodynamics, Kinetics, and Mechanisms of the Co-Removal of Arsenate and Arsenite by Sepiolite-Supported Nanoscale Zero-Valent Iron in Aqueous Solution, *Int. J. Environ. Res. Public Health.*, **2022**, 19. https://doi.org/10.3390/ijerph191811401.
- R. Yadav, A.K. Sharma, J.N. Babu, Sorptive removal of arsenite [As(III)] and arsenate [As(V)] by fuller's earth immobilized nanoscale zero-valent iron nanoparticles (F-nZVI): Effect of Fe0 loading on adsorption activity, *J. Environ. Chem. Eng.*, **2016**, 4, 681–694. https://doi.org/https://doi.org/10.1016/j.jece.2015.12.019.
- Q. Du, S. Zhang, B. Pan, L. Lv, W. Zhang, Q. Zhang, Bifunctional resin-ZVI composites for effective removal of arsenite through simultaneous adsorption and oxidation, *Water Res.*, **2013**, 47, 6064-6074. https://doi.org/https://doi.org/10.1016/j.watres.2013.07.020.
- X. Luo, C. Wang, S. Luo, R. Dong, X. Tu, G. Zeng, Adsorption of As (III) and As (V) from water using magnetite Fe₃O₄-reduced graphite oxide-MnO₂ nanocomposites, *Chem. Eng. J.*, **2012**, 187, 45-52. https://doi.org/https://doi.org/10.1016/j.cej.2012.01.073.
- M.C.S. Faria, R.S. Rosemberg, C.A. Bomfeti, D.S. Monteiro, F. Barbosa, L.C.A. Oliveira, M. Rodriguez, M.C. Pereira, J.L. Rodrigues, Arsenic removal from contaminated water by ultrafine δ -FeOOH adsorbents, *Chem. Eng. J.*, **2014**, 237, 47-54. https://doi.org/https://doi.org/10.1016/j.cej.2013.10.006.
- X. Sun, C. Hu, X. Hu, J. Qu, M. Yang, Characterization and adsorption performance of Zr-doped akaganéite for efficient arsenic removal, *J. Chem. Technol. Biotechnol.*, **2013**, 88, 629-635. https://doi.org/https://doi.org/10.1002/jctb.3878.
- L. Feng, M. Cao, X. Ma, Y. Zhu, C. Hu, Superparamagnetic high-surface-area Fe₃O₄ nanoparticles as adsorbents for arsenic removal, *J. Hazard. Mater.*, **2012**, 217-218, 439-446. https://doi.org/10.1016/j.jhazmat.2012.03.073.
- G. Zhang, Z. Ren, X. Zhang, J. Chen, Nanostructured iron(III)-copper(II) binary oxide: A novel adsorbent for enhanced arsenic removal from aqueous solutions, *Water Res.*, **2013**, 47, 4022-4031. https://doi.org/https://doi.org/10.1016/j.watres.2012.11.059.
- H.-J. Hong, W. Farooq, J.-S. Yang, J.-W. Yang, Preparation and evaluation of Fe-Al binary oxide for arsenic removal: comparative study with single metal oxides, *Sep. Sci. Technol.*, **2010**, 45, 1975-1981.
- L. Hao, T. Zheng, J. Jiang, Q. Hu, X. Li, P. Wang, Removal of As (III) from water using modified jute fibres as a hybrid adsorbent, *RSC Adv.*, **2015**, 5, 10723-10732.
- A.K. Mishra, S. Ramaprabhu, Ultrahigh arsenic sorption using iron oxide-graphene nanocomposite supercapacitor assembly, *J. Appl. Phys.* 112 (2012) 104315.