Supplementary Information

Utilization of Agricultural Waste to Herbicide Removal: Magnetic BEA Zeolite Adsorbents Prepared by Dry-Gel Conversion Using Rice Husk Ash–Derived SiO_2 for Paraquat Removal

Vanpaseuth Phouthavong[a], Takeshi Hagio[a,b,*], Jae-Hyeok Park[b], Supinya Nijpanich[c], Teeranun Srihirunthanon[d,e], Nutchanan Chantanurak[d,e], Kanchanok Duangkhai[d,e], Ratana Rujiravanit[d,e], Vanseng Chounlamany[f], Kesiny Phomkeona[f], Long Kong[g], Liang Li[g], Ryoichi Ichino[a,b]

[a]Department of Chemical Systems Engineering, Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

[b]Institute of Materials Innovation, Institutes of Innovation for Future Society, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8601, Japan

[c]Synchrotron Light Research Institute (Public Organization), Nakhon Ratchasima 30000, Thailand

[d]The Petroleum and Petrochemical College, Chulalongkorn University, Bangkok 10330, Thailand

[e]Center of Excellence on Petrochemical and Materials Technology, Chulalongkorn University, Bangkok 10330, Thailand

[f]Department of Chemistry, Faculty of Natural Sciences, National University of Laos, Vientiane Capital, Laos

[g]School of Environmental Science and Engineering, Shanghai Jiao Tong University, 800 Dongchuan Road Shanghai 200240, China

***Corresponding author:** Takeshi Hagio, email: hagio@mirai.nagoya-u.ac.jp

❖ **Extraction of RHAS from RHA**

As-received RHA was refluxed with 4M HCl and continuously stirred at 110°C for 3 h in order to remove metals and impurities, followed by washing with distilled water until a neutral pH, filtration and drying at 60°C. Then, it was treated with 5M NaOH (110°C, 3 h) to obtain a Na_2SiO_3 solution. RHAS was then precipitated from the solution after cooling down to room temperature by adding 5M H_2SO_4 until pH reached 2. The precipitated RHAS was collected, washed using deionized water, and dried in a hot air oven at 60°C.

❖ **Calculation of BEA zeolite mass percentage in the calcined magnetic RHAS-BEA sample**

The Fe_3O_4 content in the as-synthesized magnetic RHAS-BEA sample (before calcination, BC) was calculated to be 20.9 wt% using weight of the obtained precursor gel, the amount of gel used for DGC and weight of the as-synthesized product totally obtained. In this case, we neglect the phase change of Fe_3O_4 to α-Fe_2O_3 by calcination, therefore the Fe_3O_4 amount in the sample before and after calcination did not significantly change. Amount of Fe_3O_4 in the sample obtained after calcination (AC) can be calculated using **Eq. S1**.

$$(W)_{Fe3O4\_AC} \approx (W)_{Fe3O4\_BC} = 0.209 \times (W)_{BEA/Fe3O4\_BC} \qquad \textbf{(Eq. S1)}$$

Weight of RHAS-BEA after calcination at 450°C for 12 h was 71.8 wt% of the as-synthesized RHAS-BEA (before calcination). The loss of weight represents the burned-off SDA, while the remaining part was considered to be the BEA zeolite phase. The amount of BEA in calcined samples can be calculated by using **Eq. S2**.

$$(W)_{BEA\_AC} = 0.718 \times (W)_{BEA\_BC} = 0.718 \times ((W)_{BEA/Fe3O4\_BC} - (W)_{Fe3O4\_BC}) \qquad \textbf{(Eq. S2)}$$

Substitute W_{Fe3O4_BC} in **Eq. S2** with **Eq. S1**,

$$(W)_{BEA\_AC} = 0.718 \times ((W)_{BEA/Fe3O4\_BC} - 0.209 \times (W)_{BEA/Fe3O4\_BC}) \qquad \textbf{(Eq. S3)}$$

For example, when 0.2008 g of as-synthesized magnetic RHAS-BEA was calcined, the amount of BEA in calcined sample was calculated using **Eq. S3**

$$(W)_{BEA\_AC} = 0.718 \times (0.2008\ g - 0.209 \times 0.2008\ g) = 0.1140\ g$$

The amount of the sample remained after calcination was found to be 0.1570 g, therefore weight percentage of BEA in the calcined magnetic RHAS-BEA is (0.1140 g/0.1570 g)×100 = **72.6 wt%**.

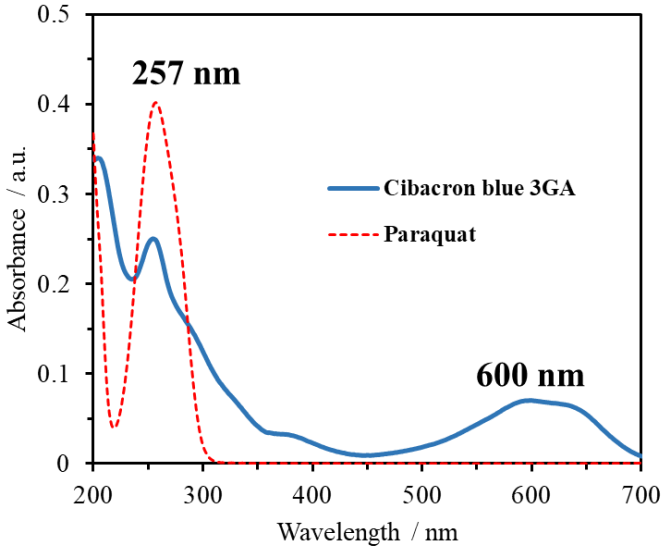

Fig. S1. UV-Vis absorption spectra of paraquat and Cibacron blue 3GA.

Table S1 Intraparticle diffusion model parameters of paraquat adsorption on RHAS-BEA and magnetic RHAS-BEA.

	Intraparticle diffusion model parameter values					
	RHAS-BEA			Magnetic RHAS-BEA		
	1st Step	2nd Step	3rd Step	1st Step	2nd Step	3rd Step
k_{id} (mg/(g·min$^{1/2}$))	9.2327	5.534	0.6142	13.198	3.7999	0.4327
C	23.471	32.114	54.787	10.977	31.018	46.566
R^2	0.8690	0.9725	0.9328	0.9987	0.9246	0.9999