Supplementary Information

Aminoclay-templated nanoscale zero-valent iron (nZVI) synthesis for efficient harvesting of oleaginous microalga, *Chlorella* sp. KR-1†

Young-Chul Lee,†[a,e] Kyubock Lee,†[b] Yuhoon Hwang,[c] Henrik Rasmus Andersen,[c] Bohwa Kim,[b] So Yeun Lee,[b] Moon-Hee Choi,[d] Ji-Yeon Park,[b] You-Kwan Oh*[b] and Yun Suk Huh*[a]

[a]Department of Biological Engineering, College of Engineering, Inha University, Incheon 402-751, Republic of Korea. Fax: +82-42-865-3613; Tel: +82-42-865-3610; E-mail:yunsuk.huh@inha.ac.kr

[b]Clean Fuel Department, Korea Institute of Energy Research, 152 Gajeong-ro, Yuseong-gu, Daejeon 305-343, Republic of Korea. Fax: +82-42-860-3495; Tel: +82-42-860-3697; E-mail: ykoh@kier.re.kr

[c]Department of Environmental Engineering, Technical University of Denmark, Bygningstorvet, Building 115, 2800 Kgs. Lyngby, Denmark

[d]Department of Beauty and Cosmetology, Graduate School of Industry, Chosun University, Gwangju 501-759, Republic of Korea

[e]Department of Civil and Environmental Engineering (BK21 program), KAIST, 291 Daehakno, Yuseong-gu, Daejeon 305-701, Republic of Korea

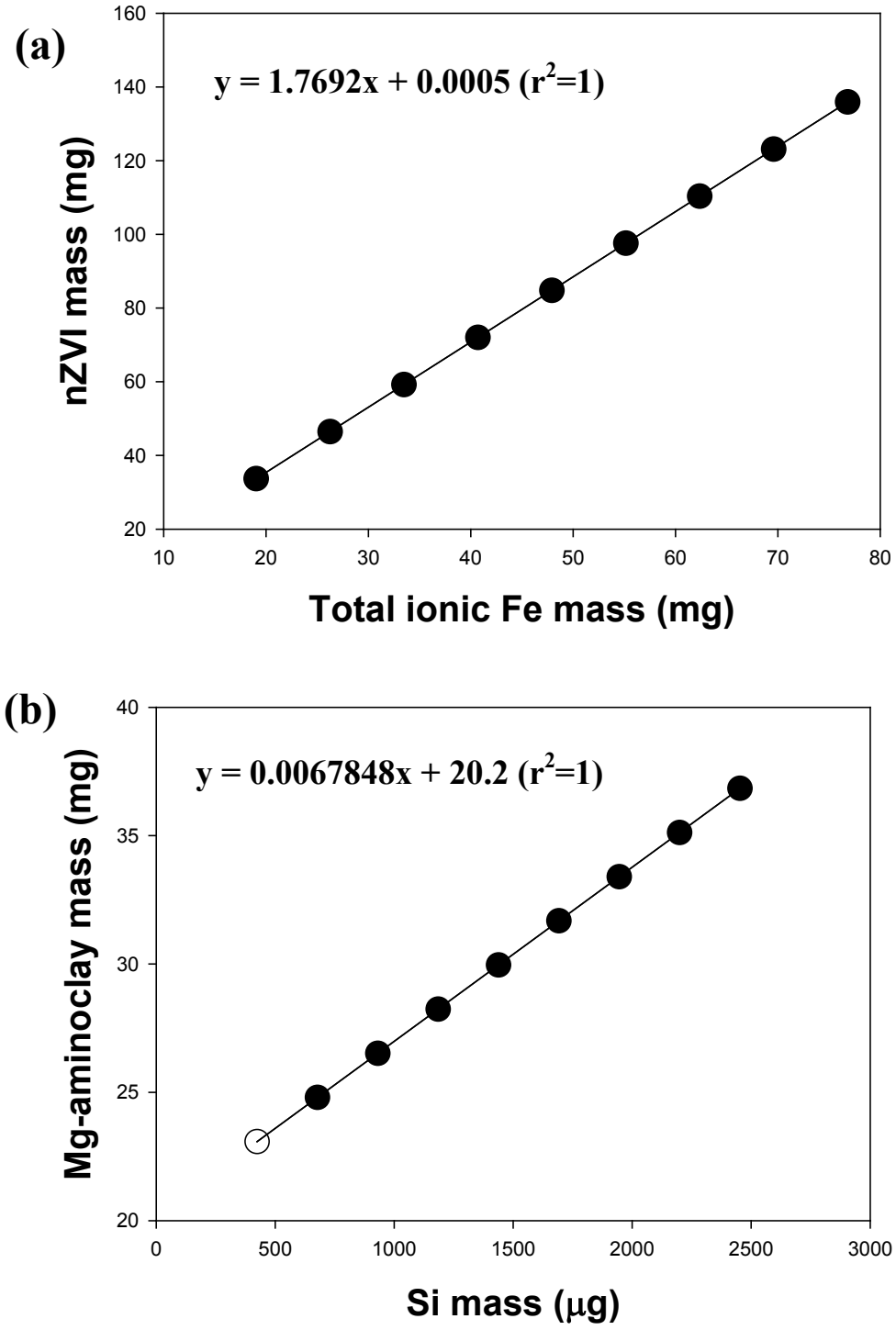

Figure S1. The calibration curves of total ionic Fe (mg) after acid digestion versus nZVI (mg) (a) and Si concentration (μg) versus aminoclay (mg) (b) by measurement of inductively

coupled plasma atomic emission spectrometer (ICP-AES). Note that in (a), nZVI recovery (%) after acid digestion indicates approximately 56.5%.

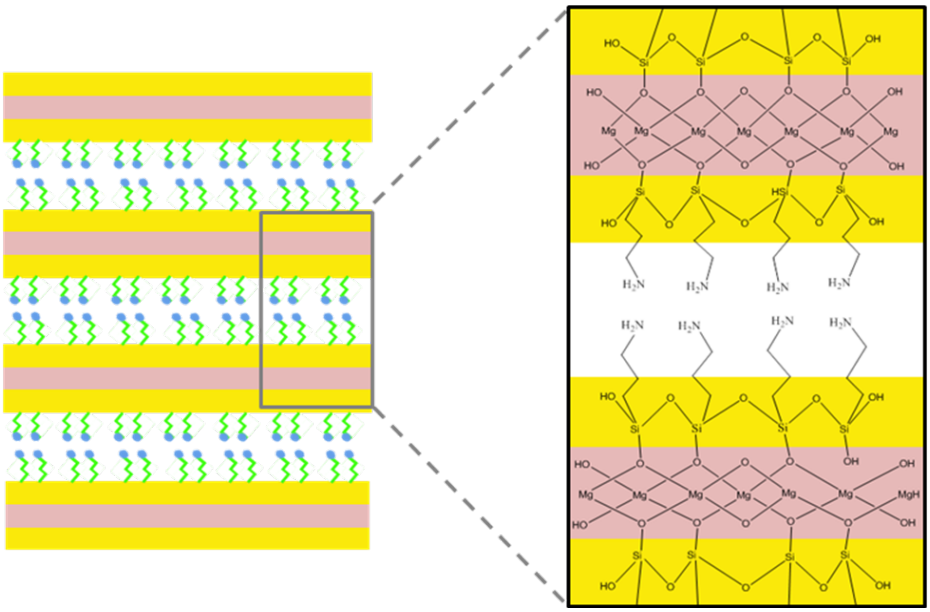

Figure S2. The suggested approximate unit structure of aminoclay, reproduced with permission of reference 10.

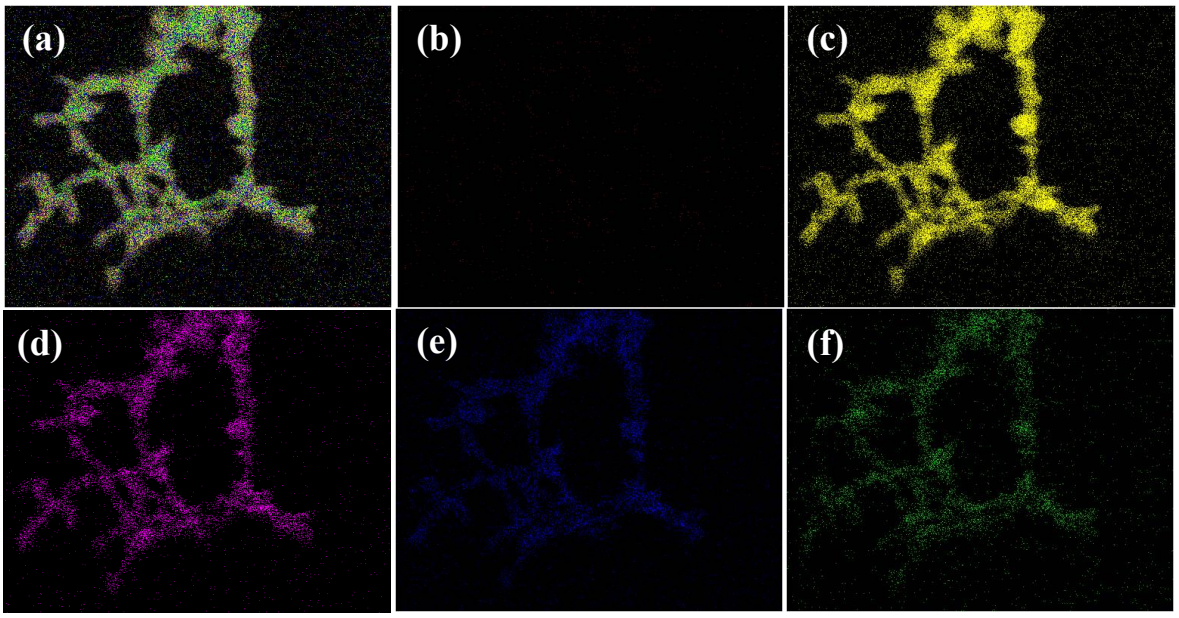

Figure S3. TEM images of elemental mapping in aminoclay-nZVI nanocomposite (ratio1.0): (a) overlay image from (b) to (f), (b) Fe, (c) O, (d) Mg, (e) Si, and (f) Cl.

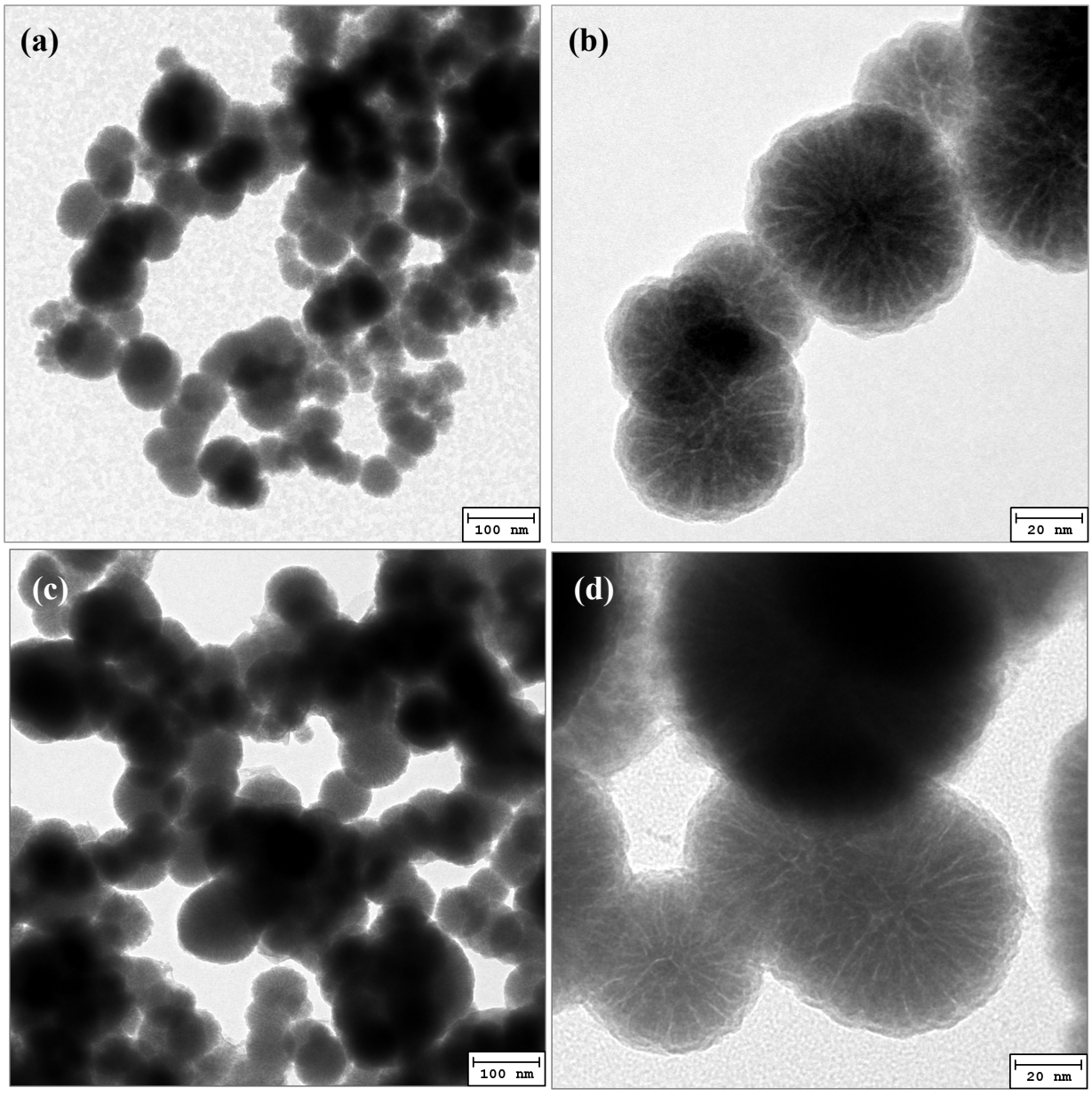

Figure S4. Transmission electron microscopic (TEM) images of aminoclay-nZVI composites (ratio 5.0) (a,b), and (ratio 7.5) (c,d).

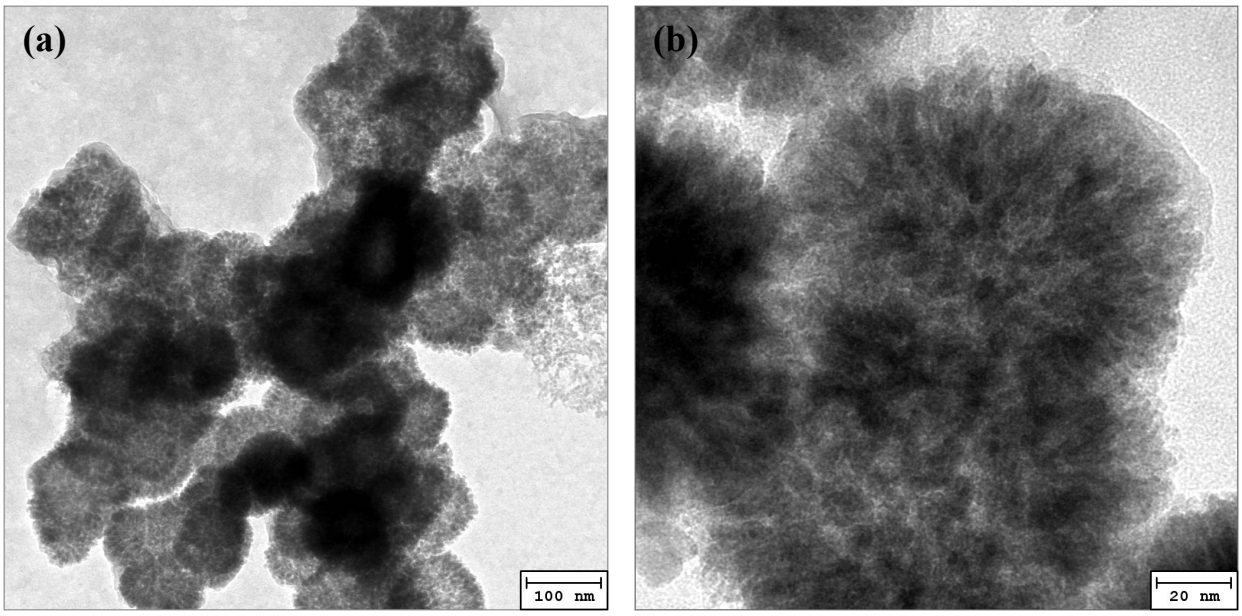

Figure S5. Transmission electron microscopic (TEM) images of aminoclay-nZVI composite (ratio 1.0) after 1 week aging.

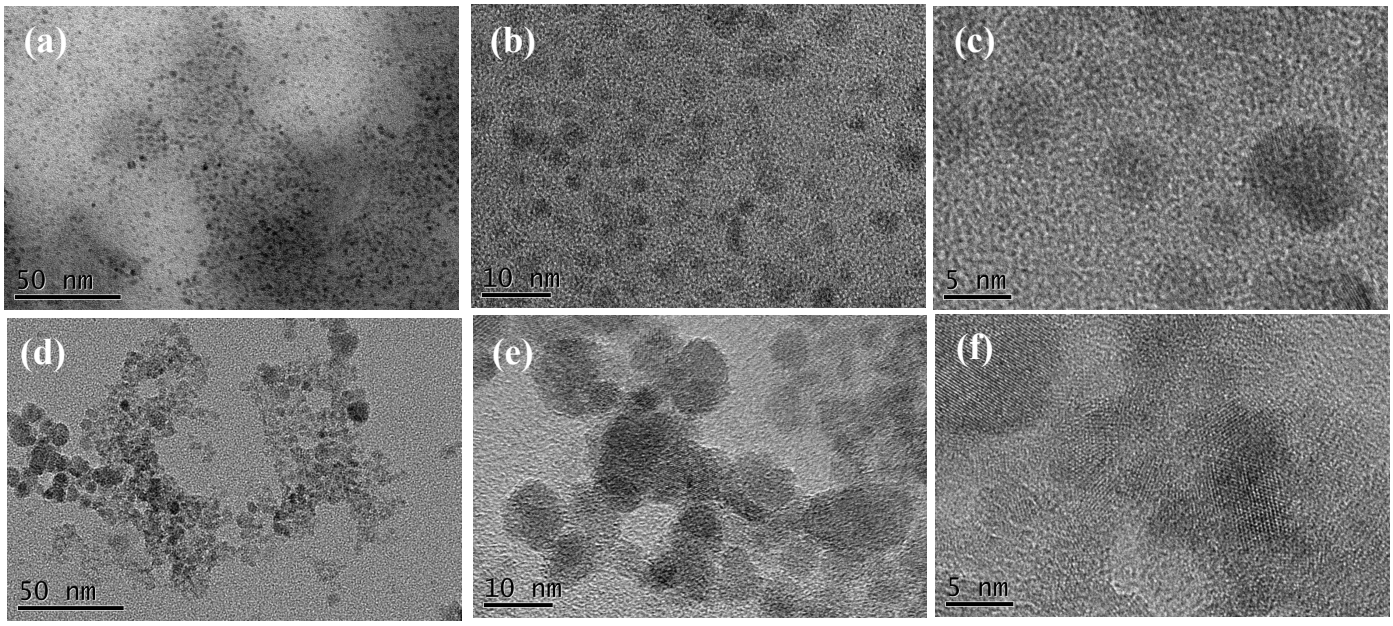

Figure S6. Transmission electron microscopic (TEM) images of Fe_3O_4 synthesis without aminoclay (a-c) and in the presence of aminoclay (0.5 mg/mL) (d-f) by co-precipitation method of $FeCl_2$ and $FeCl_3$.

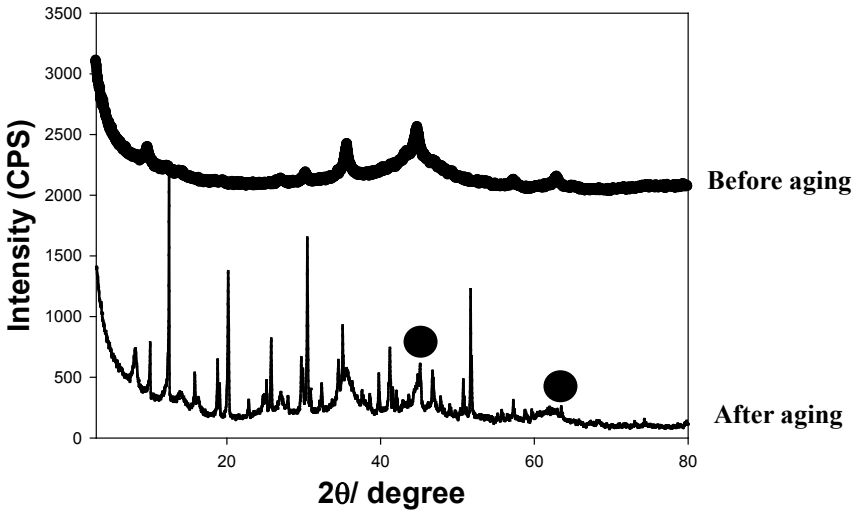

Figure S7. Powder X-ray diffraction (XRD) patterns of aminoclay-nZVI composite (ratio 1.0) before/after 1 week's aging in aqueous solution. Note that the symbol of black filled circles marked standard cubic Fe(0) phase.

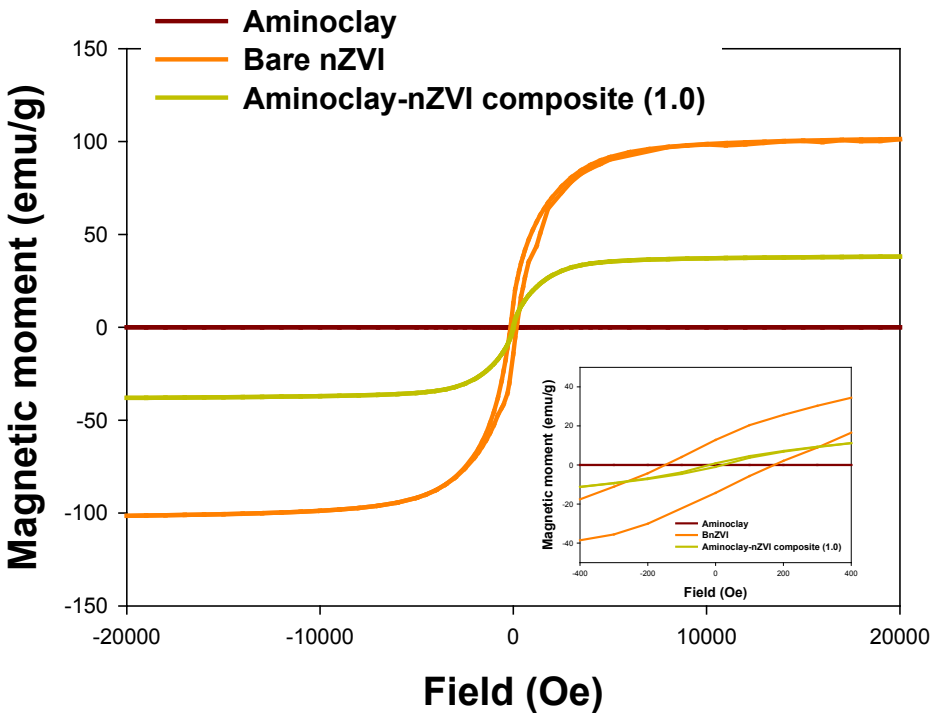

Figure S8. Room temperature hysteresis loops of aminoclay, bare nZVI, and aminoclay-nZVI composite (ratio 1:0). Inset displays the hysteresis of zoom region between -400 and 400 Oe.

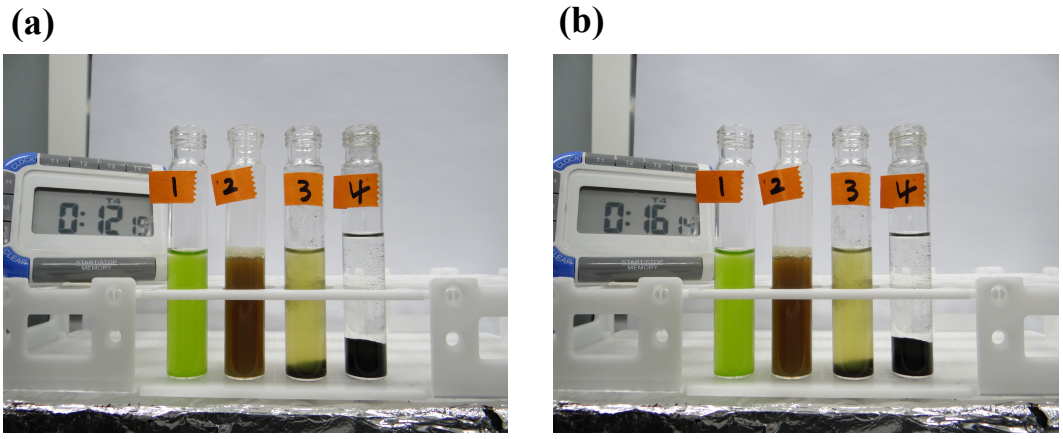

Figure S9. Microalgae harvesting for 1-4 after 5 min (a) and 10 min (b) under magnetic field on bottom. 1: fresh *Chlorella* sp. KR-1, 2: Fe-based aminoclay as a control which has no magnetic property, 3: bare nZVI, and 4: aminoclay-nZVI composite (ratio 1.0).

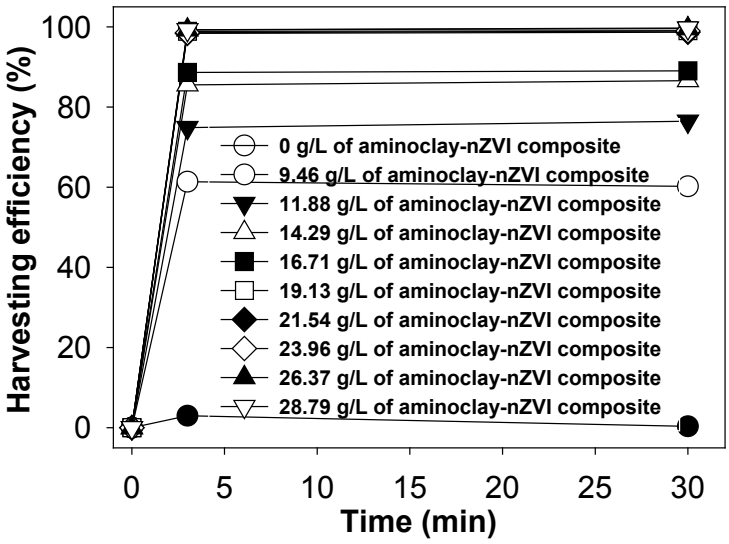

Figure S10. Harvesting kinetics according to aminoclay-nZVI loading (ratio 1.0).

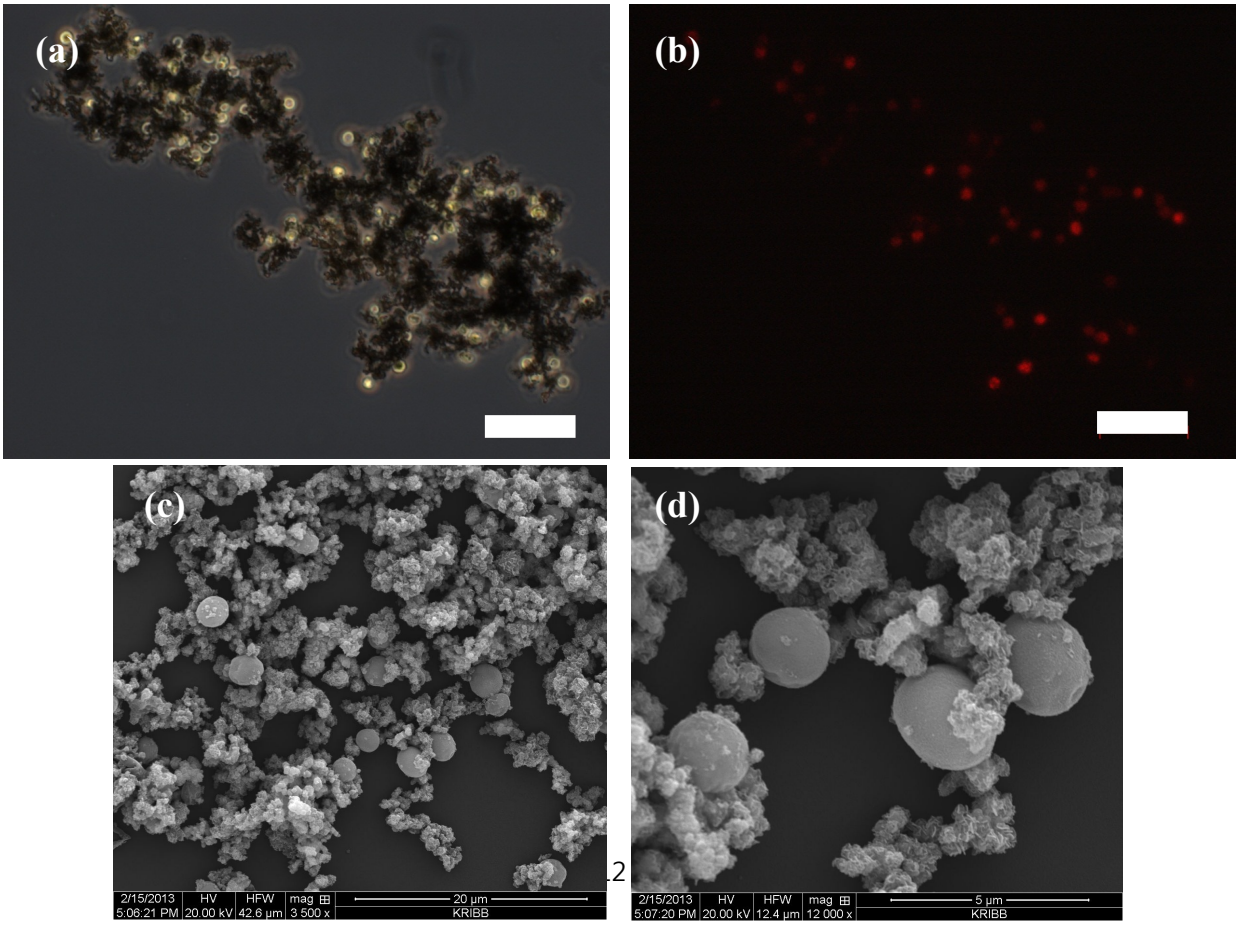

Figure S11. Optical microscopic (a) and its fluorescence (b) images of flocculated *Chlorella* sp. biomass, scanning electron microscopic (SEM) images (c,d) by aminoclay-nZVI composite (ratio 1.0). Scalar bars in (a) and (b) = 20 μm.

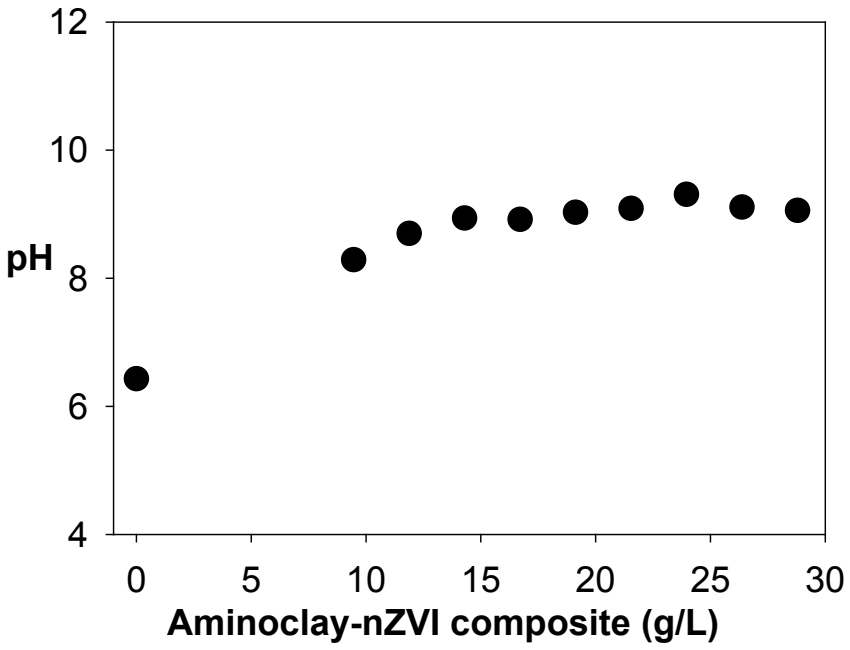

Figure S12. The pH variation in media according to loading of aminoclay-nZVI composite (ratio 1:0).

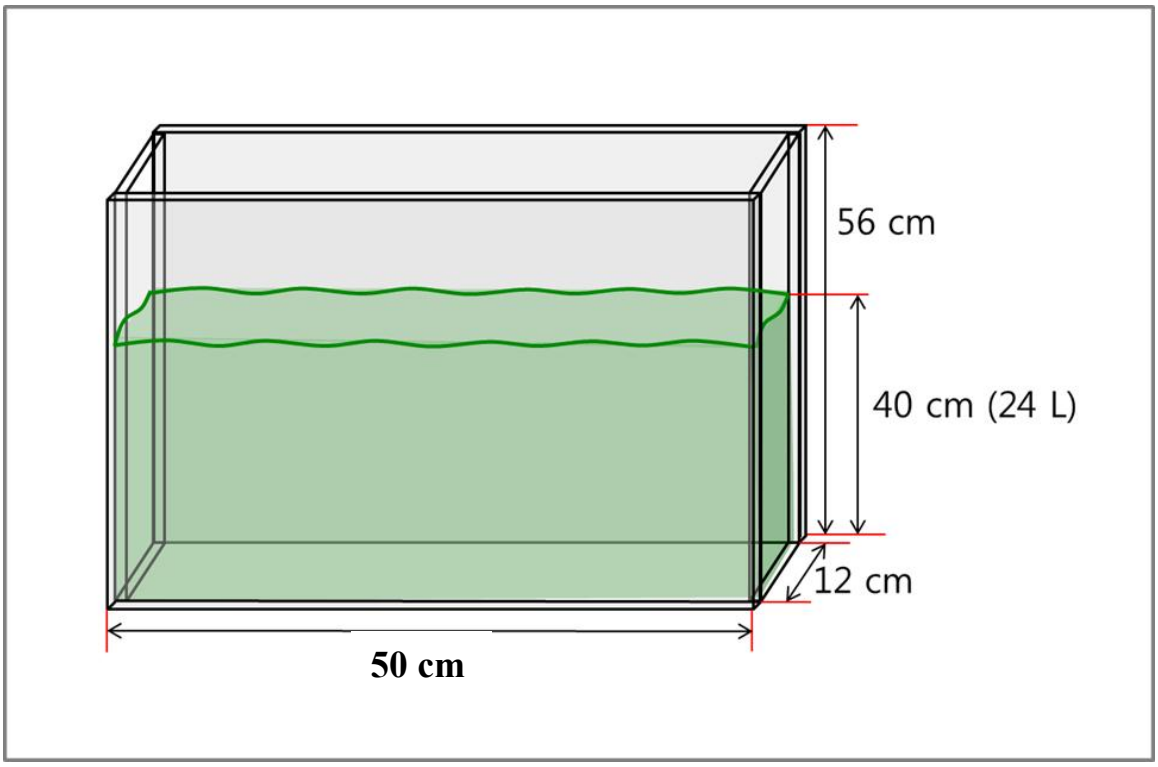

Figure S13. Schematic reactor for 24L microalgae harvesting.

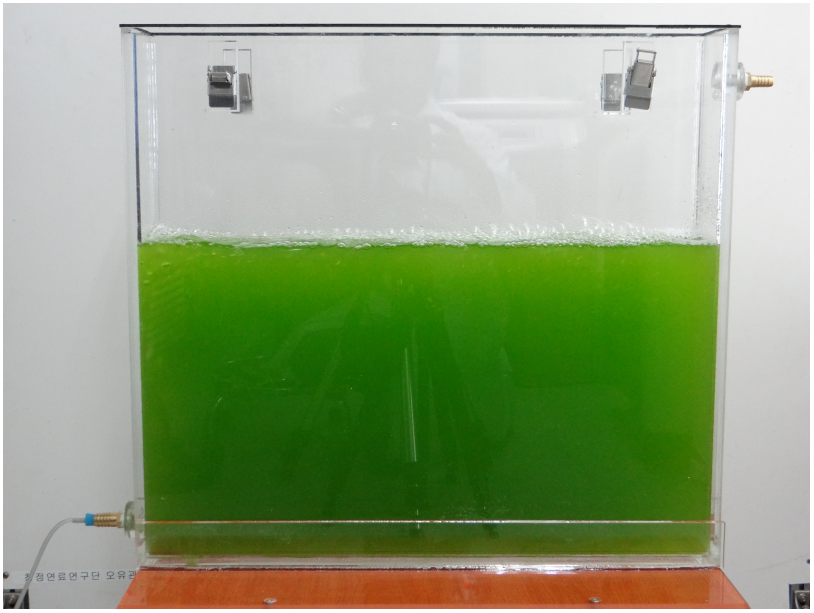

Figure S14. Dica photograph of air bubbled 24L microalgal solution in the plastic container

before injection of aminoclay-nZVI composite.