1 Repurposed marble dust as a promising adsorbent for modelling removal of

- 2 methylene blue from aqueous solutions
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14 S1.1 Experimentation

15 Marble slurry was collected from RIICO Industrial area Chittorgarh, Rajasthan (India) and dried

16 in open environment. The dried marble powder was cleaned repeatedly with DI water in order to

17 eliminate unwanted dirt and water-soluble impurities. The slurry was parched in a hot air oven at

18 353 K for 12 hours to remove excess water. In order to reduce the particle size, the marble dust

19 was mechanically pulverized in planetary ball mill followed by activation through annealing at

20 1123 K for 4 hours in a muffle furnace. The calcination releases Carbon Dioxide and form Calcium

21 Oxide from Calcium Carbonate (Eq.1).

22
$$\operatorname{CaCO}_3 \xrightarrow{\Delta} \operatorname{CaO} + \operatorname{CO}_2$$
 (1)

The batch adsorption experiments were executed by following the conventional batch bottle technique. The stock solution of MB was diluted in requisite amount to prepared the working solution. 0.1M HCl and 0.1M NaOH were added to maintain the requisite pH of initial reaction solution. The reagent bottles were filled with 20 mL of dye solution with required amount of AMD. The reaction system was kept in water bath shaker at 200 rpm at a particular temperature. After keeping it for the requisite amount of time, the AMD was extracted out from the system by centrifugal separation at 10000 rpm for 10 min and the overlying layer was subsequently used for estimation of final concentration with UV spectroscopy.

31 Fig.S1 Plot of predicted versus actual values of removal percentage of MB by AMD.



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33 Fig.S2 Thermodynamic studies of MB adsorption on AMD at different temperatures 303, 313,

34 323K (a) plot of lnK_C versus l/T for calculation of ΔG° , ΔH° and ΔS° ($R^2=0.9472$) (b) plot of ln(l-1)

35 θ and 1/T for calculation of S^* and E_a ($R^2=0.9874$)



Fig.S3 (a) Henry's Isotherm plot at 303K (R^2 =0.9880), 313K (R^2 =0.9931) and 323K (R^2 =0.9946) temperatures for different concentrations from 10 to 70 mg L⁻¹ at 0.55g AMD dose for 135 min at pH=14 (b) plot of lnK_H versus 1/T for elucidation of Δ H° value (R^2 =0.9323)



Fig.S4 XPS Survey plot of AMD before and after adsorption [10 mg L⁻¹ initial MB concentration
with 1 g AMD dose for 135 min time at 313 K temperature at pH 6].



44 Fig.S5 Regeneration studies conducted for MB removal by AMD [30 mg L⁻¹ initial MB

45 concentrations at pH 6 for 1g AMD dose for 135 min at 313K]



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