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## **Supporting information**

## Shape and size controlled synthesis of Cu nanoparticles-wrapped on RGO nanosheet catalyst and their outstanding stability and catalytic performance in the hydrogenation reaction of dimethyl oxalate

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## Synthesis of GO NSs using modified Hummer method

Modified Hummers method was utilized to synthesize graphene oxide from the oxidation of high purity graphite powder. 96.0 ml Conc H<sub>2</sub>SO<sub>4</sub> was added gradually to a mixture of 2 gm purified graphite flakes and 1 gm of sodium nitrate with keeping in ice bath. After around 30 min of vigorous magnetic stirring, 6 gm of KMnO<sub>4</sub> was added gradually and very carefully and stirred for 2 h under 20 °C as maintained by using ice bath. The mixture was placed in an oil bath whose temperature was maintained around 35 °C by a hot plate and magnetically stirred for 20 h after which the mixture turned into heavy brownish pasty color. Then, 150 ml di- water was added gradually within vigorous stirring. When the temperature increased with huge toxic gas bubbling, the more di-water was added until the solution diluted with 240 ml. When the diluted suspension color changed into brilliant yellow, 5 ml of  $H_2O_2$  as strong oxidizing agent was added to it. After 2 h of continuous stirring, the mixture was filtered and washed by rinsing and centrifugation with mixture of 10% HCl. Then ultra-pure ethanol and di-water was added for several times to increase the pH value towards neutral. The resulting filtered mixturewas dried in oven for 24 h approximately at temperature 70 °C, and finally, solid GO powder was obtained.

| Type of Catalyst               | S <sub>BET</sub> (m2/g) | Pore    | Cu+/(Cu++Cu <sup>0</sup> ) % | <b>TOF</b> value           |
|--------------------------------|-------------------------|---------|------------------------------|----------------------------|
|                                |                         | diamete |                              | ( <b>h</b> <sup>-1</sup> ) |
|                                |                         | r (nm)  |                              |                            |
| Cu/RGO (US)                    | 43.8                    | 8.1     | 27                           | 4.8                        |
| Cu/RGO (NH <sub>4</sub> OH)    | 21.9                    | 7.3     | 28.3                         | 15.3                       |
| Cu/RGO (US/NH <sub>4</sub> OH) | 37.4                    | 18.4    | 34                           | 17.1                       |
| Cu/RGO (10 wt%)                | 49.6                    | 8.2     | 26.3                         | 12.6                       |
| Cu/RGO (25 wt%)                | 37.4                    | 18.4    | 34                           | 17.1                       |
| Cu/RGO (45 wt%)                | 37                      | 19.1    | 26                           | 14                         |

## Table S1 Physico-chemical properties of the prepared catalysts

| Ref       | <b>Reaction temperature (°C)</b> | H2/DMO Ratio mol mol <sup>-1</sup> |
|-----------|----------------------------------|------------------------------------|
| 1         | 200                              | 80                                 |
| 4         | 210                              | 300                                |
| 12        | 80                               | 17.5                               |
| 13        | 220                              | 110                                |
| 32        | 220                              | 80                                 |
| 38        | 180                              | 150                                |
| 39        | 210                              | 200                                |
| This work | 210                              | 200                                |

**Table S2** Reaction condition for the catalysts used in the comparison of the catalytic activity (Related to Table. 1)



Fig. S1 TEM images for the Cu/RGO catalyst (US)



Fig. S2 XPS survey and analogues high resolution images of Cu 2p for Cu/RGO catalysts (a,b) synthesized in different reaction condition and (b) synthesized in presence of ultrasound and ammonia within different Cu wt% loading.



Fig. S3 XPS and HR-XPS to as-prepared, reduced and spent Cu/RGO catalyst to show the shift in the bending Energy values after reducing and used catalyst.



Fig. S4 H2-TPR for Cu/RGO catalysts synthesized (a) in different reaction condition and (b) synthesized in presence of ultrasound and ammonia within different Cu wt% loading.



Fig. S5 MG and DMO conversion ratio for the spent Cu/RGO catalyst after 300 h of reaction time.



Fig. S6 TEM and HRTEM image for the spent Cu/RGO catalyst