# **Supporting Information**

# Heterocyclic aminopyrazine-reduced graphene oxide coated carbon cloth

## electrode as an active bio-electrocatalyst for extracellular electron transfer in

# microbial fuel cell

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### Synthesis of graphene oxide (GO)

GO was prepared through oxidation of graphite powder (Sigma - Aldrich, India) by Hummers Method.<sup>51,52</sup> Graphite weighing 500 mg was mixed with 23 mL of  $H_2SO_4$  and 10 mL of  $HNO_3$  in a measuring flask at 5°C temperature. KMNO<sub>4</sub> (3 g) was added slowly to the mixture and stirring was allowed for 30 minutes at a controlled temperature (< 15°C). The mixture was then heated to 35°C for 3 h resulting in pasty brownish color, and was then slowly diluted with 40 mL of deionized water (DI). The solution was further diluted with 200 mL of DI after 12 h followed by a final addition of 3 mL  $H_2O_2$  (30% wt). The GO mixture was purified by centrifugation at 1500 rpm for 30 min, followed by rinsing with DI water several times. The mixture was vigorously sonicated for 2 to 3 h to exfoliate the stacked GO, followed by filtering and drying to obtain GO powder.

### Synthesis of reduced graphene oxide (r-GO)

The r-GO was prepared according to the procedure as described in the literature. <sup>52, 53</sup> An amount of 100 mg GO was completely dispersed in DI water for 2 to 3 h using an ultrasonic bath. The pH of the solution was adjusted to 10 by 1 M KOH solution, and 1 mL of hydrazine hydrate was added subsequently. The mixture was stirred using a magnetic stirrer at 95°C for 24 h. The resulting black colored solution was washed thoroughly with DI water to remove excess amount of hydrazine, and the final r-GO product was filtered and oven dried at 80°C to obtain a powder form.

### Synthesis of aminopyrazine-reduced graphene oxide (r-GO-Apy) hybrid

An amount of 50 mg of GO and 25 mg of Apy (Sigma-Aldrich, India) was dispersed in 75 mL of DI water by using an ultrasonic bath. The solution was acidified with (pH-2) HCl and stirred for 12 h. The product mixture was thoroughly washed with DI water until the filtrate was colorless, and the pH was adjusted to 10 by ammonium hydroxide solution (5%). Afterwards, 0.2 mL of hydrazine hydrate was added to reduce GO into reduced r-GO. The subsequent solution was kept for 1 h at 95°C to form r-GO-Apy hybrid. <sup>S2, S3</sup>

### Preparation of r-GO-Apy on carbon cloth electrode

The r-GO-Apy hybrid (4 mg mL<sup>-1</sup>) was dispersed in DI water using the ultrasonic bath for 30 min followed by the addition of nafion solution (5% by wt.). The resulting stable solution was painted over on plain carbon cloth (Synergic India Pvt. Ltd.) by means of the brush coating technique. The wet carbon cloth (CC) was allowed for drying in an oven at 150°C for 10 min. During the drying process, the water gets removed from the wet CC, and the r-GO-Apy hybrid forms an intertwined random complex on the cloth surface. The simple "brush coating and drying process" was repeated several times, and the required area of r-GO-Apy coated on CC (r-GO-Apy –CC) was fabricated. Subsequently, the r-GO-Apy-CC hybrid was annealed at 300°C for 2 h.<sup>54</sup> For comparison, PCC with the same geometric area was used as a control electrode.

#### References

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