## **Supplementary Information**

# Water-dispersible graphene for selective and rapid ion mercury (Hg<sup>2+</sup>)-rejecting membranes

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### Discussion of the origin of the Raman modes in graphene

The Raman spectrum of graphene produced through liquid phase exfoliation (LPE) shows, as fingerprints, G (~1585 cm<sup>-1</sup>) <sup>1,2</sup>, D (~1380 cm<sup>-1</sup>) <sup>1,2</sup>, D' (~1620 cm<sup>-1</sup>) <sup>3</sup> and 2D (~2700 cm<sup>-1</sup>)<sup>2</sup> peaks.<sup>2,4,5,6,7,8</sup> The G peak, positioned at ~1585 cm<sup>-1</sup>, corresponds to the  $E_{2g}$  phonon at the Brillouin zone centre.<sup>2,4</sup> The D peak is due to the breathing modes of sp<sup>2</sup> rings and requires a defect for its activation by double resonance.<sup>1,9,10</sup> Double resonance happens as an intra-valley process, *i.e.*, connecting two points belonging to the same cone around K or K',<sup>2,4,5</sup> giving origin to the D' peak.<sup>2,4,5</sup> The 2D peak is the second order of the D peak, <sup>2,4,5</sup> and it appears also in the absence of D peak, since no defects are required for the activation of two phonons with the same momentum, one backscattered from the other.<sup>11,12</sup> Moreover, the 2D peak is a excitation wavelength-dependent single peak (centred at ~2680 cm<sup>-1</sup> at excitation wavelength of 514 nm) for single-layer graphene (SLG),<sup>4,2,5</sup> whereas is a superposition of multiple components, the main being the 2D<sub>1</sub> and 2D<sub>2</sub> components, for few-layers graphene (FLG).<sup>2,4,5</sup> In graphite, the intensity of the 2D<sub>2</sub> band is twice the 2D<sub>1</sub> band,<sup>2,4,13</sup> while for graphene the 2D band is a single and sharp Lorentzian band,<sup>4</sup> which is roughly four times more intense than the G peak.<sup>4</sup> Taking into account the intensity ratio between 2D<sub>1</sub> and 2D<sub>2</sub> –I(2D<sub>1</sub>)/(I(D<sub>2</sub>)–, it is possible to estimate the flake thickness.<sup>14</sup>

### Raman spectroscopy analysis of graphene produced through liquid phase exfoliation in Methyl-2-pyrrolidone

**Fig. S1** shows a representative Raman spectrum of a sample consisting of graphene flakes produced by conventional LPE in Methyl-2-pyrrolidone (NMP).<sup>15,16,175,18</sup> As discussed in the main text, the Raman peaks attributed to G, D, D' and 2D modes of graphene are identified. These Raman signatures are used to uniquely identified the peak ascribed to graphene in the as-produced graphene functionalized with cationic rhodamine 6G – R6G– (R6G-FG).



Fig. S1. Raman spectrum of graphene flakes produced by conventional LPE in NMP

### Energy dispersive X-ray spectroscopy analysis of membrane based on graphene flakes produced LPE in NMP

**Fig. S2** shows energy dispersive X-ray spectroscopy combined with scanning electron microscopy (SEM-EDS) analysis for a membrane based on graphene produced by conventional LPE in NMP (graphene mass loading of 35.8 g m<sup>-2</sup>) after the passage of 263.2 L m<sup>-2</sup> of the Hg(ClO<sub>4</sub>)<sub>2</sub> solution ([Hg<sup>2+</sup>] = 10 ppm). In contrast to the membranes based on R6G-FG, the elemental analysis does not reveal any deposit of Hg, in agreement with its poor Hg<sup>2+</sup> rejection capability (~10%, Fig. 5b of the main text).



**Fig. S2.** a) Top-view SEM images a membrane based on graphene flakes produced by conventional LPE in NMP (graphene flakes mass loading of 35.8 g m<sup>-2</sup>) after the passage of 263.2 L m<sup>-2</sup> of the  $Hg(ClO_4)_2$  solution ( $[Hg^{2+}] = 10$  ppm). b,c) Elemental mapping of C and Hg for the SEM image and d) the corresponding EDX spectrum of the R6G-FG membrane imaged in a).

#### Photoluminescence analysis of R6G-FG dispersions

**Fig. S3** shows the PL spectra of the R6G-FG dispersions in water before and after the addition of  $Hg(CIO_4)_2$ . The addition of small amounts of a  $Hg(CIO_4)_2$  aqueous solution with 10 ppm of  $Hg^{2+}$  promotes the quenching of the PL emission of the R6G molecules in R6G-FG. However, a further increase of the  $Hg^{2+}$  concentration leads to a partial recovery of the PL emission, indicating that R6G molecules are released from the graphene, which also effectively quenches the PL of the R6G molecules through physisorption interactions (see **Fig. 2h**). Notably, the non-monotonic PL behavior with increasing the amount of  $Hg^{2+}$  is not displayed by R6G solution, in agreement with previous literature.<sup>19</sup> Overall, these additional PL data supports the competitive adsorption working mechanism detailed in the manuscript.



**Fig. S3.** PL spectra of R6G-FG in water before (red) and after the addition of  $Hg^{2+}$ : 0.2 ppm (green) and 0.6 ppm (light green). All the PL spectra were normalized to the absorbance of the samples at the excitation wavelength (400 nm).

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