# Supporting Information

## Satellite Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-Au SERS probe for trace Hg<sup>2+</sup> detection

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Figure S1. HR-TEM image of Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub> microsphere.



Figure S2. Zeta-potentials during FA functionalization.



**Figure S3.** (A) UV-vis spectra of CR. (B) The calibration curve of CR from 0 to 100 μM. The inset red point was CR concentration after FA functionalization.

#### Determination of maximum CR molecule density on the FA surface

#### (a) CR molecule on each FA microsphere

According to UV-vis experiment (Figure S5), the amount of CR molecular functionalized on 1 mL FA substrate was

$$N_{CR1} = C_{CR} \times V_{CR}$$

 $\approx (100 - 63.3) \times 1 \times 10^{-6} \times 6.02 \times 10^{23} \times 10^{-3}$ 

 $\approx 2.21 \times 10^{16}$ 

There are 1mg Fe<sub>3</sub>O<sub>4</sub> in 1 mL FA substrate, the amount of Fe<sub>3</sub>O<sub>4</sub> was

$$N_{Fe_{3}0_{4}} \approx \frac{m_{Fe_{3}0_{4}1}}{m_{Fe_{3}0_{4}2}}$$

$$= \frac{1}{\frac{4}{3}\pi \times 155 \times R_{Fe_{3}0_{4}}^{3} \times \rho_{Fe_{3}0_{4}}}}$$
$$= \frac{1}{\frac{4}{3}\pi \times 155^{3} \times 5.14 \times 10^{-18}}}$$
$$\approx 1.25 \times 10^{10}$$

The amount of CR molecule functionalized on each FA sphere was

$$N_{CR2} = \frac{N_{CR1}}{N_{Fe_3}o_4}$$
$$\approx 1.77 \times 10^6$$

## (b) Surface area of each FA microsphere

$$N_{Au} \approx \frac{4\pi}{\Phi}$$
$$\approx \frac{4 \times \pi \left(R + r + \frac{gap}{2}\right)^2}{\pi (r + \frac{gap}{2})^2}$$
$$= \frac{4 \times (115 + 10 + 3.5)^2}{(10 + 3.5)^2}$$
$$\approx 362$$

$$S_{FA} = S_{Fe_3O_4} + N_{Au}S_{Au}$$

$$=4\pi R^2 + 362 \times 4\pi r^2$$

Where  $\Phi$  represents the deflection angle, Gap is distance between Au NPs, R is the radii of the Fe<sub>3</sub>O<sub>4</sub> NP, r is the radii of the Au NP (Figure S6). Here, Gap, R and r are experimentally determined as Gap=7 nm, R=115 nm and r =10 nm. Based on these parameters, we obtained that the amount of Au NPs bound to an Fe3O4 sphere is about 362. Then surface area of each FA microsphere is about 756939.

### (c) CR density on FA surface

$$D_{CR} = \frac{N_{CR2}}{S_{FA}}$$
$$\approx \frac{1.77 \times 10^6}{7.57 \times 10^5}$$

≈ 2.34

The CR molecule density functionalized on each FA sphere can be estimated to be about 2.34 per nm<sup>2</sup>.



Figure S4. Estimation of the CR density on FA surface.

Elt.	Line	Intensity (c/s)	Atomic %	Conc	Units	MDL3-sig	
Si	Ka	76.56	14.381	4.75	wt.%	0.237	
Fe	La	77.84	62.134	40.82	wt.%	1.268	
Au	La	153.97	23.231	53.83	wt.%	.995	
Hg	La	1.54	0.255	0.60	wt.%	1.111	
			100.000	100.00	wt.%		Total

 Table S2 EDS for sample FA-CR after Hg<sup>2+</sup> adsorption

Elt.	Line	Intensity (c/s)	Atomic %	Conc	Units	MDL 3-sig	
Si	Ka	88.62	13.887	4.45	wt.%	0.222	
Fe	La	89.49	60.890	38.80	wt.%	1.471	
Au	La	187.18	23.653	53.16	wt.%	0.806	
Hg	La	11.33	1.571	3.59	wt.%	0.877	
			100.000	100.00	wt.%		Total



Figure S5. TEM and SEM images of FA-CR before (A, C) and after (B, D) Hg<sup>2+</sup> adsorption.



Figure S6. SERS spectra (A) and peak intensity at 1159 cm<sup>-1</sup> (B) of FA-CR with  $1x10^{-5}$  M Hg<sup>2+</sup>

mixing time from 1 min to 60 min. Signal collection time was 1 s.



**Figure S7**. Peak intensity at 1159 cm<sup>-1</sup> of FA-CR BK and FA-CR with  $1x10^{-5}$  M Hg<sup>2+</sup>. Signal collection time was 5 s.