Supporting Information

Fabrication of Trans-scale Bimetallic Synergistic Enhanced Raman Scattering Substrate with High Surface-enhanced Raman Scatting Activity

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1. Experimental section

Electron beam evaporation The evaporation of silver film on the inverted pyramid pits substrate was carried out on an electron beam evaporation system (DZS-500, Shenyang scientific instrument Co., Ltd) at a rate of $5.5 \sim 6.5$ Å/s (duration is set to 50 s). The inverted pyramid pits silicon wafer substrate and gold film coated inverted pyramid pits substrate were obtained from MaRS technology Co., Ltd. The inverted pyramids exhibit a fixed apex angle (70.5° along the (111) direction of Si) and the period is 2 µm.

Raman detection The Raman experiments were performed on a Renishaw inVia Reflex equipped with peltier-cooled charge-coupled device (CCD) detectors and a Leica microscope. Samples were excited with a DPSS laser (532 nm) with a spot size of approximately 2 μ m and power is about 50mW. Subsequently, the Raman scattering of the analytes were tested. The stock solution of 10⁻³ M R6G (Rhodamine 6G, Sigma, analytical grade) and 1000 ppm BPA (Bisphenol A, Sigma, analytical grade) were prepared in ethanol, respectively. Different concentrations of R6G and BPA were diluted with ethanol. For Raman detection, 10 μ L of R6G or BPA working solution were dropped on the surface of substrates. After these substrates dry at room temperature, the Raman spectra can be collected.

2. Characterization of TBSERS



Figure S1. Typical SEM images of TBSERS.

3. R6G Raman scattering collected on silicon wafer surface



Figure S2. 1M R6G Raman scatting collected on silicon wafer surface (1s integration times with 0.25 mW of 532 nm laser radiation focused with a ×50 objective).



4. Raman signal mapping view of TBSERS substrate

Figure S3. SERS spectra 3D view of 10⁻⁶ M R6G solution collected on TBSERS substrates; B) Raman signal mapping view of 10⁻⁶ M R6G at 1649 cm⁻¹.

5. The Ultraviolet-visible absorption spectra of BPA solution.

Ultraviolet-visible absorption spectra of BPA (100 ppm in ethanol) was recorded on TU-1901 UV-Vis spectrophotometer (Beijing Purkinje General Instrument Co., Ltd., China).



Figure S4. UV absorption spectra of BPA solution (100 ppm in ethanol).

6. Detection of BPA in bottled water sample using the proposed method

The bottled water including mineral water and pure drinking water were purchased from a local supermarket. They were stored unopened at room temperature until detection. As shown in Table R1, the concentrations of BPA in the spiked water samples determined by the proposed method were in good agreement with their added concentrations, along with the quantitative recoveries of 90.0–120%.

Bottled water	Background Content (ppm)	Concentration		Recovery	RSD
		Spiked (ppm)	Found (ppm)	(%)	(%)
mineral water 1	ND	0.5	0.6	120	6
mineral water 2	ND	1	1.2	120	3
mineral water 3	ND	5	4.7	94.0	5
pure drinking water 1	ND	0.5	0.5	100	5
pure drinking water 2	ND	1	0.9	90	4
pure drinking water 3	ND	5	5.2	104	4

Table R1. Detection of BPA in bottled water sample using the proposed method (n = 5)

ND: not detected