Electronic Supplementary Material (ESI) for Reaction Chemistry & Engineering.

Supporting information for:

Microplasma synthesized gold nanoparticles for surface enhanced Raman spectroscopic detection of methylene blue

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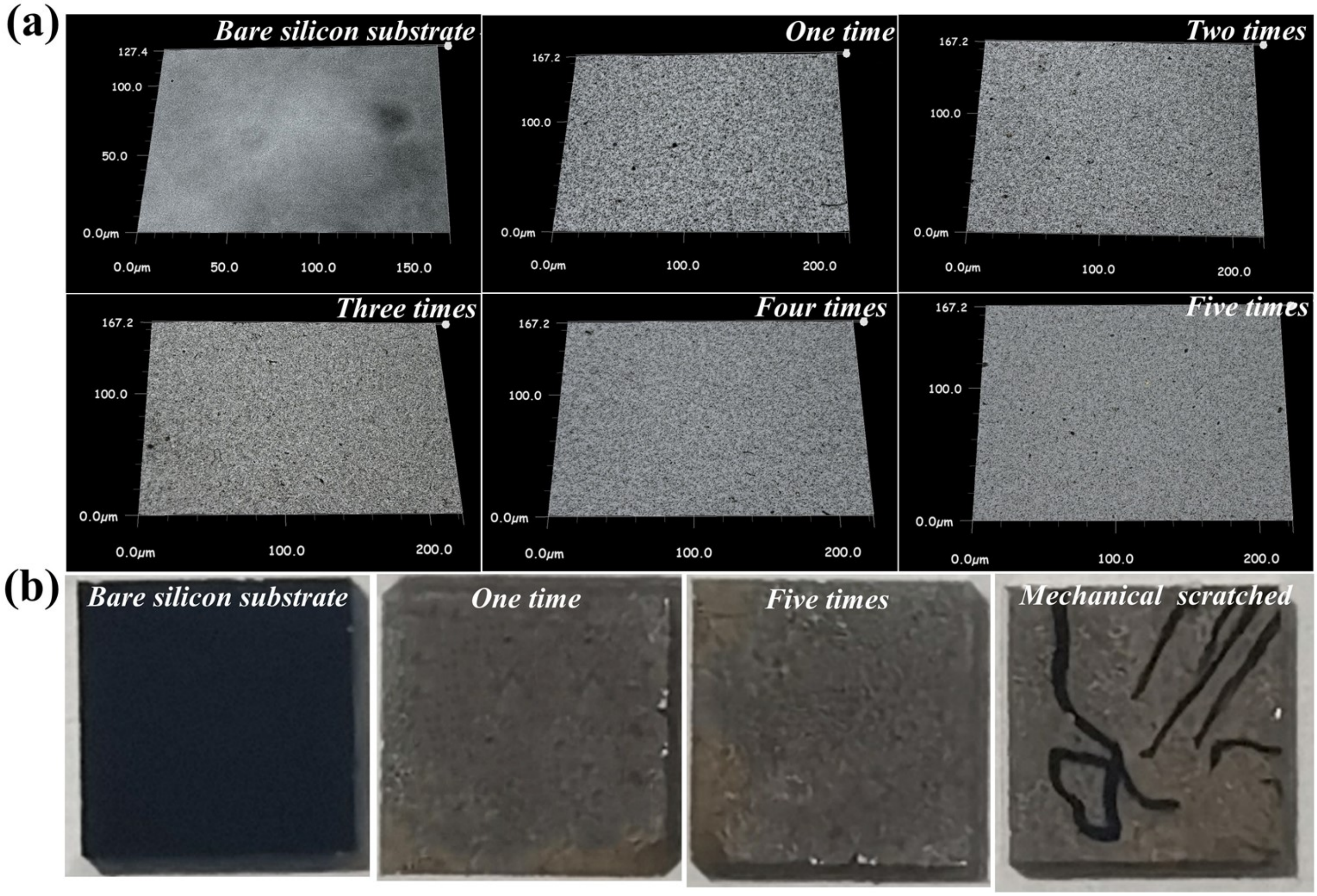

Fig. S1 (a) Representative SDFM images of the SERS substrates after different times of usage; (b) The photographs of the SERS substrates after different treatments.

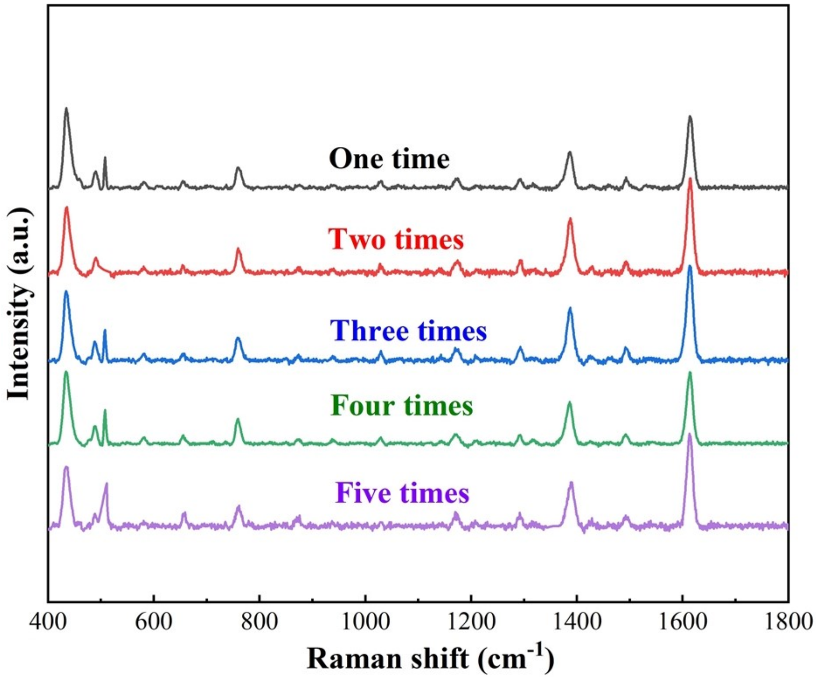

Fig. S2 Raman spectra of the MB molecules detected using the SERS substrates after various recycle times.

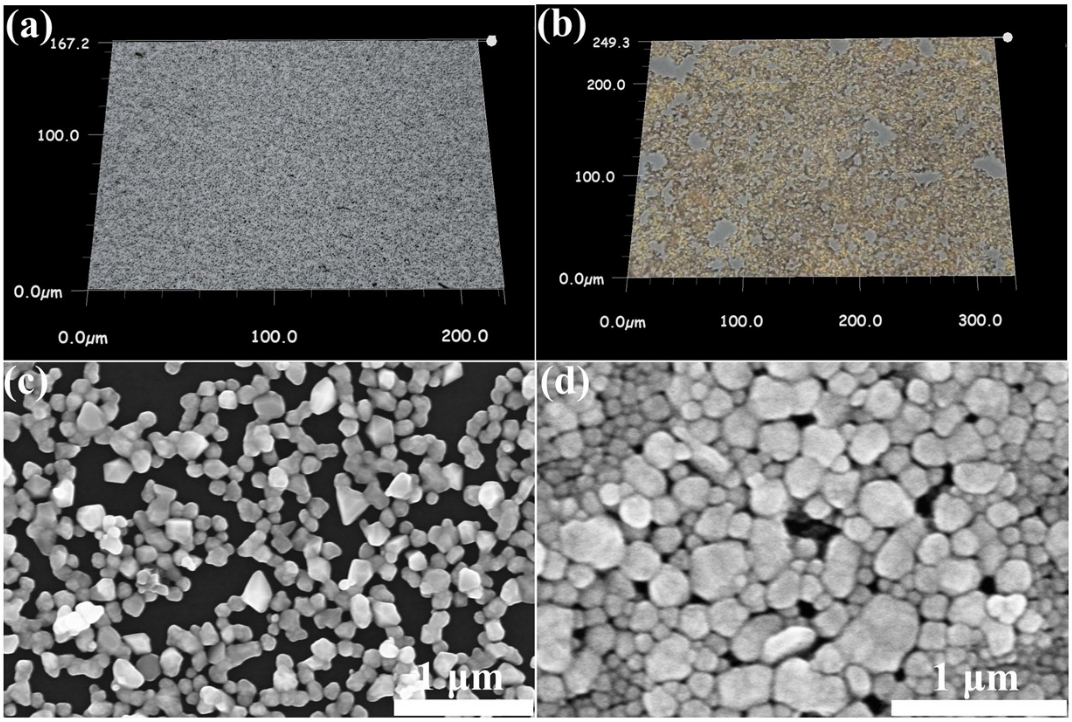

Fig. **S3** (a-b) SDFM images of the AuNPs-based SERS substrates dried for 2h: (a) at room temperature, (b) at 80 °C; (c-d)

The corresponding SEM images of the SERS substrates dried for 2h: (c) at room temperature, (d) at 80 °C.

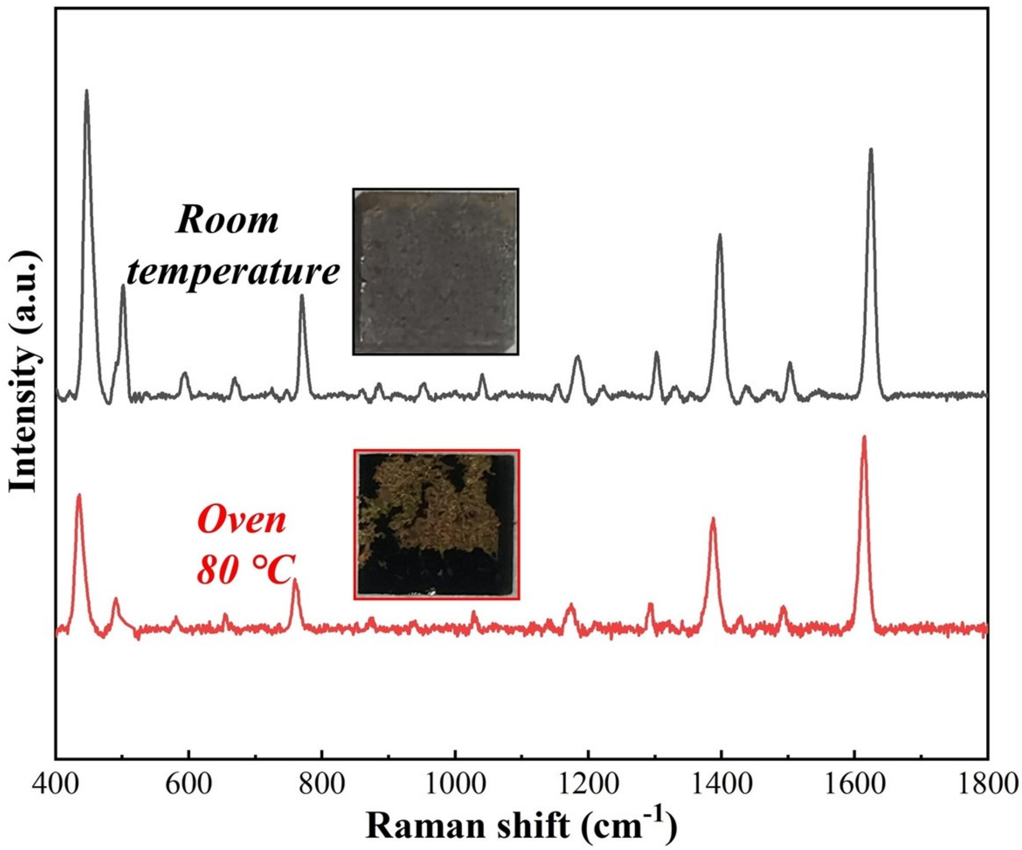

Fig. **S4** SERS spectra of the MB molecules absorbed on substrates dried at room temperature as well as at 80 °C.

Table S1 Summary of emission lines from the OES spectrum

Species	System	Transition	Wavelength
OH	3064 Å system	$A\ ^2\Sigma^+ \rightarrow X\ ^2\Pi$	309.2 nm
N	Second positive system	$C\ ^3\Pi_u \rightarrow B\ ^3\Pi_g$	335.9 nm (0,0), 357.1 nm (0,1)
			380.2 nm (0, 2)
Au	Au I		397.7 nm, 405.4 nm
			472.1 nm, 516.9 nm
H	Balmer series	n→2s,2p	493.6 nm (H_β), 655.9 nm (H_α)
O		$3p^5P \rightarrow 3s^5S$	778.1 nm
N_2			592.6 nm
H_2O			618.76 nm
Ar	Ar I	4p→4s	696.8 nm ($1s_5$-$2p_2$), 707.1 nm ($1s_5$-$2p_3$)
			738.6 nm ($1s_4$-$2p_3$), 750.3 nm ($1s_5$-$2p_1$)
			763.7 nm ($1s_5$-$2p_6$), 772.3 nm ($1s_3$-$2p_2$)
			794.5 nm ($1s_3$-$2p_4$), 826.9 nm ($1s_2$-$2p_2$)
			843.4 nm ($1s_4$-$2p_8$)

Table S2 An overview of methods regarding Raman detection of MB molecules using the SERS method.

Materials	Methods	Particle size	Detection limit (M)	EFs	Advantages	Drawbacks	Reference
AuNPs	Microplasma	4~12 nm	10^{-11}	1.06×10^{8}	Simple, efficient, and green process High purity products	Low throughput	This work
Au-Cu alloy	Chemical reduction	~8 nm	~	3.2×10^{2}	Products with narrow size distribution and good reproducibility	Chemical contaminations Poor composition control	1
Au-Ag alloy	Laser ablation	4~11 nm	10^{-9}	$\sim10^{7}$	Simple, facile, and efficient process High purity products	Costly and complex setup Energy consuming process	2
Polymer encapsulated rhenium nanoparticles	Chemical reduction	~1.7 nm	10^{-5}	5.18×10^{2}	Effective, low-cost, flexible process Stable products	Toxic reducing agent Seriously aggregated products	3
MoS_2/TiO_2 nanoflowers	Hydrothermal	2~3 μm	10^{-13}	2.09×10^{6}	Simple and flexible process Controlled product properties	Complex operation Time/energy consuming process	4
Silver nanoparticles	Chemical reduction with physical evaporation	~300 nm	~	4.2×10^{7}	Simple process Stable products with good reproducibility	High temperature and time consuming process Products with impurities	5
Ag-Au NPs	Laser ablation	11~23 nm	10^{-10}	~	Efficient and flexible process High purity products	Costly and complex setup Energy consuming process Non-uniform aggregated NPs	6
Core/shell Ag-Au alloys	Chemical reduction and seed mediated growth	2.8~24.1 nm	10^{-7}	1.2×10^{7}	Simple and low-cost process Controlled product properties	Time consuming process Products with impurities and broad size distribution	7

References

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