Supporting Information

Highly Dense and Accessible Nanogaps in Au-Ag Alloy Patterned Nanostructures for Surface-Enhanced Raman Spectroscopy Analysis

Taeksu Lee[a], Soongeun Kwon[a] and Jae-Jong Lee[a,*]

[a] *Department of Nano Manufacturing Technology, Korea Institute of Machinery and Materials (KIMM), Daejeon 305-343, Republic of South Korea*

E-mail: jjlee@kimm.re.kr

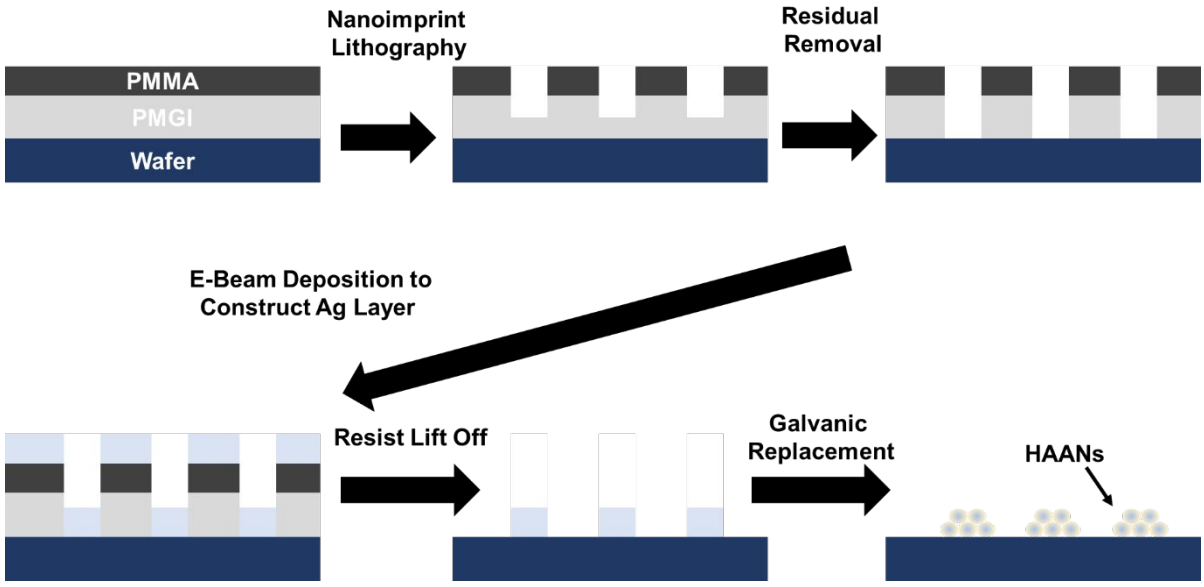

Figure S1. Schematic illustration of overall process in this research: Nanoimprint lithography (NIL) was performed and resist residual was removed by consecutive dry etching and wet chemical dissolution of PMGI (polymethylglutarimid) E-Beam deposition was conducted to construct Au layer. Lift-off process to erase resist with AZ-MIF 300 developer was performed to generate SDAs. Thereafter, HAuCl4 aqueous solution was injected to HAANs via galvanic replacement.

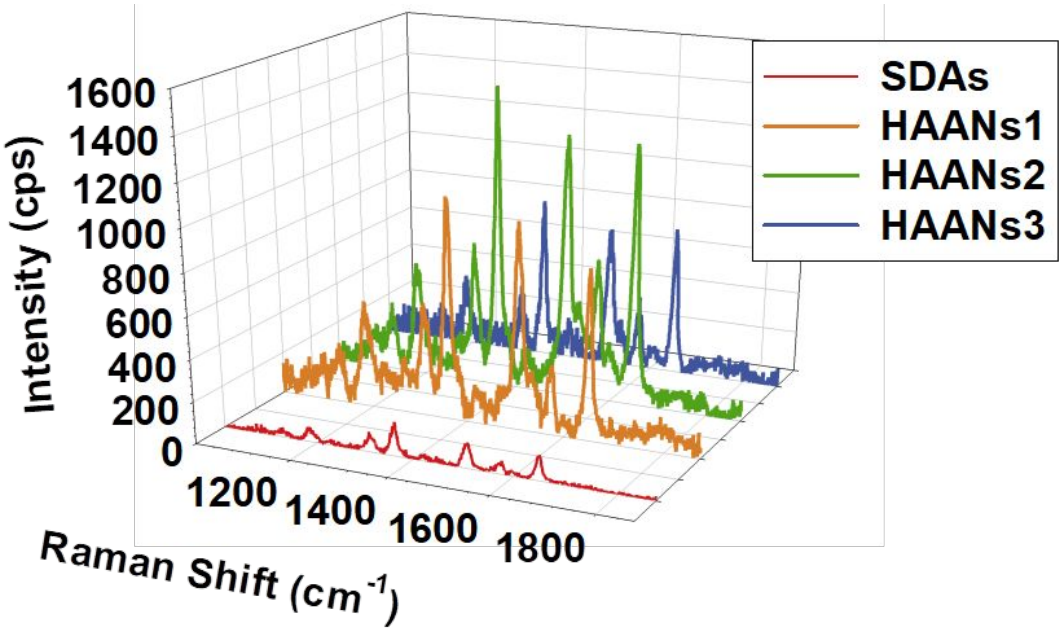

Figure S2. Representative SERS spectra of 10^{-5} M rhodamine 6G treated SDAs and HAANs (wavelength : 514 nm, laser power : 0.1 mW and integration time : 10 sec)

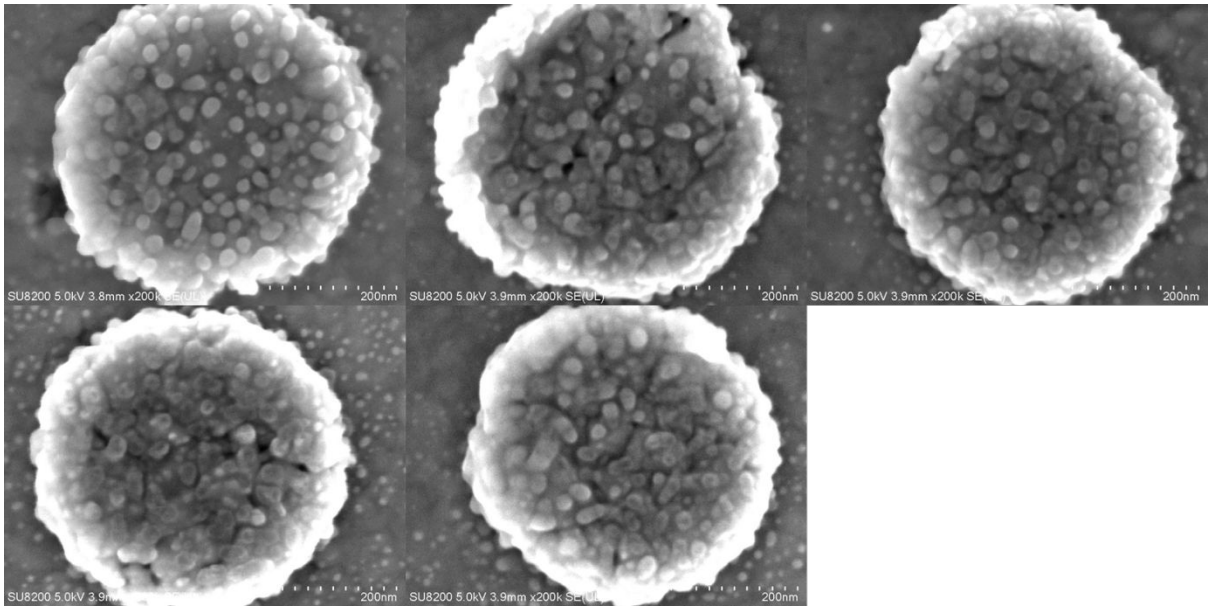

Figure S3. Representative SEM images of HAANs2 to count and measure nanogap

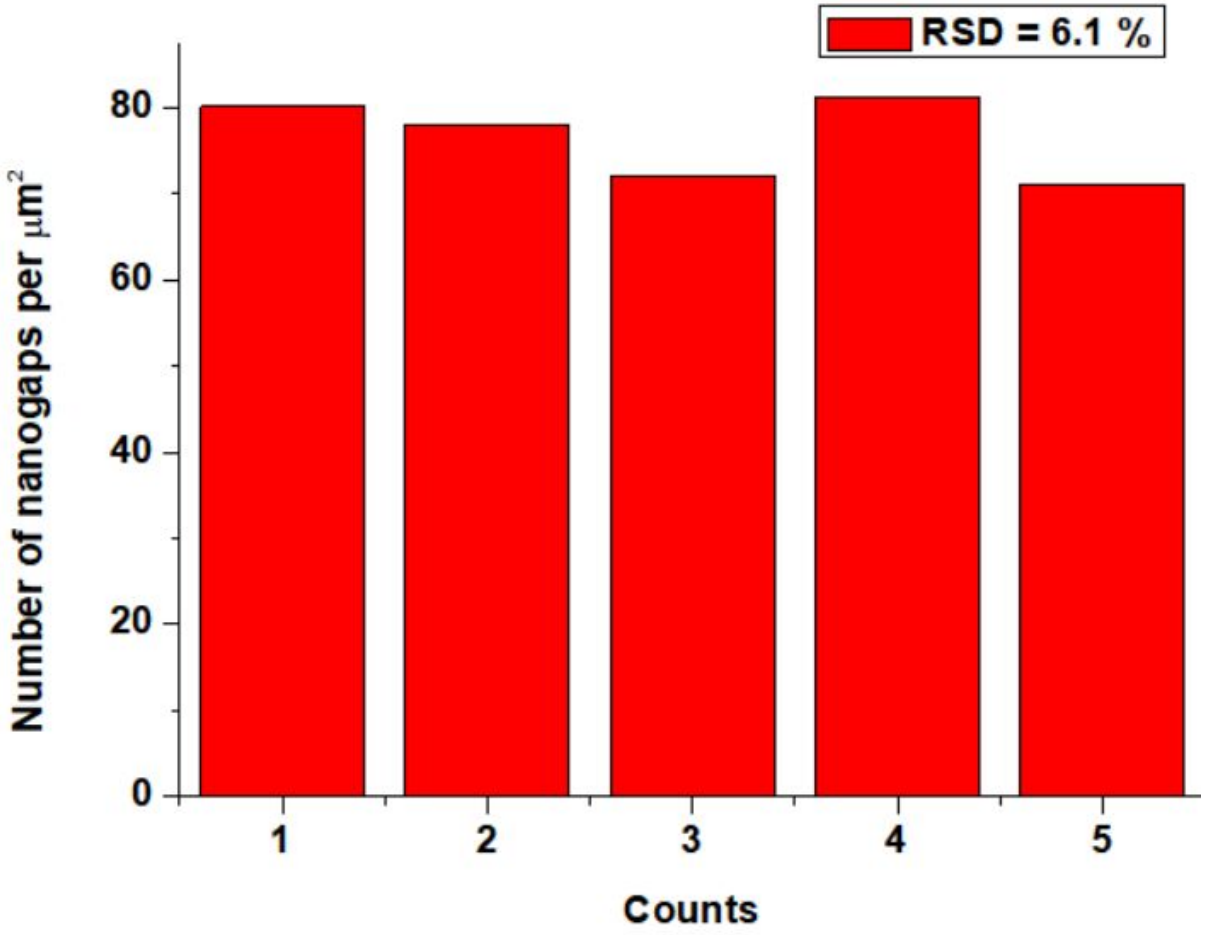

Figure S4. The statistical analysis of the number of nano-gaps with unit area (1 μm^2) at different positions of HAANs2.

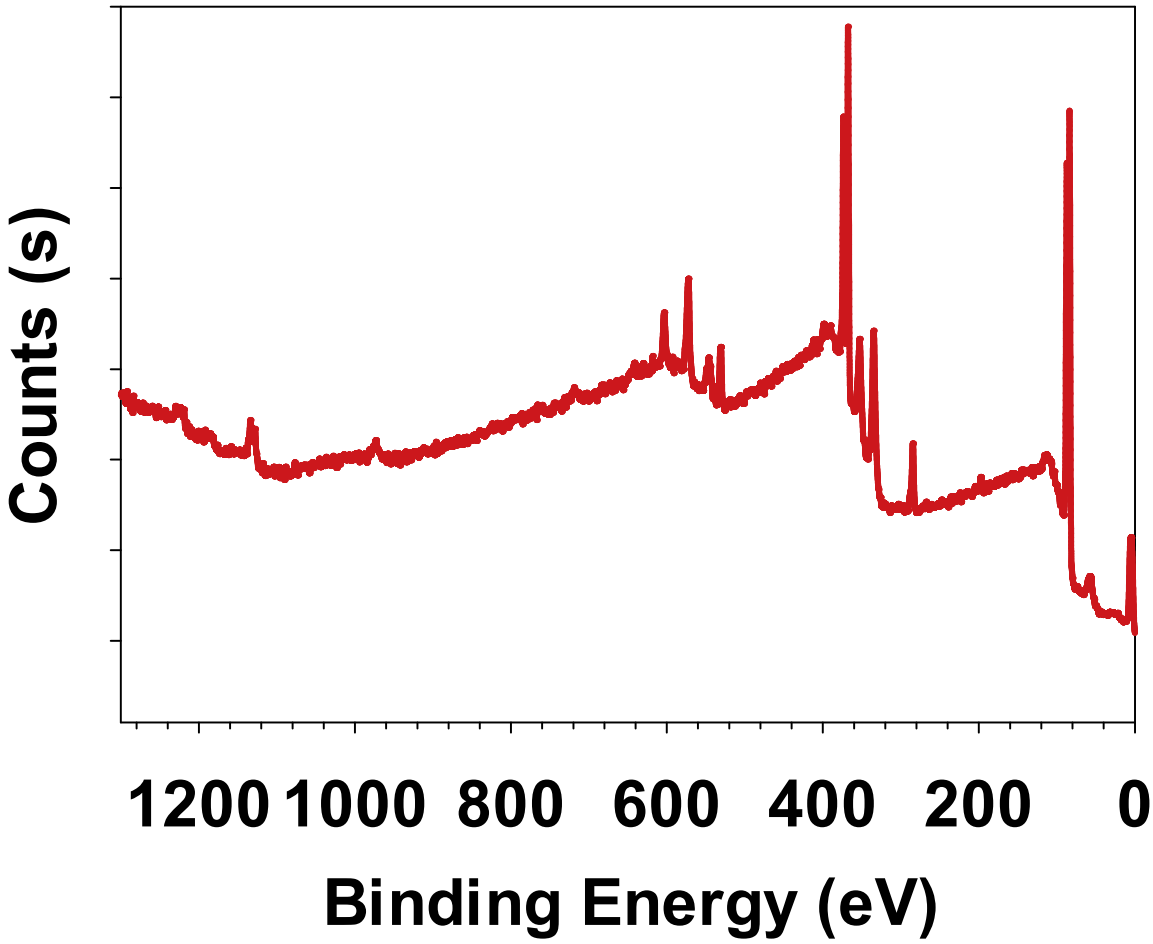

Figure S5. Overall X-ray photoelectron spectra of HAANs2.

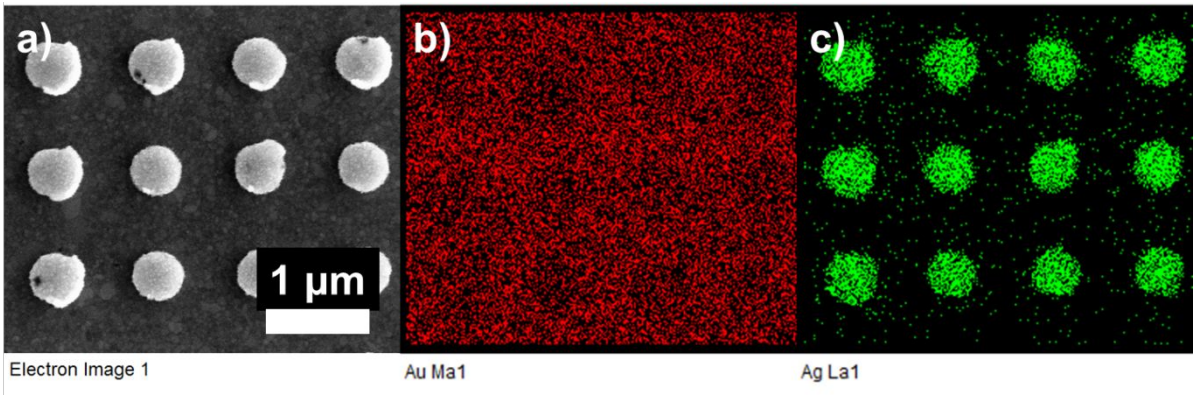

Figure S6. a) SEM images of HAANs2 and corresponding elemental mapping analysis of HAANs2. b) Red color indicates Au and c) green color reveals silver.

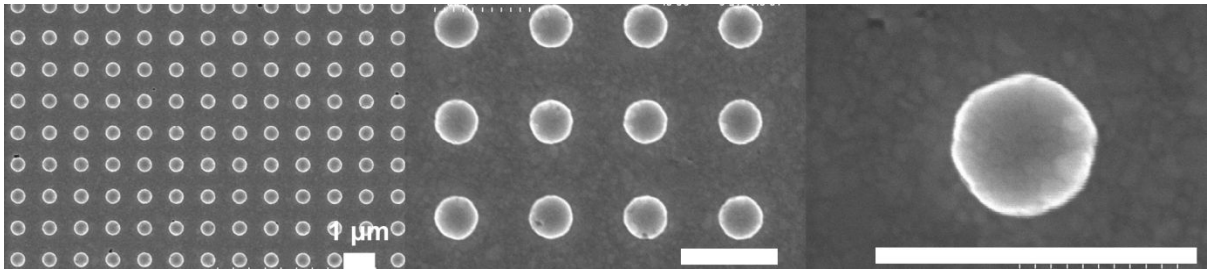

Figure S7. Representative SEM images of PVP-HAANs2. SDAs were first immersed in 1 wt % PVP solution for overnight and underwent galvanic reaction when all the conditions were as same as those of HAANs2.

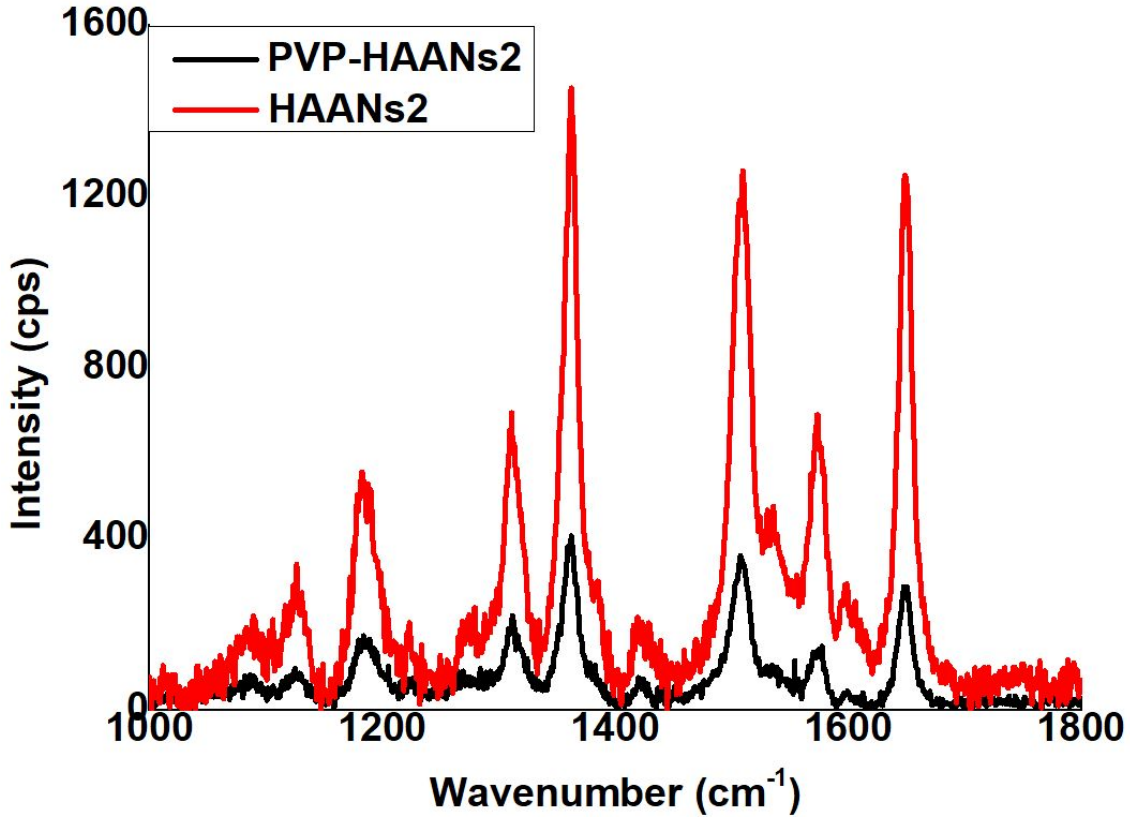

Figure S8. Raman spectra of PVP-HAANs2 (black line) and HAANs2 (red line) after 10^{-5} M R6G treatment (wavelength : 514 nm, laser power : 0.1 mW, integration time : 10 sec).

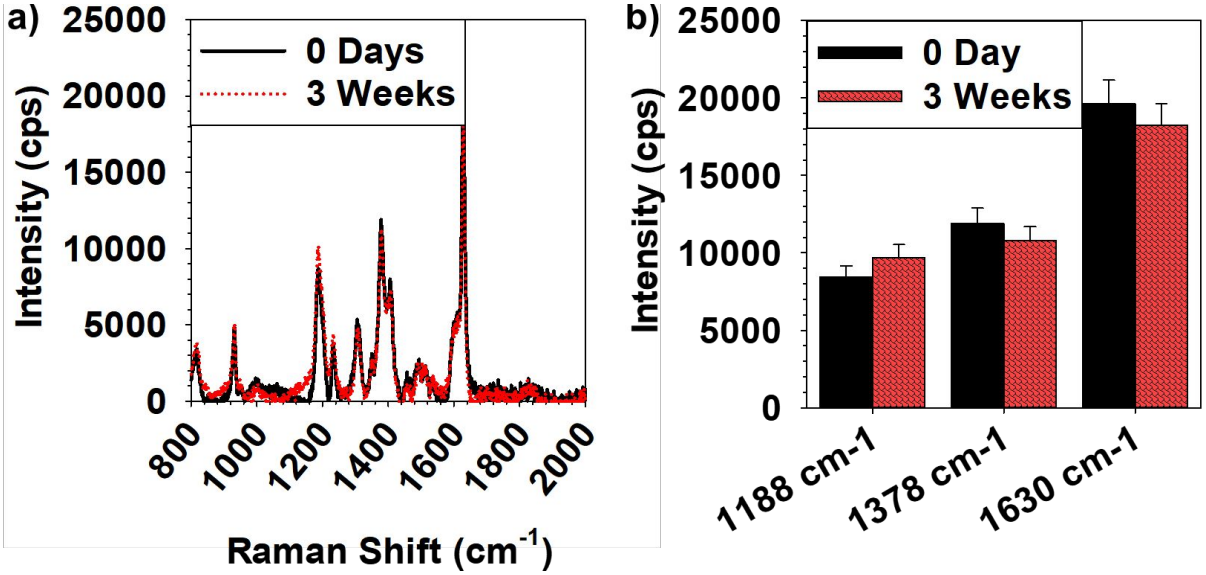

Figure S9. Time dependent stability of HAANs2. The HAANs2 were stored at room temperature and 10^{-5} M MG was used to obtain Raman signal.

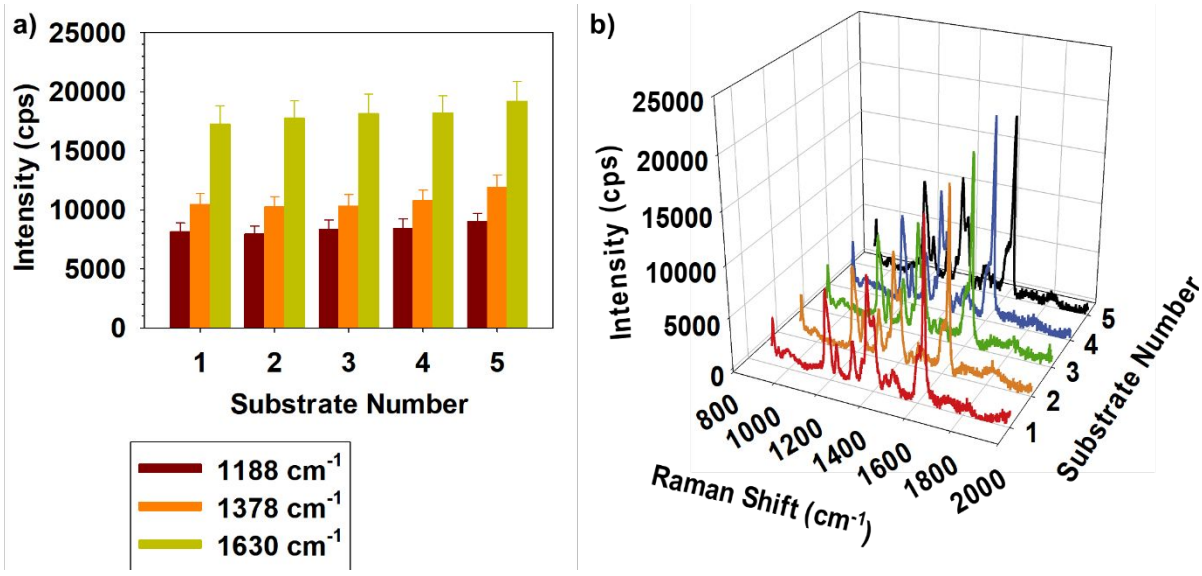

Figure S10. Synthetic reproducibility of HAANs2. The peak intensities of 10^{-5} M MG were measured from different 5 substrates from different SDAs, under 514 nm laser irradiation.

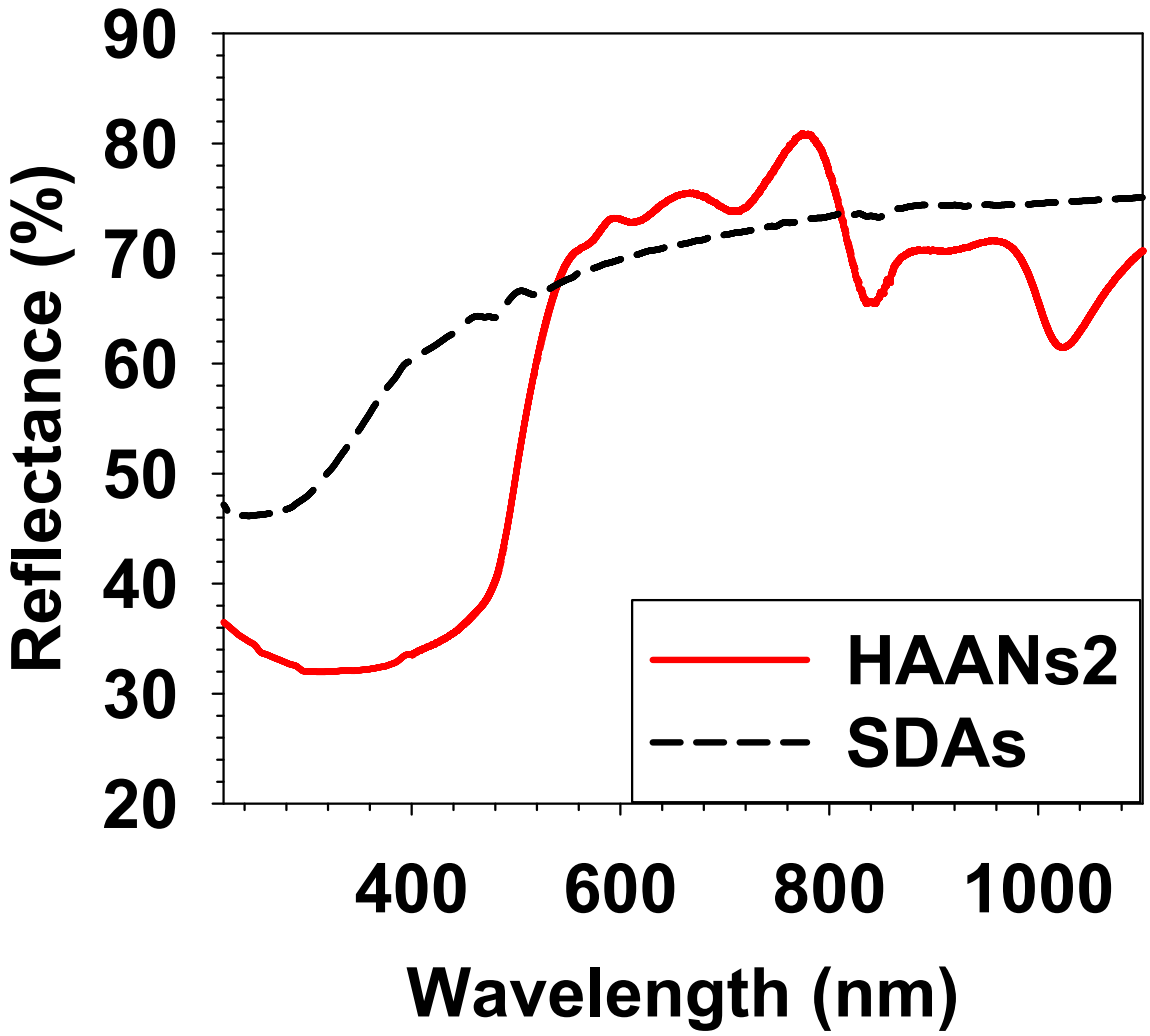

Figure S11. The reflectance spectrum of SDAs (black dashed line) and HAANs2 (red line).

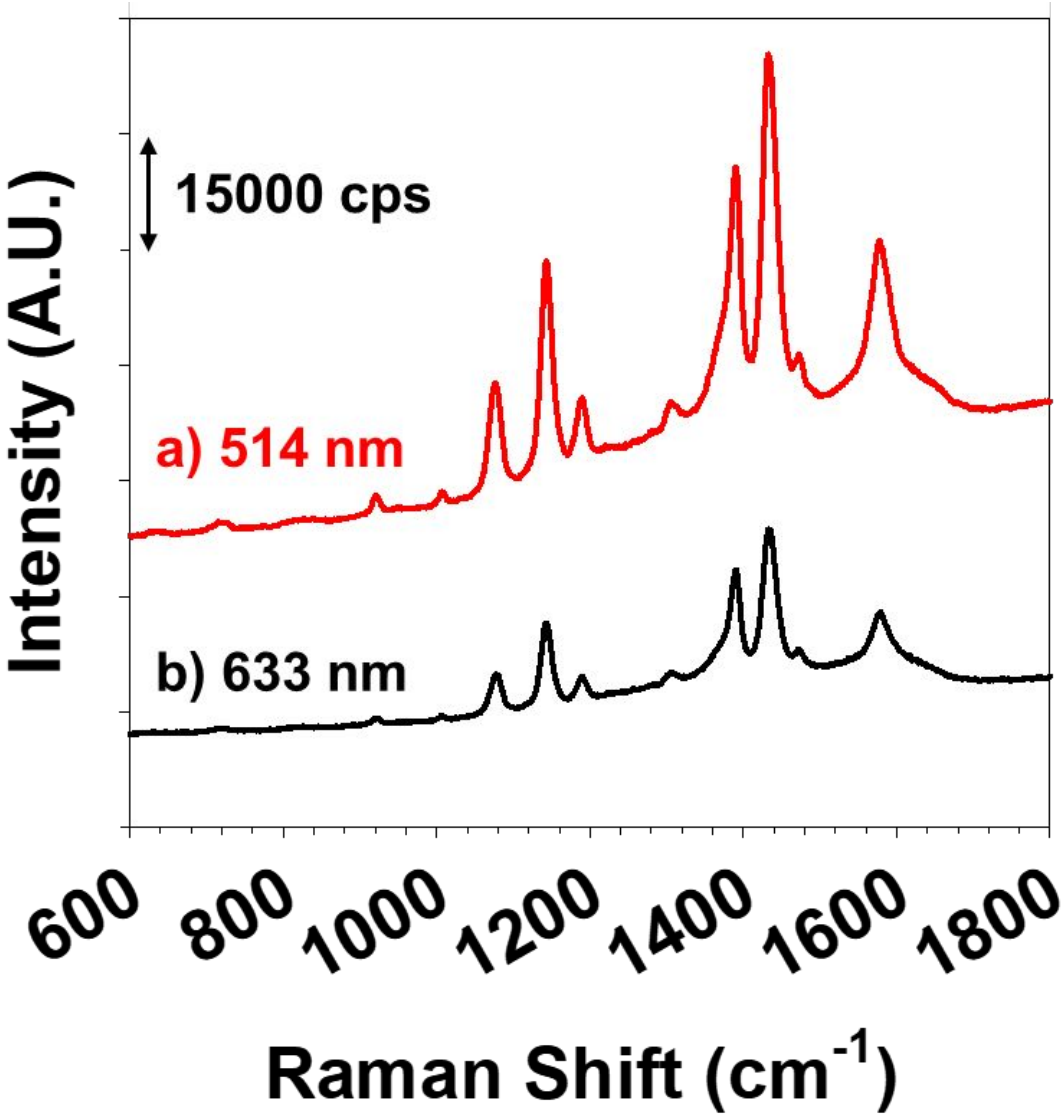

Figure S12. Representative Raman Spectra of 1×10^{-5} M p-ATP treated HAANs2 under the irradiation of a) 514 nm and b) 633 nm laser.

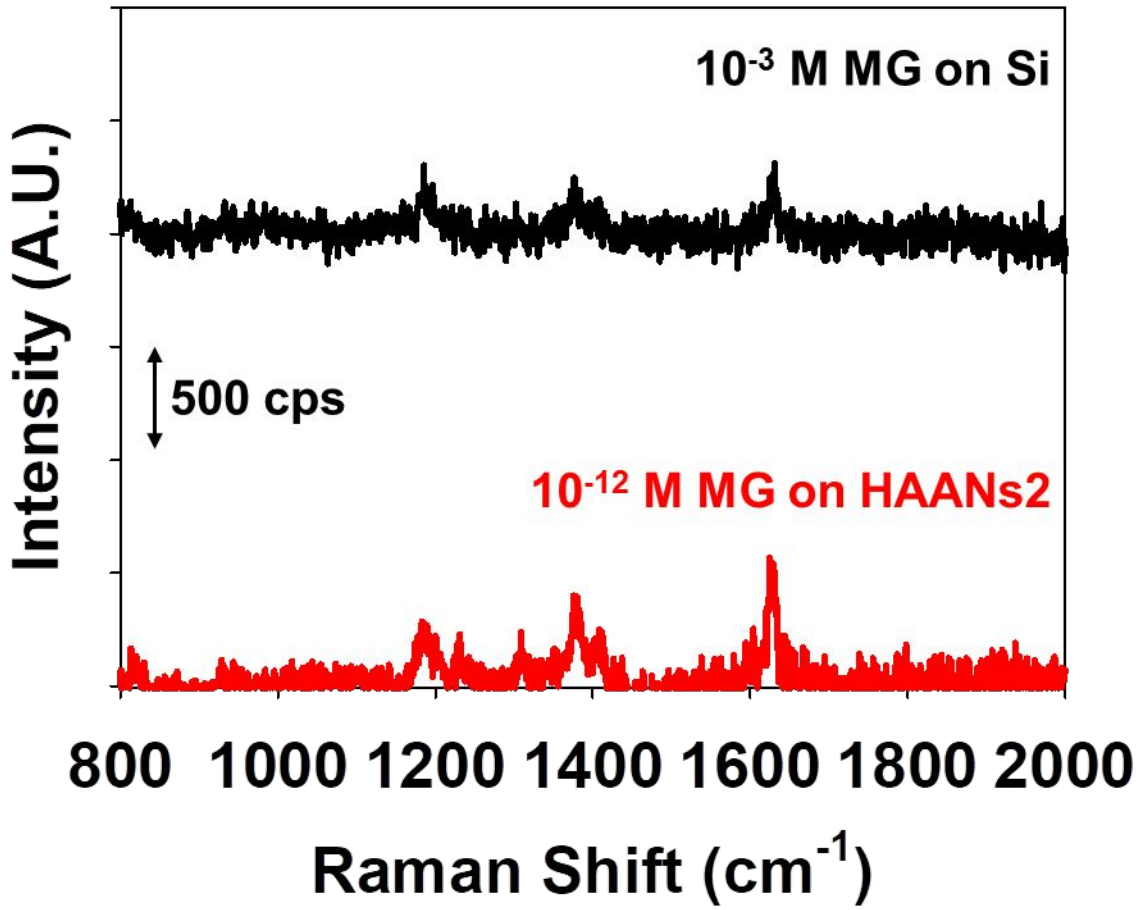

Figure S13. Representative Raman spectra of 10^{-3} M MG on bare Si wafer and 10^{-12} M MG on HAANs2

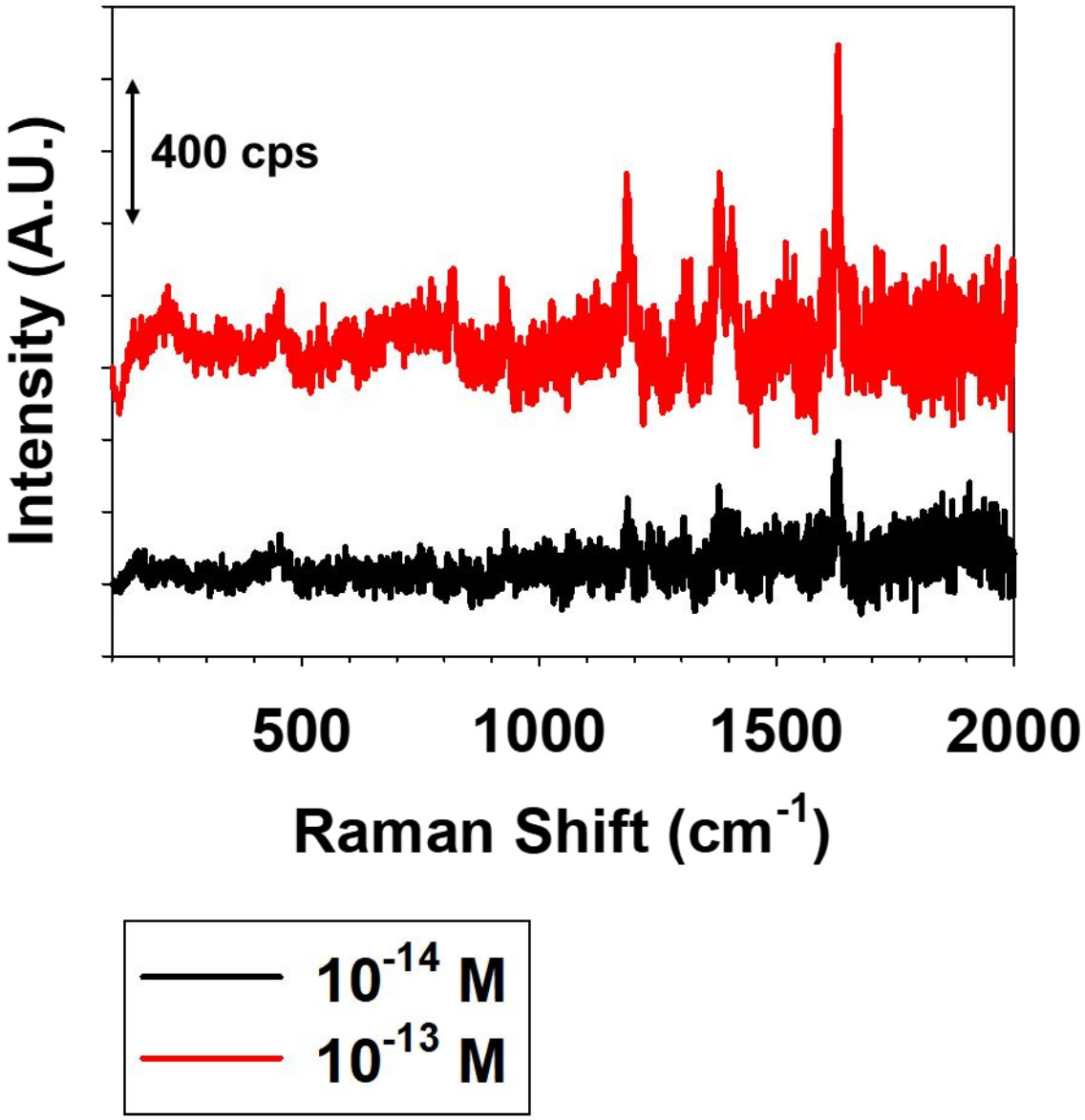

Figure S14. SERS spectra of 1×10^{-14} M and 1×10^{-13} M of MG treated HAANs2 in **Figure 6a.**

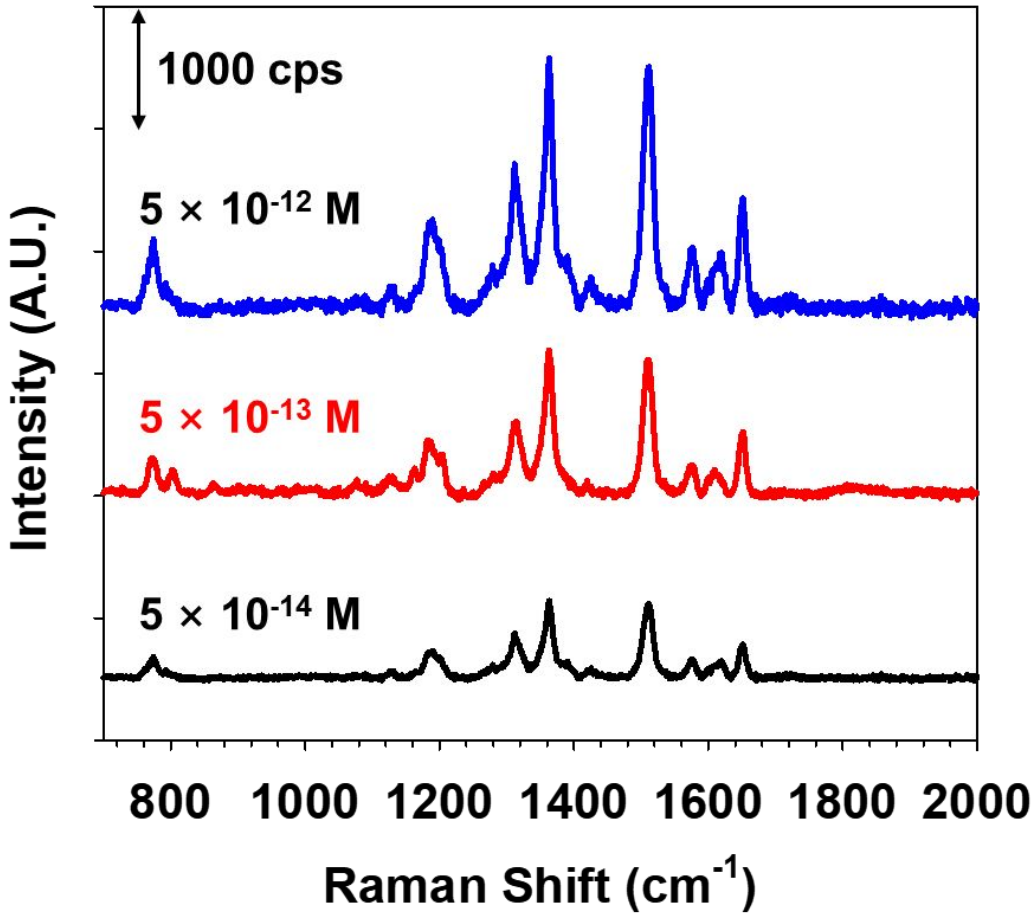

Figure S15. Different SERS spectra of HAANs2 with respect to the different rhodamine 6G concentrations (5 x 10^{-14} M to 5 x 10^{-12} M) to determine limit of detection (LOD) under 514 nm irradiation.