

Supplementary Materials

Trends in Application of SERS Substrates beyond Ag and Au, and Their Role in Bioanalysis

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Remark

Our classification of substrates into conventional and non-conventional is based on

- 1) Historic perspective of SERS development since initially SERS was discovered on silver rough film and most of the first, including the most cited, applications of SERS in biodetection were conducted on silver or gold substrates/nanoparticles (glucose and anthrax biomarker by Van Duyne's group, PSA by Porter's group, and many others)
- 2) We also did a Scopus search for “SERS” and “silver” or “gold” or “silicon” or “copper” or “aluminum” or “platinum” or “palladium” For instance Scopus shows 11,406 document results TITLE-ABS-KEY (sers AND silver) The numbers of documents found for each of those substrate are 11406 for Ag, 9974 for Au, 2256 for Si , 1066 for Cu , 811 for Al, 474 for Pt, 205 for Pd. Therefore, so called “conventional” “Au and Ag substrates are on average about 11 times more common in SERS research literature than average “non-conventional” SERS substrate. Since there is a factor x4.5 difference between search results for Au and for Si, there is hardly much doubt where to draw a border between conventional and non-conventional substrates. Even gold substrates alone are likely to be about twice as common in title, abstract and keywords of SERS related publications as all other 5 above mentioned “non-conventional substrates” Therefore term “conventional” (synonym of the most common or the most frequently used) for silver and gold substrate materials is well justified.

Table S1. Multi Elemental Silicon-based SERS substrates (NP: Nanoparticles).

Substrate	Analyte	Enhancement Factor	LOD (mol/L)	Details (Laser, power; acquisition time, RSD and etc)	Ref
AuNP@Si nanowire paper	R6G	1.00E+05	1.00E-08	633 nm; RSD <0.15. Detection limit for a pesticide: 2.5×10^{-7} M; acq t: 10s	[1]
AuNP@ Si	Phenylalanine	2.60E+04		785 nm; Laser power: 4 mW; acq t: 10 s	[2]
AuNP@ porous Si	p-MBA	3.70E+08		785 nm; RSD 6.7%; acq t: 4s; Laser power 5mW; Au layer is 30nm; with the incr. in the thickness of the Au layer intensity signal decr.	[3]
AgNP@Si	p-MBA	1.00E+08		785 nm; power: 5mW; acq t: 10s Ag layer thickness: 100nm; RSD 7%;	[4]
Al ₂ O ₃ @AgNP @Si nanocones	R6G	1.00E+09	1.00E-12	532 nm; Power: 0.05mW Al ₂ O ₃ thickness 2nm; good stability after 5 months.	[5]
AgNP@Si wafer	Pb ²⁺	2.00E+06	9.90E-11	633 nm; Power: 0.2mW; acq t: 1s, LOD for Hg ²⁺ : 8.4×10^{-10}	[6]
AgNP@Si nanowires	R6G	1.00E+07	1.00E-09	785 nm	[7]
AgNP@meso porous Si	R6G	1.00E+10	1.00E-12	514.5nm; acq t: 10s; detection range: (10^{-8} M; 10^{-12} M)	[8]
AgNP@meso porous Si	Cy5	1.00E+07	5.00E-09	514.5nm; acq t: 10s	[9]
AuNP@porous Si	Penicillin G	5.00E+07	1.00E-09	532 nm; Power: 10mW; acq t: 3s RSD 4.5%;	[10]
Graphene@A gNPs@Si (Aptosensor)	ATP	8.30E+06	1.00E-12	633 nm; Power: 20mW; acq t: 1s; substrate size: 140nm; RSD: 10.5%	[11]
Si nanowires@GrapheneNP	R6G	1.00E+07		785 nm; Si is d=50nm cylinder; Au d=60nm sphere on the top of cylinder	[12]
CuNP@Si wafer	R6G	2.29E+07	1.00E-09	633 nm; acq t: 1s; RSD <20%; CuNP d=20nm	[13]
AgNP@Si nanowires	R6G	6.00E+05	1.00E-11	633 nm; power: 1 mW ; acq t: 10s; SiNW thickness is 20 um; AgNP diameter is 40-120nm	[14]
AgNP@porous Si photonic crystals	R6G	1.20E+06		633 nm; power 1mW; acq t 3s; RSD 8%; 3.58 times better EF than just porous Si w/o photonic crystals.	[15]

	Picric Acid		1.00E-08	532 nm	[15]
AgNP@Si nanowires	R6G	4.12E+09	5.00E-09	532 nm; power: 0.05 mW; acq t: 10s;	[16]
AgNP@mesoporous Si	R6G; Cy5	1.00E+08	1.00E-11	514.5 nm; acq t: 10s;	[17]
Ag@porous Si wafers	R6G	2.00E+08	1.00E-10	514.5 nm;	[18]
Ag@mesoporous Si	Cy3	5.30E+12	1.00E-14	785 nm; acq t: 10s; size of AgNP 5-100nm;	[19]
AgNP@porous Si	NAGase	1.06E+05	1.97E-06	785 nm; SERS recovery in comparison to FL technique is 85–98%	[20]
PdNP@PSi	imidacloprid (insecticide)	1.20E+05	1.00E-09	785 nm; acq t: 20s; substrate stable for at least two weeks;	[21]
AgNP@PyrSi	R6G	1.20E+09	1.00E-10	532 nm; acq t: 10s; laser spot: 2um;	[22]
Au@porousSi	R6G	1.00E+06	1.00E-09	785 nm; acq: 10s	[23]
AuNP@Si nanowire arrays	RhB	1.00E+09	1.00E-11	514 nm; power: 0.5mW; acq t: 10s	[24]
AgNP@Si nanowires	R6G	2.30E+08	1.00E-14	633 nm; 8mW; acq t: 50s.	[25]
SiNP@AgNP @SiC (sandpaper) (Immunoassay)	PSA (prostate specific antigen)	3.14E+05	5.60E-17	785 nm; Si NP size: 50 to 200 nm; LOD: tumor markers PSA: 1.79 fg/ml; AFP:0.46 fg mL ⁻¹ ; CA19-9 1.3 × 10 ⁻³ U mL ⁻¹	[26]
Fe ₃ O ₄ @AgN P@Si nanopillars	malachite green	1.00E+05	1.00E-08	532 nm; acq t: 10s; thermoelectrical cooling CCD	[27]
AuNP(core)@SiO ₂ (shell)	Pyridine	1.00E+06		632.8 nm; power: 1mW; Au core: 55 nm; silica shell: 23 nm; two dimers distance is 2nm	[28]
PtNP(core)@SiO ₂ (shell)	Pyridine	1.00E+05		632.8 nm	[28]
PdNP@porous Si	R6G	100		488 nm; power: 5 mW; acq t: 30s; Pd size 40-86nm;	[29]
PtNP@porous Si	R6G	33		488 nm; power: 5 mW; acq t: 30s; Pt size 56-68 nm;	[29]
PtNA@Si wafers	R6G	5.00E+04		514.5 nm; Power: 0.17 mW, acq t: 30 s; 632.8nm laser has half of 514.5nm's EF	[30]
PdNP@porous Si	TNT	2.54E+06	1.00E-07	514 nm; power:10 mW, acq t:60 s min gap size (4nm) at etching	[31]

				time of 6min; by weight: Si60%;Pd37%	
Ag&Pd alloy NP@Gradient Porosity Si	E.Coli	2.30E+05	1 Cfu/ml	785 nm; acq t: 10s; best immersion time: 180s; Ag & Pd NP 10nm	[32]
PdNP@ pyramidal Si	R6G	1.91E+06	1.00E-11	632.8 nm; power: 0.1mW; acq t: 10 s; Pd thick in 60min is 300nm	[33]
AuNP@FePt @SiO ₂	Adenine	2.00E+07		633 nm; overall substrate: 30nm	[34]
AuNP@Si nanorods	R6G	3.30E+07	1.00E-10	633 nm; power: 1.3 mW; acq t:5 s; RSD 3.9–7.2%; SiNR length: 1.3 μm, diameter of 200 nm	[35]
AuSiO ₂ nanorods @ silica gel	R6G	3.60E+09	1.00E-16	532 nm; power: 1 mW; acq t: 10 s; Silica gel : 1-2nm, AuSiO ₂ (70 nm core— 102 nm shell); LOD of CEA antigen 0.86 fg/ml	[36]
Au(core)@Si O ₂ (shell)	glucose	1.20E+08	1.00E-12	532 nm; power: 20mW; acq t: 10s; AuNP 36nm; silica layer 1- 2nm best EF than thicker silica.	[37]
AgNP@Si wafer	R6G	2.90E+07		514 nm; acq t: 1s; RSD <15%; AgNP size is 40nm;	[38]
AgNC@SiO ₂	R6G	1.26E+06		532 nm; power: 0.15mW; acq t: 1s; Ag NCs (~50 nm); SiO ₂ layer is 1.4nm	[39]
AuNP@stellat e mesoporous SiO ₂ (Immunoassay)	Ferritin antigen	1.68E+06	6.67E-17	785 nm; power: 14mW; acq t: 1s; uNPs (d=10nm) covering SMSiO ₂ (d=100nm); RSD 22.94%; LOD of ferritin 3.16*10^-14 g/ml	[40]
glu@3- MPBA@AuN anoFlower@S i wafer (biosensor)	miRNA 122		7.75E-18	785 nm; size of AuNF@Si is 100nm; linear range 10aM- 100pM; RSD 0.59% for 6 different samples	[41]
SiO ₂ (core)@AuNP s (shell) Immunoassay	E. coli O157:H7		100 cells/ml	785 nm; power: 10 mW, acq t: 10 s; substrate is 180nm; assay time 15min; SERS based LFIA strip	[42]
AuNP@SiNP	RhB	1.00E+06	1.00E-10	785 nm; power: 2mW; acq t: 20s; Au film (10nm) on self assembled SiNP (120nm)	[43]
AuNP@PSiO ₂	R6G	6.50E+07		633 nm; power: 1mW; 350nm pores; thickness 60nm; RSD 12%;	[44]

SiO_2 NP@AuNP	R6G			633 nm; power: 1mW; adding SiO_2 increases EF by 3 times.	[45]
SiO_2 NP@AgNP	R6G			514nm; power: 1mW; with SiO_2 3 times better EF than bare AgNP	[46]
$\text{AgNP@Au}(\text{core})@\text{SiO}_2(\text{shell})$	RdB	1.03E+05	1.00E-11	785 nm; acq t: 10s; core diameter of 70 nm and a 5 nm silica shell.	[47]
Ag@ SiO_2 Nanocubes	Melamine for LOD R6G for EF	1.93E+07	4.76E-07	632.8 nm; acq t: 30s; SiO_2 shell is 2.56nm; AgNP has d=56nm; RSD 10.3%; LOD 0.06mg/L melamine	[48]
Ag@SiNWs	4-ABT	1.10E+06		632.8 nm; Power: 4.4mW; acq t: 1s; Ag-coated wire d=150nm; Si d=110nm; RSD 14%	[49]
AgNano assemblies@Si	R6G	6.00E+13	1.00E-16	532 nm; power: 0.6mW; acq t: 0.1s; RSD: 7.3%	[50]
Graphene@AgNP@Si fiber	BPA (bisphenol A)	6.00E+06	4.38E-09	633 nm; acq t: 4s; AgNP size 60nm; LOD 10^{-6} ppm; for 60 sample study recoveries of BPA from 89% to 116%	[51]
Graphene@Si nanowires	R6G	1.00E+05	1.00E-07	532 nm; power: 1mW; acq t: 10s; NW length 2.56 um; RSD 2.6%	[52]
AgNP@mesoporous Si	R6G		1.00E-09	514.5nm; power: 5mW; acq t: 10s; mesoporous Si 100 times better SERS signal than crystalline Si.	[53]
AgNP@Si nanowires	R6G	3.00E+08	1.00E-08	514 nm; power: 0.5mW; SiNWs size 1.7um; AgNP d=5-8 nm	[54]
AgNP@Si	BPE (bipyridyl ethylene)	1.00E+15	1.00E-10	632.8nm; power: 5 mW; acq t: 5 s; AgNP d=12-22nm;	[55]
AgNS@Cu ₂ O @Si nanocones	4-ATP	1.00E+07	1.00E-14	785nm; power: 2mW; acq t: 1s; nanocones: 600-700nm	[56]
Au film@PSi	R6G	1.00E+12	1.00E-15	785nm; RSD: 6.2%; d=160nm	[57]
Au&Ag alloy NP@Si	MB (molecular beacon)	1.00E+10	1.00E-11	785nm; 30mW; acq t:5s; RSD <15%	[58]
Ag film@Laser Textured SiNP	4-methylbenzene thiol	5.50E+06		532nm; Ag optimum thickness 20nm	[59]
AgNP@Si Nanopillars	R6G for LOD; PhSH (thiophenol) for EF	2.40E+08	1.00E-13	785nm; Si nanopillars of size 80 nm, height 1 um, periodicity 100 nm	[60]

Graphene@Ag film@Laser Textured SiNP	R6G	2.60E+07	1.00E-10	532nm; power: 10mW; acq t: 2s; Ag 30nm; stable for more than 50 days	[61]
AgNP@ZnO nanowires @Si nanorods	R6G	1.40E+07	1.00E-12	532nm; power: 0.37mW; acq t: 5s; RSD 6%; SiNR d=250nm; ZnO NW d=80nm	[62]
SiC@Ag(film) @AgNPs (Immunoassay)	PSA, PSMA, and hK2	1.14E+07	1.50E-08	785nm; LOD 0.46 fg mL ⁻¹ (PSA); 1.05 fg mL ⁻¹ (PSMA) and 0.67 fg mL ⁻¹ (hK2)	[63]
AgNP@Si	N. gonorrhoeae	1.00E+08	LOD 100 cfu/ml	785nm; acq t: 6s; Si wafers (3mmx3mm used) & AgNP layer 100nm	[64]
Au@ZnO@Si	Neoprotein		1.60E-09	785nm; power: 5mW: acq t: 10s; Au layer 90nm; ZnO layer; 1.4um	[65]
AgNP@Si wafer	miRNAs	2.00E+06	1.00E-18	633nm; power: 0.2mW; acq t: 1s	[66]
AgNP@ Si wafer	TNT	6.40E+06	1.00E-12	514 nm, acq t: 1s	[67]
AuNP@Si Immunoassay sandwich	hIgG		3.00E-12	633 nm and 785nm	[68]
AuNanothorn @Si wafer	R6G	1.9E+07	4.50E-13		[69]
AuNP@Si	R6G	2.5E+08	1.00E-11	633 nm, power: 0.94mW	[70]
AgNC@SiO ₂ @PMHS paper	adenine	6.55E+06	8.90E-10	633 nm, power: 0.3 mW, acq t: 2 s, stability up to 90 days	[71]

Table S2. SERS substrates using pure Si with/or SiO₂ as a substrate.

Substrate	Analyte	Enhancement Factor	LOD	Details (Laser, power; acquisition time, RSD and etc)	Ref
black Si	PATP-to-DMAB	1.00E+03	1.00E-06	532nm; para-aminothiophenol (PATP)-to-4,4'-dimercaptoazobenzene (DMAB) reaction was observed.	[72]
Si Nano Wires	R6G		1.00E-04	633nm; power: 8mW; acq t: 50s;	[25]
SiO ₂ Nano Wires	Interleukin 10	4.00E+07	5.55E-12	415.5nm; power: 24.6 mW; acq t: 10 s; LOD 0.1ng/ml ; EF of Au@SiO ₂ substrate is 2.7*10 ³	[73]
Laser irradiated Si structure	Malachite green	500		533 nm; acq t: 1s; spongiform Si structures of average dimension of 300 nm	[74]
Si Nano Spheroids	R6G	5.38E+06		785 nm; acq t: 3s; substrate is composed of both crystalline and amorphous Si and amorphous SiO ₂ .	[75]
Si@SiO ₂	HeLa cancer	1.00E+07	1.00E-12	785 nm; acq t: 180s/line; in situ live bioimaging	[76]

Table S3. Platinum and/or Palladium based SERS substrates.

Substrate	Analyte	Enhancement Factor	LOD	Details (Laser, power; acquisition time, RSD and etc)	Ref
Anodic Al oxide@ Pt Metal Film	Methylene blue	1.88E+05	1.00E-09	532nm; power: 0.01mW; acq t: 1s; RSD 7.14% for 5 signals	[77]
PtNP@AgNP@ Ge wafers	R6G	9.10E+06		633 nm; acq t: 1s; RSD <10% for 100 spots; Pt w% is 8.57%	[78]
PtNP	pyridine	2000		632.8 nm; size of PtNP 10-20nm	[79]
PtNP (core @ SiO ₂ (shell))	Pyridine	1.00E+05		632.8 nm; EF obtained for electrochemically roughened platinum electrodes is about 100.	[28]
AgNP@4-ABT@Pt	4-ABT	790		514.5nm; acq t: 30s; Pt surface 400 nm × 400 nm × 150 nm.	[80]
Pt@TiO ₂ Nanotube Arrays	R6G	4.30E+04	2.00E-05	532 nm; power: 0.5mW; acq t: 5s; polydopamine layer is on vertically aligned TiO ₂ nanotube.	[81]
AgNanoCubes @ pATP@Pt (sandwich)	p-ATP	4.10E+06		532 nm; no signal w/o AgNC; signal increases as the thickness of Pt film from 42 to 90nm	[82]
PdNP @ AuSSV (sphere segment void)	pyridine	7.80E+03		632.8nm; power: 3mW; EF decreases with the thickness of Pd layer	[83]
PtNP @ AuSSV (sphere segment void)	pyridine	4.90E+03		632.8nm; power: 3mW; EF decreases with the thickness of Pt layer	[83]
PdNP @ porous Si	R6G	100		488 nm; power: 5mW; acq t: 30s; size of PdNP 40-86 nm	[29]
PtNP @ porous Si	R6G	33		488 nm; power: 5mW; acq t: 30s; size of PtNP 56-68 nm	[29]
PtNA@Si wafers	R6G	5.00E+04		515nm; power: 0.17 mW; acq t: 30 s	[30]
AuNP shell @ PdNP core	p-ATP	5.00E+04		632.8nm; power: 1mW; 135 nm Au covering 0.7 nm Pd (about two atomic layer of Pd)	[84]
PdNP@porous Si	TNT	2.54E+06	1.00E-07	514 nm; power: 10mW; acq t: 60s; w%: Si is 60%; Pd is 37%	[31]

Ag&PdNP@Gradient porosity Si	E.Coli	2.30E+05	1 CfU/ml	785 nm; acq t: 10s; NP is 10nm; optimum 180s immersion time	[32]
Au&Pd alloy@Cu ₂ O/Cu O	CV	5.00E+05	1.00E-06	488 nm; acq t: 10s; size:160nm; EF increases with the thickness of alloy; 100% self-recovery in 1 min	[85]
PdNP@Si	R6G	8.54E+05	1.00E-10	682.8nm; power: 0.1mW; acq t: 10s; Pd thickness is 300nm	[33]
PdNP@ pyramidal Si	R6G	1.91E+06	1.00E-11	682.8nm; power: 0.1mW; acq t: 10s; Pd thickness is 300nm	[33]
PdNP@ Ge	R6G	3.48E+05	1.00E-09	682.8nm; power: 0.1mW; acq t: 10s; Pd thickness is 64.4 nm	[33]
Polyvinyl alcohol scaffold@PdNW	MB (methylene blue)	1.29E+02	1.00E-05	532 nm; Diameter: 55–70 nm, length; 3–4 μm	[86]
CTAB@Pd nanocubes	MB (methylene blue)	1.93E+03	1.00E-06	532nm; side length: 37 ± 5 nm	[86]
DNA@PdNP	MB (methylene blue)	1.20E+05	1.00E-08	532 nm; size of the PdNPs is 3.5- 5 nm.	[86]
AuPdNanoBrambles	Crystal Violet	5.37E+06	8.00E-08	633 nm; power: 0.5mW: acq t: 10s; w% of Au is 46%, Pd is 54%	[87]
PdAu Hollow NanoChains	R6G	3.50E+05	1.00E-08	633nm; power: 1.7mW; acq t: 10s; w% of Pd is 4 times more Au	[88]
PdNP@Au	4-ABT	4.80E+03		632.8nm; power: 10mW; acq t: 10s; triangular 100nm Pd with height of 300-400nm	[89]
PdNP	4-ABT	1.90E+09	1.00E-10	633 nm; power: 10mW; acq t: 5s;	[90]
Nanocrystalline Pd films@Si	thiophenol	1.20E+05		632.8 nm; power: 5mW; acq t: 10s; NP size 50-60nm	[91]
Ag/Pd alloy NP	R6G	2.62E+08	1.00E-09	532 nm; power: 30mW; w% of Pd is 18%; SERS activity dec with inc in w% of Pd; stable for 20 days.	[92]

PdNP	4-mercaptopyridine;	1.70E+05		514nm; power: <0.5mW; acq t: 10s;	[93]
Flake PdNP@Au surface	4-ABT	1.50E+03		632.8nm; power: 8.5mW;	[94]
PdNP	pyridine	1.80E+03		632.8nm; power: 7mW; size 50nm	[95]
AuNP@FePt@SiO ₂	adenine	2.00E+07		633 nm; substrate diameter 30nm	[34]
Ag/Au/Pt Nanocages	Rhodamine 3B	4.70E+09	1.00E-15	532 nm; acq t: 10s; w% Ag: 63%, Au: 16%; Pt 21%,	[96]

Table S4. Performance of Gold and Silver as SERS substrates.

Substrate	Analyte	Enhancement Factor	LOD	Details (Laser; power; acquisition time; RSD and etc)	Ref
Ag	4-Mercaptobenzoic acid	7.33E+07	1.00E-10	785 nm,	[97]
Ag Nano Sheets@opal	R6G	6.00E+07	1.00E-14	514 nm, power: 0.17mW; Ag nanosheets are 370nm	[98]
Ag Nano Cubes	R6G	2.00E+08	1 molecule detection	532 nm, power: 60mW; acq t: 5s	[99]
AuNP	4-Mercaptobenzoic acid	1.10E+09	1-2 molecule detection	785 nm; AuNP with hollows 20 to 45 nm,	[100]
AuAg alloy nanoporous film	R6G	3.00E+08	1.00E-12	785 nm; power: 0.1mW; EF of hotspots: 10E+10 to 10E+11, w% of alloy Au 79%, Ag 21%	[101]
Au Nano Flowers	MUC 4 (pancreatic cancer marker)	7.30E+06	1.00E-13	785 nm; LOD was 0.1ng/ml; MUC 4 MW was taken to be 1000kDA	[102]
Ag/Au Nano Cubes	R6G	6.10E+09	1.00E-16	532 nm; acq t: 10s;	[103]
AuNP	hIgG		2.80E-11	633 nm; power: 5mW; acq t: 5s; Immunoassay sandwich; antigen used; AuNP 60nm; were modified with Raman reporters (4-NBT)	[68]
AuNP@cysteine	TNT	1.0E+09	2.00E-12	670nm, power: 2mW	[104]
Ag nano ribbons	TNT	1.0E+07	2.50E-08	532 nm, 1ps laser and input energy of 1200mJ was used to synthesize Ag nano ribbons.	[105]

Au nanofilms/cicada wing	R6G	2.2E+06	1.00E-08		[106]
Ag pyramids	R6G	9.1E+05	1.00E-10	532 nm	[107]
Ag&AuNP	R6G	5.0E+08	2.00E-14	785 nm, power: 1mW	[108]
AuNP	R6G	5.8E+07	2.00E-12		[109]
AuNP	R6G		3.00E-09	638 nm, acq r: 1s	[110]
AuNP@polyster ene	R6G	1.8E+08		473 nm, size of Au 5nM	[111]
AgNP@cicada wings	R6G	1.0E+06	1.00E-07	785 nm, power: 0.325mW, acq t: 1s	[112]
AgNP@filter paper	adenine	1.00E+07	1.60E-07	785 nm	[113]
Ag@chitosan flakes	adenine		1.20E-11	633 nm	[114]

Table S5. The average SERS performance of Si, Pt, Pd based substrates and pure Ag, Au substrates in detection of R6G (analyte).

Substrate	Average EF		Average LOD (M)		References
	Arithmetic	Geometric	Arithmetic	Geometric	
Pure Si	5.38×10^6 (only one EF)		10^{-4} (only one LOD)		[25,74]
Si with metals	2.0×10^{12} (33; 6×10^{13})	1.8×10^7 (33; 6×10^{13})	5.0×10^{-9} (10^{-16} ; 10^{-7})	2.2×10^{-11} (10^{-16} ; 10^{-7})	[1,5,7,8,12-18,22,23,25,29,33,35,36,38,39,50,52-54,57,61,62,69,70]
Pt	2.3×10^6 (33; 9.1×10^6)	2.8×10^4 (33; 9.1×10^6)	2.0×10^{-5} (Only one LOD available)		[29,30,78,81]
Pd	4.4×10^7 (100; 2.6×10^8)	4.6×10^5 (100; 2.6×10^8)	2.4×10^{-9} (10^{-11} ; 10^{-8})	3.2×10^{-10} (10^{-11} ; 10^{-8})	[29,33,88,92]
Al+Au	8.1×10^6		7×10^{-10} (10^{-10} ; 10^{-9})	3.16×10^{-10} (10^{-10} ; 10^{-9})	[115]; [116]; [117]; [118]
Al+Ag	5.39×10^7 (10^7 ; 9.77×10^7)	3.13×10^7 (10^7 ; 9.77×10^7)	2.50×10^{-7} (10^{-15} ; 10^{-6})	1.78×10^{-11} (10^{-15} ; 10^{-6})	[119-123]
Au	8.0×10^7 (2×10^6 , 2×10^8)	2.8×10^7 (6×10^7 , 2×10^8)	4.3×10^{-9} (2×10^{-12} ; 10^{-8})	3.9×10^{-10} (2×10^{-12} ; 10^{-8})	[106,109-111]
Ag	6.6×10^7 (9×10^5 ; 2×10^8)	1.0×10^7 (9×10^5 ; 2×10^8)	3.34×10^{-8} (10^{-14} ; 10^{-7})	4.6×10^{-11} (10^{-14} ; 10^{-7})	[98,99,107,112]

Table S6. The Clinical Performance of Gold and Silver SERS substrates.

Substrate	Diagnosis of	Analyte, biomarker	Sensitivity	Specificity	Accuracy	N of sample (positie+, negative -)	Ref
AuNP	7 different cancer cells	cells			81.2%	245	[124]
AuNP	Cervical cancer	Serum	100%	100%	100%	36 (24+,12-)	[125]
AuNP	Oral squamous cell carcinoma	Serum	80.7%	84.1%	82.5%	280 (135+, 145-)	[126]
AuNP	Colorectal cancer	Serum	97.4%	100%	98.8%	83 (38+,45-)	[127]
Au chip	Lung Cancer	Saliva	100%	100%	100%	127 (61+,66-)	[128]
AuNP	Prostate cancer	Urine	100%	89%	95%	18 (9+,9-)	[129]
AuNP	Colorectal Cancer	Serum,carcino embryonic antigen	90%	100%	91.8%	98 (80+,18-)	[130]
AgNP	Colorectal Cancer	Purified serum, Albumin	100%	100%	100%	206 (103+, 103-)	[131]
AgNP	Liver cancer	Serum	92.3%	100%	97.3%	75 (26+, 49-)	[132]
	Nasopharyngeal cancer	Serum	96.0%	88.0%	90.7%	75 (25+, 50-)	[132]
AgNP	Coronary heart disease	Urine	80.9%	92.1%	84.1%	220 (157+, 63-)	[133]
AgNP	Prostate Cancer	Serum, PSA	95%	93.8%	94.2%	120 (80+, 40-)	[134]
AgNP	Breast Cancer	Saliva, sialic acid	94%	98%	96.1%	206 (100+, 106-)	[135]
AgNP (SERS-CRISPR)	SARS-COV-2	RNA extracts, viral N gene	87.5%	100%	97.3%	112 (24+, 88-)	[136]
AgNP	Prostate Cancer	Serum & PSA	100%	87.5%	94%	54 (30+,24-)	[137]

Overall performance of the methods shown below are used in the review to compare the efficiency of Au, Ag substrates in clinical diagnosis in comparison to non-conventional SERS substrates as Si, Fe, Al and etc.

Table S7. Multi Elemental Aluminum-based SERS substrates. Abbreviations: AAO - anodic aluminium oxide.

	Ref	Author et. al	Year	Analyte	Substrate	Analytical parameters
Al+Ag	[138]	Chang et al.	2020	R6G	ZnO: Al/Ag heterostructure	LOD 10^{-10} M
	[119]	Chang et al.	2019	R6G	Al-doped ZnO@SnO ₂ @Ag heteronanowires	LOD 10^{-12} M Substrate can maintain similar SERS-enhancing effect even after five cycles.
	[120]	Shan et al.	2014	R6G	AAO/Al-based Ag nanostructure arrays	EF 9.77×10^7
	[122]	Das et al.	2021	R6G	silver capped aluminium nanorods	LOD 10^{-15} M EF 10^7
	[123]	Zhang et al.	2018	R6G	AgNPs/MnO ₂ @Al	LOD 10^{-6} M
	Geometric mean for LOD: 1.78×10^{-11} M					
Geometric mean for EF: 3.13×10^7						
Al+Au	[115]	Hou et al.	2014	R6G	Au nanoparticles on hexagonally patterned bowl-shaped-dimples on Al foil	LOD 10^{-9} M
	[116]	Sui et al.	2016	R6G	Au-CuCl ₂ -AAO	LOD 10^{-10} M EF 2.3×10^7
	[117]	Nielsen et al.	2009	R6G	Gold Nanostructures with Sub-10 nm Gaps on a Porous Al ₂ O ₃ Template	LOD 10^{-9} M
	[118]	Choi et al.	2010	R6G	Au-AAO	EF 0.81×10^7
	Geometric mean for LOD: 3.16×10^{-10} M					
Geometric mean for EF: 8.10×10^6						

Table S8. Multi Elemental Copper-based SERS substrates. Abbreviations: CV- crystal violet, R6G – rhodamine 6G, MB – methylene blue.

	Ref	Author et al.	Year	Substrate morphology	Analytes	Analytical parameters
Cu+Ag	[139]	Fodjo et al.	2012	Ag/b-AgVO ₃ nanobelts deposited on copper foil (Cu@Ag/b-AgVO ₃)	carbaryl carbofuran isoprocarb propoxur (carbamate pesticides)	LOD: 2.5×10 ⁻¹² M 10×10 ⁻¹² M 50×10 ⁻¹² M 75×10 ⁻¹² M EF 10 ⁶ RSD 4.3%
	[140]	Dai et al.	2021	Cu-coated fabric, Cu-Ag-coated fabric	CV	LOD 10 ⁻⁸ M EF 2×10 ⁶
	[141]	Liu et al.	2020	Complex of CuO@Ag nanowires on Cu mesh (Cu/CuO@Ag)	R6G	LOD 10 ⁻¹⁵ M EF 4.88×10 ¹¹
	[142]	Rao et al.	2017	Cu–Ag–PVA (with different Ag contents)	R6G MB	LOD: 3.30×10 ⁻⁸ R6G, 1.60×10 ⁻⁸ MB EF 10 ⁷ -10 ⁸
	[143]	Zhang et al.	2018	graphene/bilayer silver/Cu sandwich	CV, R6G	LOD: 10 ⁻⁹ CV, 10 ⁻⁸ R6G EF 1.19×10 ⁵ RSD <5.9% R ² 0.935 for CV R ² 0.974 for R6G
	[144]	Sravani et al.	2021	Ag–Cu alloy microflowers	R6G	LOD 10 ⁻²¹ M EF 10 ⁸
	[145]	Fu et al.	2020	Superhydrophobic nanostructured Cu with Ag NPs	R6G	LOD 10 ⁻¹³ M EF 1.2×10 ⁵
	Geometric LOD: 5.30×10 ⁻¹² M Geometric EF: 1.76×10 ⁷					
Cu+Au	[146]	Bankowska et al.	2016	Au–Cu layer on photoetched GaN	pyridine, 4-mercaptopbenzoic acid	EF 5×10 ⁵
	[147]	Sakir et al.	2020	Cu NSs from Au nanoparticles (NPs) that were attached to the substrate through a layer of end-grafted	R6G	LOD 10 ⁻⁸ M EF 2.95×10 ⁵ RSD 9.4% R ² =0.973

				poly(ethylene glycol).		
[148]	Chen et al.	2017	Au/Cu hybrid nanostructure arrays	Urea	LOD 10^{-3} M	
[149]	Manish et al.	2020	Au–Cu alloy nanostructures	MB	EF 1.2×10^3	
[150]	Siva et al.	2020	Flower-Shaped Au–Cu Nanostructures	CV	LOD 10^{-10} M EF 0.21×10^6	
Geometric LOD: 1.00×10^{-7} M Geometric EF: 7.81×10^4						

Table S9. The list of SERS studies of different Al-based substrates from 2012 to 2020. Abbreviations: NAP – naphthalene, TEPS – triethoxyphenylsilane, CV – crystal violet, R6G- Rhodamine 6G, $(\text{Ru}(\text{bpy})_3)^{2+}$ - tris(bipyridine)ruthenium(II), MH - 6-mercaptop-1-hexanol, BPE -trans-1,2- bis(4-pyridyl)-ethylene, AlFON - aluminum film-over nanosphere.

Substrate	Substrate preparation	Analytes	Analytical parameters	Ref
Al NP-film	Thickness of Al nanosheets from which Al NPs were produced – 2 nm. Al nanosheets deposited on Al film at a rate 8 Å / s under 10^{-6} Pa pressure	Adenine, CV (325 nm)	For adenine: EF 3.62×10^5 , LOD 10^{-6} M For CV: EF 4.1×10^5 , LOD 10^{-7}	[151]
Al nanocrystals	SERS and normal Raman spectra were measured under 0.0038, 0.038, 3.8 mW laser power and 10, 40, 10 s integration time. The SERS spectra on Al substrate were averaged over 50 scans.	ssDNA (785 nm)	EF 10^5 - 10^6 LOD 2×10^{-6} M	[152]
Al nanodots	Polymeric nanostructures were fabricated using the Nanoscribe® Photonic Professional (Nanoscribe Inc., Germany). All structures were written on square glass substrates (width = 22 mm and thickness \approx 0.13 to 0.16 mm).	NAP, TEPS (532 nm)	EF 7×10^4 EF 1.7×10^4	[153]
Aluminum film-over nanosphere (AlFON)	nanosphere sizes: 170 nm silica spheres (Bangs Laboratories, SS02N, washed once and diluted prior to use) and 210 nm carboxylated latex/polystyrene spheres (Life Technologies, C37486, used as received).	Adenine (229 nm), $(\text{Ru}(\text{bpy})_3)^{2+}$ MH (355 nm), BPE (405 nm)	EF 10^3 - 10^5	[154]
Al nanovoids	Large areas of Al nanovoids were fabricated via nanoimprint lithography. A layer of 50 nm Al was evaporated onto the nanovoids via E-beam evaporation (Lesker) to form the film of Al nanovoids.	Adenine (244 nm)	EF 5×10^3	[155]

Al nanovoids	optical power density on the sample was 1500 W/cm ² . The exposure time was 30 s, and 10 acquisitions were performed for each spectrum.	Adenine (488 nm, 785 nm)	EF 10 ⁶	[156]
Al bow-tie nanoantenna	50 nm thick aluminum bowtie antenna was formed by two equilateral triangles of 100 nm side length with 20 nm apex to apex gap distance.	Liquid benzene (258.8 nm)	EF ~10 ⁵	[157]
Geometric mean for LOD: 5.85×10 ⁻⁷ M, geometric mean for EF: 1.27×10 ⁵				

Table S10. The clinical performance of the SERS studies with different substrates.

Substrate	Diagnosis of	Analyte	Sensitivity	Specificity	Accuracy	N of samples	Ref
SiO ₂ (core)@Ag(shell) (immunoassay)	SARS-COV-2	Serum,IgM/IgG	100%	100%	100%	68 (19+, 49-)	[158]
SiC@Ag(film)@AgNPs (immunoassay)	Prostate cancer	Serum, (biomarkers : PSA, PSMA, hK2)	70%	-	70%	10+	[63]
	benign prostate hyperplasia	Serum, PSA, PSMA, hK2	60%	-	60%	10+	[63]
	Healthy samples	Serum, PSA, PSMA, hK2	-	75%	75%	12-	[63]
AgNP@Si	N. gonorrhoeae	Urine, N. gonorrhoeae	100%	100%	100%	120 spectra (60+, 60-)	[64]
Au@ZnO@Si	Neisseria meningitidis	Cerebrospinal fluid, neoprotein	95%	98%	-	N/A	[65]
AgNP@Si wafer	Breast cancer	Serum, 9 miRNAs	-	-	85%	25, (7+, 6+, 5+, 4+, 3-)	[66]
Si@SiO ₂	HeLa cancer	cells	86%	94%	-	N/A	[76]
AgNP@pyramidal Si	Lung Cancer	serum	100%	90%	95%	100 (50+, 50-)	[159]
AgNP @ porous Si	Breast Cancer	Serum	93.3%	96.7%	95%	60 (30+, 30-)	[160]
Si wafers @ Ag @ AuNP	Hepatocellular carcinoma	Serum	100%	100%	100%	80 (20+, 60-)	[161]
SiO ₂ @ AuNP	Lung Cancer	Serum	-	-	90.7%	116 (58+, 58-)	[162]

2D Au–Si substrate and upper Ag@4-MBA@Au core-shell nanoparticles.	Breast Cancer	miRNA-21 and miRNA-155 in serum	100%	100%	100%	60 (30+, 30-)	[163]
Ag coated Si Nanopillar	Lung Cancer	-	87%	83%	85%	N/A	[164]
Au cubes on Si	SARS-CoV-2 virus	Saliva	95%	95%	95.2%	42 (21+, 21-)	[165]
Ag @ Si	Neoplastic Tissue	Salivary gland homogenates	97%	89%	93%	6 (3+, 3-) 120 spectra	[166]
Al foil	Colorectal Cancer	Serum	83%	83%	83.3%	60 (30+,3 0-)	[167]
AgNP@Al foil	Prostate Cancer	Serum, PSA	87%	100%	94%	54 (30+,2 4-)	[137]
AgNP@Al plate	Prostate Cancer	Serum, PSA	84.9%	100%	91.3%	161 (93+,6 8-)	[168]
Ag Colloid@Al foil	bladder cancer, NMIBC, MIBC	Serum	-	-	93.3%	149 (27+,3 2+, 60+,30 -)	[169]
AgNR wrapped with Al ₂ O ₃	lung adenocarcinoma (LAC)	Serum	98.1%	97.6%	-	190 (108+, 82-)	[170]
AgNP@Al plate	bladder cancer	Serum	-	-	96.4%	88 (48+,4 0-)	[171]
	differentiating bladder cancer	Serum	-	-	95.4%	88 (25+,2 3+, 40-)	[171]
AgNP@Al plate	Bladder Cancer	Serum	90.9%	100%	94.5%	91 (55+,3 6-)	[172]
Ag colloid@Al foil	Prostate cancer, benign	Plasma, PSA	95.8%	100%	97.9%	28 (14+,1 4-)	[173]

	prostatic hyperplasia						
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Note: The sensitivity/specificity/accuracy of the overall substrate group is presented in the following way: average (min; max). The number of samples: All samples (positive “+”, negative “-”) NP – nanoparticles, PSA - Prostate Specific Antigen. Ref 124 is included in the table, but the parameters reported in this paper are not used for the calculation of averages, since the number of patients tested in the study (10 to 12) is too low to consider the reported parameters as conclusive statistical values. Abbreviations: non-muscle-invasive bladder cancer (NMIBC) and muscle-invasive bladder cancer (MIBC).

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