

**Supplementary information for:**  
**Artificial intelligence and machine learning for plasmonic and surface-enhanced sensing**

Ailsa Geddis<sup>1</sup>, Hannah Williams<sup>1</sup>, Saba Bashir<sup>1</sup>, Jason Malenfant<sup>1</sup>, Caroline Dubois<sup>1</sup>, Louis Hamlet<sup>1</sup> and Jean-François Masson\*<sup>1</sup>

**AUTHOR AFFILIATIONS:**

<sup>1</sup>Département de chimie, Institut Courtois, Centre interdisciplinaire de recherche sur le cerveau et l'apprentissage, Quebec Center for Advanced Materials, Regroupement québécois sur les matériaux de pointe, Université de Montréal, C.P. 6128 Succ. Centre-ville, Montréal, Québec, Canada, H3C 3J7.

\*Corresponding author: [jf.masson@umontreal.ca](mailto:jf.masson@umontreal.ca)

**ORCID:**

Ailsa Geddis : 0000-0002-0161-1968

Saba Bashir : 0000-0001-9595-7048

Jason Malenfant : 0009-0002-2694-1872

Caroline Dubois : 0009-0003-8195-4438

Jean-Francois Masson : 0000-0002-0101-0468

**CONTENTS:**

S1: Summary table for protocols for tuning colloidal silver nanoparticles

S2: Reference Table for Clinical and Diseases Plasmonic Sensing with ML and AI

S3: Reference Table for Environmental Plasmonic Sensing with ML and AI

S4: Reference Table for Food Plasmonic Sensing with ML and AI

S5: Reference Table for Materials Characterisation

S6: Reference list for S1

NB reference numbers in S2-5 correspond to main article.

## S1: Summary of Protocols for Tuning Colloidal Silver Nanoparticles

Shape/LSPR	Method of Synthesis	Methodology impact on Morphology	LSPR peak: shape shift
<b>Sphere, quasi-spherical</b> 400-520	<sup>1</sup> AgNO <sub>3</sub> reduction with TSC <sup>2</sup> AgNO <sub>3</sub> reduction with NaBH <sub>4</sub>	<b>Strength of reducing agent:</b> Strong (NaBH <sub>4</sub> ) : ↓ NP diameter Weaker (TSC) : ↑ NP diameter <b>[Reducing agent]:</b> High : ↓ NP diameter Lower : ↑ NP diameter	Blue: narrow Red: broad Blue: narrow Red: broad
	<sup>3, 4</sup> Photoassisted: reduction of AgNO <sub>3</sub> with citrate or NaBH <sub>4</sub> under UVA/blue light	$\lambda_{\text{radiation}}$ : 512-480 nm: disc/triangles 477-458 nm: pyramid/pentagon	Red Blue
<b>Cubes</b> 430 - 700	<sup>5</sup> AgNO <sub>3</sub> reduction with ethylene glycol and PVP	<b>[AgNO<sub>3</sub>]:</b> High : single crystal nanocubes Low : mix truncated cube, spheres, prisms <b>Time of reaction (at low [AgNO<sub>3</sub>]):</b> ↓: ↑ polydispersity	
	<sup>6</sup> AgNO <sub>3</sub> reduced with glucose and shape controlled with HTAB	<b>HTAB:[Ag(NH<sub>3</sub>)<sub>2</sub>]:</b> ↓ ratio (1-1.5): spherical particles ↑ ratio (2.5-3): sharp nanocubes	Blue: broad Red: narrow
	<sup>7</sup> AgNO <sub>3</sub> reduced with ethylene glycol and selective etching with H <sub>2</sub> S and shape controlled with PVP	<b>[H<sub>2</sub>S] Concentration for selective etching:</b> Optimal : monodisperse nanocubes ↓ H <sub>2</sub> S : nanocubes rounded sides ↑ H <sub>2</sub> S : wires, and agglomerated particles	Red: narrow Blue: Very broadened
<b>Prism</b> 340-440 and 650-700	<sup>8</sup> Silver di(2-ethyl-hexyl)sulfosuccinate (Ag(AOT)), N <sub>2</sub> H <sub>4</sub> , dodecanethiol (stabilizer)	<b>[N<sub>2</sub>H<sub>4</sub>]/[Ag(AOT)] = 2 Ratio:</b> Higher Ration : ↑ size <b>Dodecanethiol addition:</b> ↑ : ↑ hexagon size	Red Red/blue: broad
	<sup>9</sup> AgNO <sub>3</sub> reduced with TSC, NaBH <sub>4</sub> and NaOH and photo irradiated	<b>[OH<sup>-</sup>] ( release of Ag<sup>+</sup> is produced from OH<sup>-</sup>):</b> Low/moderate (slow release of Ag <sup>+</sup> ): well-defined, thin triangular prism High OH <sup>-</sup> (Ag <sup>+</sup> production nearly stops): small/ polydisperse	Red Blue: broad
	AgNO <sub>3</sub> reduced by NaBH <sub>4</sub> and TSC	<b>[NaBH<sub>4</sub>] :</b> ↓ [NaBH <sub>4</sub> ] : spherical and rodlike NPs Med [NaBH <sub>4</sub> ] : Triangular nanoprism and few spheres ↑ [NaBH <sub>4</sub> ] : small spherical AgNP	Broad Red, broad(ish) Blue, narrow

<b>Rod</b> ~400 and ~500-620	<sup>10</sup> Seeded: AgNO <sub>3</sub> , reduced by L-ascorbic acid (AA) in the presence of Au <sub>seeds</sub> , CTAB and NaOH	<b>[Au<sub>seed</sub>]:</b> ↓ [Au <sub>seed</sub> ]: ↑ nanorod aspect ratio (longer rods) <b>pH:</b> pH > pK <sub>a2</sub> of AA: nanorods pH < pK <sub>a2</sub> of AA: nanowires	Red (longitudinal band)  Longitudinal/transverse modes Transverse mode only
	<sup>11</sup> Polyol process: AgNO <sub>3</sub> reduced by ethylene glycol and PVP modified with tannic acid and CF <sub>3</sub> COOAg	<b>Temp:</b> 120 °C (EG ↓ reducing) : spheres 170 °C (optimum) : uniform Ag NRs 187 °C (too fast reduction): by-products and uncontrolled growth	Blue (no longitudinal band) Red (longitudinal band) Broad
	<sup>12</sup> AgNO <sub>3</sub> reduced by ethylene glycol and PVP	<b>Time of reaction (with AgNO<sub>3</sub> injection) :</b> ↓ time (5.0 min) : ↓ aspect ratio (shorter rods) ↑ time (6.5 min) : ↑ higher aspect ratio (longer rods)	Blue (longitudinal band) Red (longitudinal band)
<b>Branched</b> 450-700	<sup>13</sup> Au Seed mediated growth solution of AgNO <sub>3</sub> , BDAC and CTAB	<b>[BDAC]/[CTAB] :</b> ↑[BDAC]/[CTAB] (less protected end): ↑ aspect ratio (longer rods) ↓[BDAC]/[CTAB] (more protection): ↓ aspect ratio (shorter rods)	Red (longitudinal band) Blue (longitudinal band)
	<sup>14</sup> AgNO <sub>3</sub> reduced with NH <sub>2</sub> OH and TSC	<b>[HA]</b> (number of initial seeds): shorter branches <b>[TSC]</b> (faster growth along Au <sub>seeds</sub> ): longer branches <b>[AgNO<sub>3</sub>]:</b> ↓ [AgNO <sub>3</sub> ] : shorter branching ↑ [AgNO <sub>3</sub> ] : longer branching	Blue: narrow Red: broad  Blue: narrow Red: broad
	<sup>15</sup> AgNO <sub>3</sub> reduced with AA and PVP	<b>[AgNO<sub>3</sub>]:</b> ↑ [AgNO <sub>3</sub> ] (more nuclei): shorter branching ↓ [AgNO <sub>3</sub> ] (fewer nuclei): longer, more defined branching <b>[AA]:</b> ↑ [AA]: smaller branches ↓ [AA]: longer, sharper branches <b>[PVP]:</b> presence stabilizes sharp branches	Blue: narrow Red: broad  Blue: narrow Red: broadened Red: narrowing
<b>Plates</b> 450-900	<sup>16</sup> Ag <sub>seed</sub> Growth: Ag <sub>seed</sub> , AgNO <sub>3</sub> + PVP, NaBH <sub>4</sub>  The controlled oxidation with H <sub>2</sub> O <sub>2</sub> and enables oriented dissolution-regrowth	<b>Rate of H<sub>2</sub>O<sub>2</sub> addition (etchant):</b> Slow : well-defined nanoplates Fast : poor control or loss of platelet morphology  <b>[PVP] :</b> ↓ [PVP] : thicker and/or irregular shapes  <b>[NaBH<sub>4</sub>]</b> (determine initial Ag <sub>seed</sub> size): ↑ [NaBH <sub>4</sub> ] (smaller seeds) : more uniform thinner plates	Red: broad Blue: narrow  Broader  Narrow

		↓ [NaBH <sub>4</sub> ] (fewer seeds): thicker, plate	Blue: Broader
<b>Decahedra</b> 400-500	<sup>17</sup> Photochemical: Ag <sub>seed</sub> mediated from <sup>4</sup> then photochemical irradiated	<b>Time of reaction time (with Ag<sub>seeds</sub>):</b> ↑ (20-90 min) : seeds grow into fully formed decahedra <b>Reaction Temperature:</b> ↓ (~20 °C) : mainly decahedra, some plates ↑ (~80-85 °C) : ↑ polydispersity (polyhedrons, nanoplates)	Red shift  Red, narrow Broadened
<b>SiO<sub>2</sub> Shell</b> 520-725	<sup>18</sup> Hydrolysis and condensation of TEOS on AgNP	<b>[TEOS] :</b> ↑ TEOS : ↑ shell thickness	Red shift (slight)

\*Abbreviations: **AA:** L-Ascorbic Acid, **APTES:** (3-Aminopropyl)triethoxysilane, **APTMS:** (3-Aminopropyl)trimethoxysilane, **BDAC:** benzyltrimethylhexadecylammonium chloride, **CTAB:** Cetyltrimethylammonium bromide, **CTAC:** cetyltrimethylammonium chloride, **HTAB:** n-hexadecyltrimethylammonium bromide, **PVP:** poly(vinylpyrrolidone), **TEOS:** tetraethyl orthosilicate, **TSC:** trisodium citrate, **TTAB:** tetradecyltrimethylammonium

## S2: Selected Reference Table for Clinical and Diseases Plasmonic Sensing with ML and AI.

Ref #	ML function	AI algorithm(s) used	Biological Function of AI	Plasmonic Technique	Type of substrate?	Biological Analyte type	Accuracy and LOD
256	Classification	CNN	Classification of bacteria and yeast samples	[non plasmonic] Raman	/	Bacterial isolate and cells	82% isolate classification and 97% antibiotic treatment accuracy
401	Feature engineering Classification regression quantification	t-SNE unsupervised for data visualisation and quality check RF (classifier) SVM (regressor and quantification)	Chemical Taxonomy and structural elucidation for untargeted identification of unidentified molecules	SERS	Ag Nanocube array	Dropcast and dried solution of analyte	Accuracy of classification 87-100% Quantification LOD uM
403	Prediction of structure from Spectra Inverse Prediction	4 parallel RF PLS regressor  XGBoost regressor	Two-way prediction of structures and spectra of untrained molecules	SERS	Ag Film substrate	Pure analyte solution dropcasted	>90% classification accuracy
404	Classification Interpretability	PCA dimensionality reduction SVM with an RBF kernel SHAP	Bacterial classification	[non plasmonic] Raman	/	/	94% classification accuracy
277	Classification Quantification Interpretability (Attention based)	CNN with CBAM  (*CBAM) and GradCAM	Bacterial classification and quantitative diagnosis of polymicrobial UTIs	SERS	Au@Ag core-shell nanoparticles	Processed (filtered and centrifuged) isolates from urine samples	95% classification accuracy
405	Image analysis and classification/ detection	MLP and CNN	Rapid testing for single pathogen detection, smartphone image analysis	LSPR	Sandwiched plasmonic biosensor AuFilm AuNP	Human serum samples	99% classification accuracy LOD: 0.019 IU mL <sup>-1</sup>
406	Feature engineering Classification/ qualitative detection	Wavelet and Fourier transform and spectral difference MLP and SVM t-SNE analysis	Rapid testing for single pathogen detection	LSPR	/	Covid-19 swab dataset	/
407	Classification	MBAA-CNN	Rapid testing	SERS	Paper-based SERS chip with colloidal AgNP	Bacterial isolates from blood samples	98.6% species 99.5% antibiotic resistance accuracy
408	Feature extraction Data transformation classification	Wavelet packet transform Gramian angular field CNN	Bacteria identification through two-dimensional SERS	SERS	Ag-based substrate	Dropcasted cultured samples	90.6% classification accuracy
410	Image classification Data augmentation	Spectral Transformer (based on ViT) with MLP layer NoiseMix	Fast identification of bacteria phenotypes -with data augmentation to improve training	[non plasmonic] Raman	/	Bacterial isolates	96% classification accuracy for 15 classes of bacteria
411	Classification and staging	CNN	Periodontal disease staging	SERS	Ag nanoparticles	Saliva samples	99.7% classification 99.4% staging
412	classification	PLS-DA with artificial NN	Disease staging	SERS	PMN film with ZIF shell and AgNP	Exhaled breath	> 89% classification accuracy
413	Classification	kNN and nn	Structural elucidation of biomarker (for multiple	SPRi	Ti, Au, SiO <sub>2</sub> PDFTS microwell array biochips	Whole serum	LOD < 7 ng/mL

			sclerosis)				
275	Full analysis model: pre-processing, explainable quantification, spectral data interpretation	Deep Component Spectral Filtering	SERS metabolomics Biomarker identification and trace level quantification in biological matrices (and more...)	SERS	AgNPs (Hydroxylamine and Citrate coated)  AuNStar@Ag	Serum samples	/
417	classification	SVM	Cell secretome classification	SERS	Paper Microfluidic biosensor with AgNPs	Suspended Cells	96% classification validation accuracy
418	classification	PCA with DNN	Stem cell quality validation and differentiation prediction	SERS	Commercial Ag substrates	Suspended Cells	82% accuracy
419	Live classification	CNN	Dynamic extracellular environment monitoring	SERS	AuNRaspberry coated fibres	Live cells in culture	/
420	Classification and quantification	DNN	Detecting structural states of protein aggregates with immunoassay	SEIRA	Microfluidic micro-flowcell with plasmonic metasurface	Proteins in aCSF solution	/
422	Classification and staging	SVM	Classification and staging of breast cancer	SERS	Plasmonic AgNP@Au@PS chip	Blood isolated EVs	99.6% classification accuracy, 85.2% staging accuracy
423	Classification	SVM	Coronary artery disease diagnosis and prognosis determination	SERS	Gold coated glass slides	Human plasma isolated small EVs	80.2% classification accuracy
424	Classification and interpretability	PCA with CNN + SVM SHAP and PDP	Early-stage lung cancer	SERS	AuNPs	Lung cancer cell derived EVs	>91% classification accuracy for all tests
425	Classification	CNN	Exosome cancer discrimination of 6 cancer types	SERS	AuNP aggregated array chip	Exosomes isolated from plasma	>90% sensitivity and specificity
426	Classification and interpretability	CNN-MLP	Exosome cancer discrimination and biomarker profiling from interpretability	SERS	Gold nanoisland on glass substrate	Exosomes from serum	>93% accuracy
273	Interpretable classification	Grad-CAM	Spectral pattern recognition for lung cancer detection from exosomes	SERS	Gold Nanocube superlattices prepared by Marangoni effect-driven self-assembly	Exosomes isolated from cell culture	>98.95% accuracy from surface and internal biomarkers and >96.35% from solely surface
432	Image acquisition, reconstruction and segmentation through feature representation	Cycle-GAN  CNN (Vector Quantized Variational Autoencoder: VGVAE2)	For Stimulated Raman Histology (SRH) virtual staining  For feature expression, image reconstruction and segmentation for fast brain tumour detection	[non plasmonic] SRS	/	Surgical brain tissue sample	94.6% diagnostic accuracy reached (comparable to traditional histological images with 93.9% diagnostic accuracy)
433	Feature extraction by image segmentation	DenseNet based learning (DeepChem)	Image segmentation to obtain chemically labelled image from a single frame SRS image with subcellular resolution	[non plasmonic] SRS	/	Cultured cells	/
434	Decomposing and quantitative analysis then classification	MCR-ALS  with CNN (VGG-16 architecture)	Detecting arterial ageing remodelling from Raman Imaging for monitoring tissue ageing with collagen composition	[non plasmonic] Raman Imaging	/	Fixed Tissues	/
437	Classification with prediction	PLSDA, Hybrid ML-DL (VGG-16, DenseNet121, Xception, InceptionV3), Modified 2DCNN (above	Diagnosis and prognosis prediction of early-stage breast cancer with histopathology	[non plasmonic] FTIR Imaging	/	Fixed Breast Tissue Samples	/

		ML-DL + ALEXNET, LENET5)					
438	Rapid classification	CNN	pH responsive SERS chip for intraoperative glioma detection	SERS	AuNStar array with Raman reporter IR7p	H <sub>2</sub> O droplet exposed to live tissue then SERS chip	/
439	Rapid classification	SVM	Live intraoperative cancer detection	[non plasmonic] Raman	/	<i>In situ</i> intraoperative brain tissue	91% diagnostic accuracy for Glioblastoma, 97% brain metastases, 96% meningiomas
442	Data extraction augmentation Classification and regression	NN, (add realistic residuals) XGBoost	Extracting true spectra of viruses and augmenting data to improve classification and quantification (through regression)	SERS	AgNRod@SiO <sub>2</sub> array substrate	Saliva samples (some inoculated with virus)	92.3% classification and concentration prediction accuracy
274	Interpretable classification	CNN, ViT, CRIME	Quantification of serotonin in urine	SERS	Gold nanoparticles with cucurbitu[8]uril spacers	Urine and water samples	/

**Abbreviations.** RF: Random Forest; PLS: Partial Least Squares; XGBoost: eXtreme Gradient Boosting; SVM: support vector matrix; RBF: radial basis function; SHAP: Shapley Additive Explanations; CBAM: convolutional block attention module; MLP: multi-layer perceptron; CNN: convolutional neural network; t-SNE: T-distributed stochastic neighbour embedding; PMN: plasmonic metal organic framework nanoparticle film; PDFTS: Perfluorodecyltrichlorosilane; DNN: Deep Neural Network; PDP: Partial dependence plot; VGG-16: Visual Geometry Group ; MCR-ALS: Multivariate Curve Resolution Alternating Least Squares; SRS: Stimulated Raman Scattering; CRIME: Context Representative, Interpretable Model Explanation; ViT: Vision Transformer.

### S3: Reference Table for Environmental Plasmonic Sensing with ML and AI

Ref #	ML function	AI algorithm(s) used	Environmental Function of AI	Plasmonic Technique	Substrate Type	Environmental Analyte type	Accuracy and/or LOD
445	Classification and quantification	ML, logistic regression	Microplastic detection and quantification across a map image	SERS	Gold nanopocket on paper (for simultaneous filtration and detection)	Microplastic-containing solution	2.5 µg/mL PS 5.3 µg/mL PE
446	Qualitative and quantitative detection of multiple contaminants	DNN	Combined water purification and organic pollutant sensing	SERS	AgNP-ZnONR-SNF	Organic pollutants in water	1 pg/mL with 92.3% detection accuracy
447	Classification, regression and fast detection	PLS-DA and SVM-R	Recognition and real time monitoring of hazardous chemicals	SPEDS (PDUS and SERS)	AuNPs, CuS-coated Au array	17 hazardous chemicals, incl. Thiourea, Hg <sup>2+</sup> in water	10 <sup>-9</sup> M with 98.2% identification accuracy and 98.6% quantification
448	Classification and regression	BPNN, LSSVM	Rapid detection of soil-available Nitrogen	SERS	AgNP	Nitrogen as NH <sub>4</sub> -N, NO <sub>3</sub> -N in soil	30 µg/mL NH <sub>4</sub> -N, 10 µg/mL NO <sub>3</sub> -N
449	Regression	PLS	Portable, low-cost extracellular enzymatic activity monitoring in soil	SERS	AuNP	L-3,4-dihydroxyphenylalanine for secondary detection of enzyme activity in soil	Non applicable
450	Data dimensionality reduction and prediction	CNN	Concentration prediction of Thiram in soil	SERS	AuNP superlattice	Thiram in soil	10 <sup>-8</sup> g/g
452	Classification	PCA, k-NN	Pesticide Detection	SERS	AgNP on paper, prepared with extract of <i>C. Libani</i>	Thiram, phosmet, myclobutanil, and abamectin in water	1 µM
454	Regression and multiplex classification	Multinomial logistic regression	Detection of hazardous molecules	SERS	Needle-Clustered Ag@Au	3 Bipyridine pesticides + mixtures dropcast and in ground apple matrix	0.006 ppm paraquat, 0.017 ppm diquat, 0.037 difenzoquat
457	Trace levels detection and classification	PCA+k-NN	Detection of Explosives (aliphatic and aromatic) at trace levels	SERS	AuNPs on CPU PGA	Explosives (TNT, TNB, TNP, RDX, PETN, HMX)	2-6 nM aromatics, 600-900 pM aliphatics
459	Classification and concentration prediction	SVM	Pesticide classification using biomimetic SERS substrate	SERS	AgNP on PDMS coated Seashell "biomimetic sensor"	Pesticides (Thiabendazole, Thiram) swabbed from vegetable	0.35 x10 <sup>10</sup> M from bok choy, 0.25 x10 <sup>10</sup> M Chinese cabbage, 0.17 x10 <sup>10</sup> M lettuce
464	Regression and feature extraction	SVM-R, KNN, MobileNet	On-site pesticide residue detection	SERS	AuNPs	Pesticides (chlormequat chloride, paraquat)	0.03 mg/L chlormequat, 0.003 mg/L paraquat
466	Regression and Feature Extraction	SqueezeNet	Analysis of agricultural chemical residue in rice	SERS	AuNRs Superhydrophobic platform	Pesticides (chlormequat chloride, acephate)	0.05 mg/L chlormequat, 0.3 mg/L acephate
468	Classification	PCA, 4 MLs	Trace antibiotic detection	SERS	AuNP on plant leaf	Antibiotics (Ceftriaxone, Cefitofur sodium)	0.88 nM Rhodamine6G, 94% classification accuracy
469	quantification	PCA-SVM	Rapid quantification of structurally similar	CF-SERS	Ag-Cl NPs	Triazole pesticides in tobacco samples	74 ppb triadimenol, 68 ppb triadimefon

			pesticides				
--	--	--	------------	--	--	--	--

**Abbreviations.** **PLS-DA:** Partial least-squares discriminant analysis; **BPNN:** Back-propagation neural network; **SVM (-R):** Support vector machine (regression); **LSSVM:** Least squares Support vector machine; **k-NN:** *k*-Nearest neighbor; **DT:** Decision tree; **SPEDS:** Surface-plasmon-enhanced dual spectroscopies; **CF-SERS:** Capillary Force-Enhanced, Surface-Enhanced Raman Spectroscopy; **P-DUS:** plasmon-mediated differential UV-Vis spectroscopy; **PS:** Polystyrene; **PE:** Polyethylene; **TNT:** Trinitrotoluene; **TNB:** Trinitrobenzenesulfonic acid; **TNP:** Trinitrophenol; **RDX:** Royal Demolition Explosive; **PETN:** Pentaerythritol tetranitrate; **HMX:** High Melting Explosive; **AgNP-ZnONR-SNF:** Ag nanoparticles decorated, ZnO nanorods coated silica nanofibers; **AuNP:** Gold Nanoparticles; **CPU PGA:** Central Processing Unit Pin Grid Array

## S4: Reference Table for Plasmonic Food Sensing with ML and AI

Ref #	ML function	AI Model	Food Sensing Function of AI	Plasmonic Technique	Substrate Type	Food Matrix and analyte	LOD and/or accuracy
269	Multilabel classification and regression	Transformer (SERSFormer 2.0)	Simultaneous detection of multiple pesticides in fresh produce	SERS	Au@Ag core-shell NPs	Pesticides (chlorpyrifos, phosmet, carbophenothion, etc.) in spinach and strawberries	Sub-ppb (ng/g) Accuracy classification 99.9%
485	Detection and regression	1D & 2D CNNs (regression)	Trace level detection and quantification of mycotoxin in Corn Oil	SERS	Au NRods	Mycotoxin (zearalenone) in corn oil	$6.81 \times 10^{-4}$ $\mu\text{g/mL}$ (1D); $7.24 \times 10^{-4}$ $\mu\text{g/mL}$ (2D)
486	Data Augmentation and Regression	CNN-LSTM	Rapid and label-free detection of a mycotoxin in peanut oil	SERS	Au@Ag nanoparticles	Mycotoxin Aflatoxin B1 in peanut oil	0.31 $\mu\text{g/kg}$ (AFB1)
487	Regression	1D-CNN regression model	Identification and quantitative analysis of malachite green in fish	SERS	Wrinkled Ag NPs@PDMS film	Malachite green (veterinary drug) in fish scales	$1 \times 10^{-6}$ M
488	Classification and quantification	Transformer (SERSFormer)	Analysis of Hazardous pesticides in spinach	SERS	Au@Ag core-shell NPs	Pesticides (coumaphos, oxamyl, carbophenothion, thiabendazole, phosmet) in spinach	0.5 ppm (lowest tested; range 0.5–10 ppm) 98.4% classification accuracy
489	Regression	1D-CNN regression	Rapid detection and quantitative analysis of thiram (pesticide) in fruit	SERS	Corrugated Ag NPs@PDMS thin film	Thiram in Tomatoes & blueberries (peel)	$10^{-7}$ M (~0.024 ppm)
490	Classification and regression from complex mixture of molecules	RamanFormer (Transformer model with self-attention)	Multiplex quantification	Raman	/	/	/
491	Classification and regression	pSCNN (Pseudo-Siamese CNN)	Classification and quantification of a binary mixture of trace antibiotics in human milk	Label-free SERS	AgNP colloids	Antibiotics (doxycycline, tetracycline) in Human milk	$10^{-9}$ – $10^{-10}$ M (ppb) 98.85% accuracy
492	Classification and regression Interpretability	CNN + NN-EN hybrid SHAP	Discrimination and ratio quantification of multiplexed antibiotics in aqueous solution	SERS	Ag nanoparticles (~40–45 nm)	Aqueous / food-related (antibiotic mixtures) ciprofloxacin, doxycycline, levofloxacin	$\sim 10^{-8}$ M (ppb level) 98.7% classification; ratio quantification
493	Multiplex classification and regression	2D-CNN with Fusion (PLSR, Deep Forest, CNN for training)	Quantitative determination of low-conc preservatives and heavy metals in mushrooms	SERS + Fluorescence	AuNPs	Preservative (potassium sorbate) & heavy metal (Pb) in Mushroom (Tricholoma matsutake)	2.35 mg/kg (sorbate); 9.72 $\mu\text{g/kg}$ (Pb)
494	Classification	1D-CNN binary classifiers	Multiplex detection of foodborne pathogens	SERS	3D nanostructure swab	Pathogens ( <i>E. coli</i> , <i>Salmonella</i> , <i>Staphylococcus</i> , <i>Listeria</i> , <i>Bacillus</i> , <i>Yersinia</i> in Utensils & foods (meat, tomato, lemon, egg, etc.)).	38.16 ng/ $\mu\text{L}$ ( <i>E. coli</i> O157:H7) ~100% accuracy for 6 pathogens
495	Classification	ADMIN-NAS-Unet	Detection of Foodborne pathogens	SERS	Portable Raman SERS (no nanostructure detailed)	22 foodborne pathogens (22 subspecies: <i>E. coli</i> , <i>Listeria</i> , <i>Staphylococcus</i> , <i>Salmonella</i> , <i>Shigella</i> )	92.8% accuracy
496	Pre-processing Feature extraction	CNN for background noise elimination ResNet-18 CNN	Rapid and sensitive detection of <i>E. coli</i> in food from fluorescence image analysis	Fluorescence plasmonic	QD-Aptamer conjugates in microfluidic device	( <i>E. coli</i> ) in Milk & chicken	2 CFU/mL >99% accuracy; 96.7–104% recovery

497	Classification and Regression	ANN	Quality control of milk; quantitative determination of components and qualitative adulteration detection	SEIRA	Citrate-capped Ag nanoislands on glass (SEIRA)	Macronutrients in Milk (cow, camel, goat, buffalo, infant formula)	Fat (1.5–7.5%), Casein (1–5%), Lactose (1.5–7.5%), Urea (100–500 mg/L)
499	Classification	1D-CNN	Rapid differentiation and authentication of agricultural products incl. adulteration analysis	SERS	AgC NPs, Ag <sub>7</sub> Au <sub>3</sub> NPs, AgA NPs	Flue-cured tobacco, green tea, purple rice	Accuracies up to 99.4% (individuals) and 94.8% (adulterated)
500	Regression	SSA-BP Neural Network (shallow NN, optimised)	Identification of Auramine O dye adulteration in spices	SERS	AgNPs	Auramine O (illegal dye adulterant) in Saffron, Curcuma (food spices); Dendrobium officinale (medicinal herb)	0.003 mg/mL (LOD), 0.009 mg/mL (LOQ)
501	Classification Quantification	CNN (optimised), SAE, RF, SVM (compared) PLS-DA	Detection and quantification of antioxidants in edible oils	SERS	AuNP colloids	Antioxidants (BHA, BHT, TBHQ) in Edible oils	<10 mg/kg (antioxidants) >99% classification of antioxidant type; >94% concentration classification
502	Regression	1D-CNN	Predict amounts of pesticide residues in corn oil	FT-NIR and Raman in multispectral fusion	/	Solutions of chlorpyrifos in water and corn oil	~5.37 mg/kg LOD
503	Classification and quantification	ResFusionNet-TSMT (ResNet with Transformers)	Simultaneous pesticide classification and concentration quantification	SERS	Used database from ref 269		Classification accuracy 99.21%

**Abbreviations:** **LOD:** limit of detection; **Pb:** lead; **RNN:** Recurrent Neural Network; **GRU:** Gated Recurrent Unit; **LSTM:** Long Short-Term Memory network; **ANN:** Artificial Neural Network; **SAE:** Stacked Autoencoder; **NAS-Unet:** Neural Architecture Search U-Net; **pSCNN:** Pseudo-Siamese CNN; **NN-EN:** Neural Network with Elastic Net regularization; **SSA-BP:** Sparrow Search Algorithm-Back Propagation, **NAS:** Neural Architecture Search; **ADMIN:** Automated Design of Mixed Networks.

**S5: Reference Table for Materials Characterisation:** Examples of AI-integrated characterisation methods of plasmonic materials outlined in **Section 7.5**.

Ref	AI-type	Function of AI	Material System	Performance impact
504	DNN: implemented U-net architecture.	Denoising EDX elemental tomography maps, before using a 3D reconstruction algorithm.	Au and Ag nanoparticles	Superior method to classical denoising, with improved performance in 3D reconstructions
505	DL: combination of CNN and generative adversarial networks	Automating SLEEM image refinement.	Au nanoparticles-loaded SBA-15 mesoporous silica	Utilizing AI for SLEEM image quality refinement (contrast and resolution), improves the images sharpness/resolution.
506	CNN and gaussian mixture model	Dark field scattering image analysis of single particles.	Galvanic exchange reaction, and detection of Hg <sup>2+</sup> ions	Introducing outlier detection, for determination and improved single particle analysis.
507	DNN: U-Net CNN	Automated LSPR for plasmonic particle scattering recognition from dark field scattering imaging.	Au nanorods	Accurately extracting scattering information of plasmonic NP in cells from the background, with higher accuracy, stronger generalization ability, and robustness.
508	DL: NN	Non-invasive prediction of relative plasmon coupling, based on colour features from camera images. NN regress PC values from the mean R, G and B channel intensities to analyse colour intensities from the images.	Au nanoparticles functionalized on glass	Rapid, non-contact estimation of the relative plasmon coupling from camera images. With predictive performance metrics of R <sup>2</sup> = 0.8227, RMSE = 0.0902, MSE = 0.0081, and MAE = 0.0491
509	CNN	To analyse colour of calcite-assisted localization and kinetics (CLOCK) and predict Au nanorod dimensions.	Au nanorods	Valuable to differentiate between different size Au nanorods directly during optical microscopy experiments and predict the nanorods aspect ratio within RMSPE of ~10%.
510	NN	(shallow and deep NN) Classification of feature extraction from dark-field microscopy to identify the unique NP array features from dark-field microscopy images.	Au NP cluster arrays composed of Au (quasi) nanospheres ~70nm and Au nanorod aspect ratio of 3,5 and 2,4.	High accuracy using deep learning for the NP cluster array classification, with clusters of different Au nanorod aspect ratios and Au (quasi)nanospheres
511	NN	Analysis of NP shapes from in-situ XANES data, NN trained on numerical XANES spectra	Ag clusters	Extract, size, shape and distances in silver clusters, for monitoring temperature dependence of Ag aggregation
512	DL: gated recurrent neural network (GRU)	Predict the composition of multisized gold nanosphere colloids from extinction spectra	Au NP colloid	Advantageous, as GRU can read single and double peak extinction spectra
206	RF	Direct prediction of absorption from particle size.	Ag nanostructures	Using RF achieves the lowest MAE and works well with a smaller dataset.

513	DL: CNN and long short-term memory (LSTM)	Directly extract the viscosity related features, by analysing the time-series data, of tiny tracer particle rotating inside biological condensate. To then predict the viscosity.	Au nanorods	Better than traditional curve-fitting-based methods, with much higher sensitivity and accuracy.
-----	---	---	-------------	---

**Abbreviations.** **CNN:** convolutional neural networks, **DL:** deep learning, **DNN:** deep neural networks, **MAE:** mean absolute error, **MSE:** mean square error, **NP:** nanoparticle, **NN:** neural network, **RMSE:** root mean square error, **RMSPE:** root mean percent error.

## S6: References corresponding to S1

1. P. C. Lee and D. Meisel, *The Journal of Physical Chemistry*, 1982, **86**, 3391-3395.
2. J. A. Creighton, C. G. Blatchford and M. G. Albrecht, *Journal of the Chemical Society, Faraday Transactions 2: Molecular and Chemical Physics*, 1979, **75**, 790-798.
3. X. Zheng, W. Xu, C. Corredor, S. Xu, J. An, B. Zhao and J. R. Lombardi, *The Journal of Physical Chemistry C*, 2007, **111**, 14962-14967.
4. K. G. Stamplecoskie and J. C. Scaiano, *Journal of the American Chemical Society*, 2010, **132**, 1825-1827.
5. Y. Sun and Y. Xia, *Science*, 2002, **298**, 2176-2179.
6. D. Yu and V. W.-W. Yam, *Journal of the American Chemical Society*, 2004, **126**, 13200-13201.
7. A. R. Siekkinen, J. M. McLellan, J. Chen and Y. Xia, *Chemical Physics Letters*, 2006, **432**, 491-496.
8. M. Maillard, S. Giorgio and M.-P. Pileni, *The Journal of Physical Chemistry B*, 2003, **107**, 2466-2470.
9. B. Xue, D. Wang, J. Zuo, X. Kong, Y. Zhang, X. Liu, L. Tu, Y. Chang, C. Li, F. Wu, Q. Zeng, H. Zhao, H. Zhao and H. Zhang, *Nanoscale*, 2015, **7**, 8048-8057.
10. N. R. Jana, L. Gearheart and C. J. Murphy, *Advanced Materials*, 2001, **13**, 1389-1393.
11. J. Patarroyo, A. Genç, J. Arbiol, N. G. Bastús and V. Puntes, *Chemical Communications*, 2016, **52**, 10960-10963.
12. M. A. Mahmoud and M. A. El-Sayed, *The Journal of Physical Chemistry Letters*, 2013, **4**, 1541-1545.
13. B. Nikoobakht and M. A. El-Sayed, *Chemistry of Materials*, 2003, **15**, 1957-1962.
14. A. Garcia-Leis, J. V. Garcia-Ramos and S. Sanchez-Cortes, *The Journal of Physical Chemistry C*, 2013, **117**, 7791-7795.
15. Y. Wang, P. H. C. Camargo, S. E. Skrabalak, H. Gu and Y. Xia, *Langmuir*, 2008, **24**, 12042-12046.
16. R. Mravljak and A. Podgornik, *ACS Omega*, 2023, **8**, 2760-2772.
17. L.-C. Yang, Y.-S. Lai, C.-M. Tsai, Y.-T. Kong, C.-I. Lee and C.-L. Huang, *The Journal of Physical Chemistry C*, 2012, **116**, 24292-24300.
18. M. Lessard-Viger, M. Rioux, L. Rainville and D. Boudreau, *Nano Letters*, 2009, **9**, 3066-3071.