



PROMISING RESEARCH IN THE SENSING DOMAIN OF SILVER NANOPARTICLES: ITS ONGOING AND FUTURE ASPECTS

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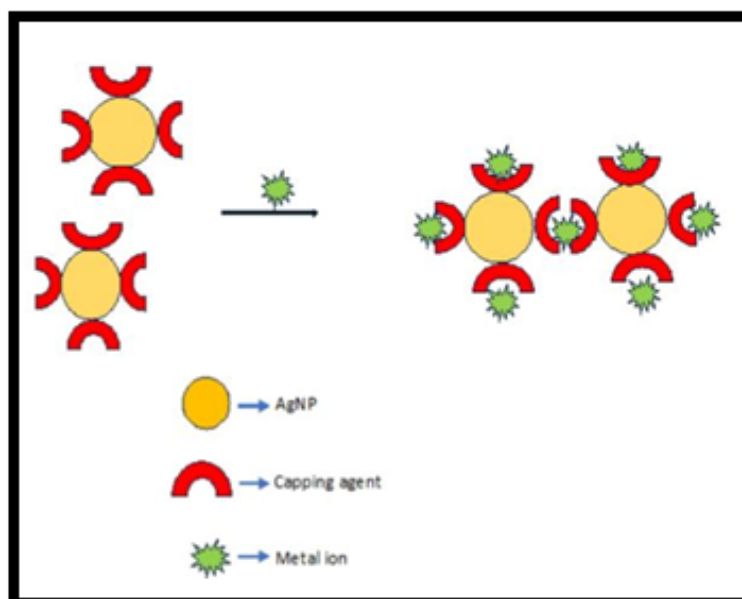
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Abstract

The usage of silver nanoparticles has increased a lot in recent years owing to their distinctive physical and chemical detection properties. Owing to their SPR bands and tuneable optical properties AgNPs based sensors have fascinated a lot, they have proved to be appropriate for their selective and sensitive colorimetric and fluorimetric probe. The current work focusses detailed study based on synthesis of AgNP and its physical, chemical, and biological detecting ability towards metal ions, in various aqueous solutions and in the environment.

Graphical Abstract



Keywords Silver nanoparticles, colourimetric and fluorescence sensors

Introduction

In recent years, the development of advanced materials and sensing technologies has greatly expanded our capabilities in detecting and monitoring various environmental pollutants, particularly heavy metal ions. Metal elements are essential components for living body [1,2]. However, any metal ion with too higher and/or lower level would destroy the balance of metabolism. Thus, quantitative determination of metal ions concentration is the key to ensure the metal ions on health level amongst them heavy metals are potential environmental pollutants as many of them are toxic even at trace ppm level [3,4]

Currently the research based on nanotechnology has increased, as the properties of materials at nanoscale are totally different that may not be predictable from those observed in bulk materials, on account of surface-to-volume and quantum size effects [5]. Nanomaterials are the structured components in which one of its dimensions (width, length or thickness) is at least in size range from 1 to 100 nm [6]. Normally, the size, shape and surface modification of metal nanoparticles comprises some of the most important factors that can dramatically affect their properties [7].

Nowadays, many analytical techniques, such as atomic absorption spectrometry, inductively coupled plasma mass spectrometry and inductively coupled plasma emission spectrometry, have been applied to the detection of metal ions. However, these methods usually need sophisticated equipments and tedious sample preparation steps. Moreover, chemosensors fabricated from fluorescent nanoparticles, such as carbon dots, graphene quantum dots, and noble metal nanoparticles or nanoclusters for sensing of metal ions have attracted wide attention due to their good selectivity, high sensitivity and easy operation [8,9]. Colorimetric assay has also been extensively utilized to detect heavy metal ions in aqueous solution because of the cost-efficient and less time-consuming procedures compared to other methods [10 -13].

The selectivity and sensitivity chemosensory for the detection of heavy metal in the environment is essential, due to the high toxicity to living organism. The sensor should be simple, portable and highly sensitive, which can sense heavy metal ions in the nano molar level. Nano sensor had been used for the identification of environmental pollutant by showing a change in their properties of nanoparticle (change in the colour) or aggregation of nanoparticle indicates the sensing of the particle [14].

Also, gold and silver nanoparticles have generated a great interest as a research tool spanning all states of matter. Particularly silver nanoparticles (AgNPs) have various significant applications in exclusive fields. It is widely used as an antibacterial and antifungal agent and have strong optical features which makes them suitable for biological sensing and imaging [15 - 17]. Now a day, noble metal nanoparticles, including Ag NPs are widely used as chemo-sensors in the detection of a number of important compounds [18].

Among the coinage metal nanoparticles (Cu, Ag, Au), it is well-known that silver has the strongest surface plasma resonance, even with the size down to 2 nm. Therefore, most of SPR-related fundamental investigations and sensing applications have focused on silver nanoparticles. Photocatalytic and antimicrobial properties of Ag NPs are contributed due to the release of silver ions from the crystalline surface of nanomaterials [19, 20]. The introduction of organic ligands onto the surface of metal nanoparticles provides the stability of these Nano entities in different solvents as well as the desired surface functionality [21-23]

NPs can be produced using physical, chemical, biological, methods by and large, two approaches are: (1) top to bottom. (2) bottom to top approaches. Metallic nanoparticles can be synthesized using different approaches, such as electrochemical methods, decomposition, microwave-assisted techniques, and wet chemical procedures [24,25]. However, the chemicals used in those methods are usually flammable and highly toxic [26], which limit their applications in many areas because they may pose risks to human health [27]. Therefore, there is an increasing need to promote a safe and environmentally friendly procedure to synthesize metallic nanoparticles without using toxic chemicals [28]. Notably, the development of effective green chemistry methods has received much attention as an alternative approach to synthesize metal nanoparticles, which can eliminate or minimize

the generation of toxic or hazardous waste materials and establish a sustainable process [29]. Conventional methods for metal ion detection usually involve expensive equipment and time-consuming procedure.

Now day, green synthesis of NPs is major focus of the researchers, bio synthesized a kind of bottom – up approach of NPs synthesis. The biological approaches used for AgNPs synthesis are plant extract which containing flavonoids, terpenoids, etc. [30]. Fig. (1) illustrates chemical structure of some different phytoconstituents responsible for the green synthesis of AgNPs.[31]

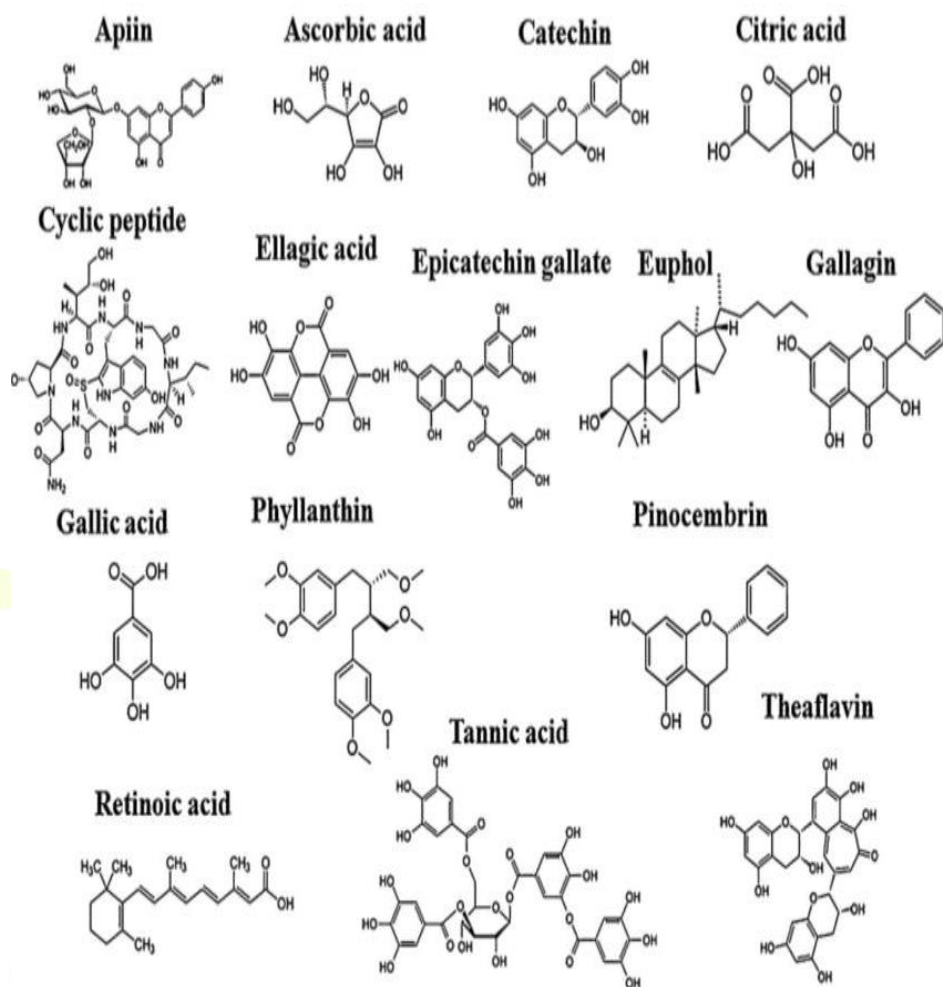


Figure 1: The chemical structure of different phytoconstituents responsible for the green synthesis of AgNPs.

Due to SPR bands and tuneable optical properties, silver nanoparticles have a strong absorption band in the visible region. Similarly, functionalized AuNPs have also been proven suitable to serve as sensitive and selective colorimetric probes for the detection of heavy metal ions. In fact, advantages of using NPs as chemosensors are proven to be a promising approach for simple and cost-effective protocols with high sensitivity and rapid tracking of valuable and toxic metal ions in environmental systems. In fact, metal nanoparticles can be converted into powerful nanoscale chemical sensors by manipulating their surface with those molecules which can introduce new functionality and also possess additional binding sites to interact with substrates, such as metal ions in solution. Therefore, detection and sensing of heavy and transition metal ions via nanoparticles [32, 33] are topics of recent interest in nanoscience.

The advantages of AgNPs-based chemosensors is their remarkable sensitivity. Even at trace concentrations, metal ions can induce significant changes in the optical properties of AgNPs, allowing for the precise quantification of metal ions in complex sample matrices. Moreover, functionalization of AgNPs with specific ligands enhances the selectivity of these sensors. they have been used to detect a wide range of metal ions,

including heavy metals in drinking water, toxic ions in food samples, and metal ions as biomarkers in clinical settings.

While AgNPs-based chemosensors offer promising advantages, they also face challenges related to stability, reproducibility, and interference from other ions in complex samples. Researchers are continuously working to address these issues and improve the practicality of these sensors.

Colorimetric sensors

Colorimetric sensor typically relies on the principles of molecular recognition, formation and signal transduction. Generally, Chemosensors are designed with specific ligands or receptors that can selectively bind to the heavy metal ions. Upon binding to the target metal ions, and formation of complex with metal ion its often result in colour change. The chemosensor undergoes a change in its physical or chemical properties. This change can be used to generate a measurable signal, indicating the presence of the metal ion. Additionally, they need to be sensitive enough to detect even trace concentrations of the ions. To quantify the concentration of heavy metal ions, the response of the chemosensor can be calibrated against known standards. This allows for accurate measurement. The sensitivity of the sensor determines its ability to detect low concentrations of heavy metal ions. The limit of detection is the lowest concentration of heavy metal ions that the sensor can reliably detect and quantify. It is an important parameter for evaluating the sensor's performance. The key to their success lies in the selective complex formation and the resulting colour change, which is directly related to the concentration of the target heavy metal ions in the sample.

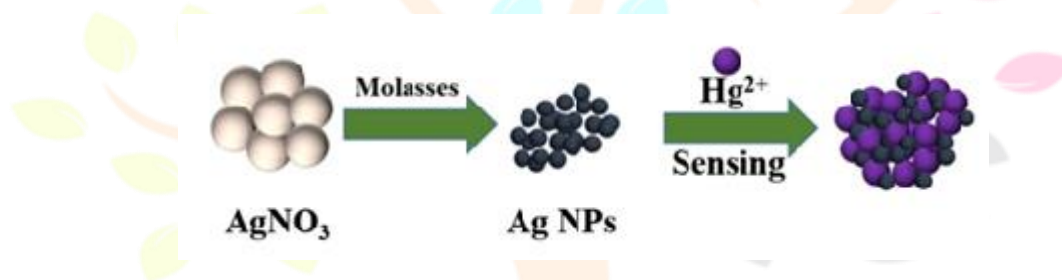
In addition to AgNPs based colorimetric probes, some other attempts have been reported for the determination of heavy metal ion. A few selected probes implemented for detection of various heavy metals are summarized in Table (1)

TABLE 1: Probes of Ag Nps based colorimetric sensor for detection of heavy metal ions

Probe	AgNPs ligand	Shape and Size	Functional groups	Limit of Detection	Metal detection	Reference No.
1.	Molasses	Spherical, 16nm	-OH , -NH C=N ,C=O	0.025 μ M	Hg ⁺²	[34]
2.	Ciric acid, Flavinoids, Hesperidin	Irregular, 55nm	-NH ₂ , C=O, -OH	0.25ppm	Hg ⁺²	[35]
3.	Methionine	8 \pm 1	Sulphur	-	Hg ⁺²	[36]
4.	Chlorophyll , terpenoids, proteins, etc	Spherical; <60nm	-NH ₂ C=O, -CH ₃ , CH ₂	2.7 μ M	Hg ⁺²	[37]
5.	Almond skin Extract Biomolecul e	Spherical	-OH , C=O Amino grp.	245 ppm	Fe ⁺²	[38]
6.	Polyphenoli c Catechins	Spherical ;63 – 69 nm.	-OH , - COOH, S = O ,C=C	20 –200 mgL ⁻¹	Hg ⁺²	[39]
7.	Vitamin B ₁₂ Almond skin extract	-	-OH , C=O, C=N , CH =CH	2 mgL ⁻¹	Fe ⁺³	[40]
8.	N – acetyl- L-Cysteine	Spherical ; 6.55 \pm 1.0 nm	-OH, COOH ,	80nm	Fe ⁺³	[41]
9.	Glutathione	8.0 nm	-COOH , - SH,-NH ₂	7.5 \times 10 ⁻⁵ molL ⁻¹	Ni ⁺²	[42]

10.	Fluorescein	11 ± 5 nm	-SH , -COOH	0.34 ppm	Hg ⁺²	[43]
11.	Tri-sodium	Spherical ; 10- 15 nm	C = O , -OH	1nm	Cr ⁺⁶	[44]
12.	L-Dopa	Flower shaped	-OH , -COOH	0.01ppm	Cr ⁺⁶	[45]
13.	β- Cyclo- Dextrin	Spherical; ~ 8±2 nm	-OH	37.5nm	Hg ⁺²	[46]
14	D.erecta fruit biomolecule	Spherical; 26-35 nm	-C= O NH ₂ ,OH	0.5 ppm	Cr ⁺⁶	[47]
15	LysinePeryl eneLysine (LPL)	-	-COO, -NH ₂	2 μM, 10 μM	Cr ⁺⁶ , Cd ⁺²	[48]

M. Gangarapu and P. Anaikutti et al. have modified silver nanoparticles (AgNPs) were sustainably synthesized using molasses for the colorimetric sensing of Hg⁺² ion, probe-1. The presence of various bioactive compounds in molasses played a significant role. Based on earlier reports and their study results, a plausible mechanism for the interaction of Ag NPs with Hg⁺² ions was proposed and shown in (Scheme 1).



Scheme 1: A plausible mechanism of Hg²⁺ sensing using AgNPs

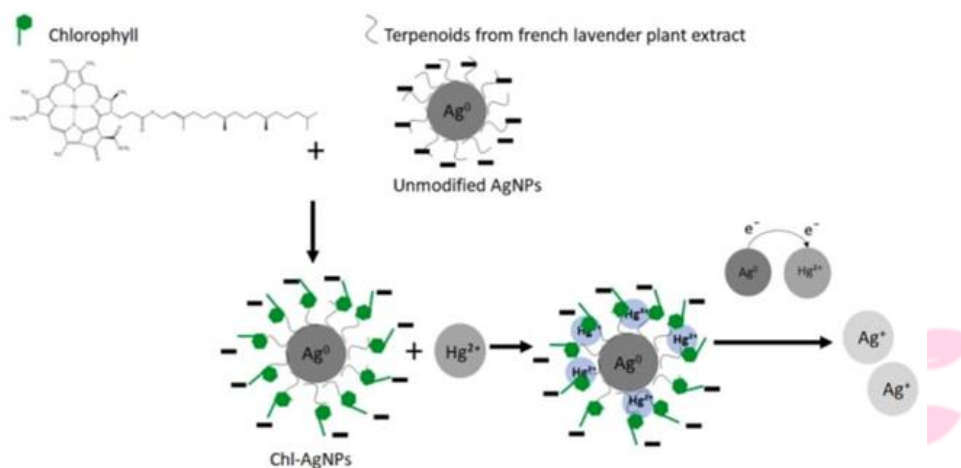
AgNPs bind with Hg (II) ion, a color change from dark brown to colorless is observed. The researchers successfully applied the AgNPs for detecting Hg²⁺ ions in real water samples, including tap water, drinking water, and river water, with good accuracy in recovery. The AgNPs also demonstrated catalytic activity, particularly for the reduction of 4-nitrophenol to 4-aminophenol in the presence of NaBH₄. The catalyst showed excellent recyclability, with a conversion efficiency of 80% after six cycles [34].

A. Aminu, S.A. Oladepo, they have reported probe-2 which is fast orange peel-mediated synthesis of AgNPs and colorimetric sensing of Hg (II) ions. Their physicochemical characterization, various factors affecting AgNPs synthesis, including silver ion concentration, extract volume, pH 10, temperature, and reaction time, were investigated. The AgNPs were characterized using UV-Vis spectroscopy, XRD, FESEM, and FTIR. They observed AgNP binding with Hg⁺² color change from golden brown to colorless. The method also showed minimal interference from other metal ions, making it suitable for environmental monitoring of mercury in drinking water [35].

The rapid colorimetric detection of Hg (II) using AgNPs in the presence of methionine was investigated by S. Balasurya, A. Syed, A. M. Thomas and Co-workers. In this probe-3, they used UV-visible spectroscopy, BET surface area analysis, scanning electron microscopy, and other methods to characterize the properties of AgNPs. The interaction of methionine AgNPs complex with Hg (II) ion was examined, showing a color change from brownish-yellow to colorless. The quantification of Hg²⁺ was carried out using different concentrations and real samples from sewage waste. They successfully detected Hg²⁺ in real samples, including sewage water, and confirmed that the presence of other metal ions did not interfere with quantification. The researchers studied the impact of

temperature, pH8, and affected by salinity on Hg^{2+} detection. They developed paper and gel tests using Ag NPs-methionine conjugate, demonstrating the detection of Hg^{2+} [36].

D. D. Yılmaz et al. synthesized probe **4** monitoring the colorimetric detection of $\text{Hg}(\text{II})$ ion using chlorophyll functionalized green silver nanoparticles in aqueous medium. The detection mechanism for Hg^{2+} involved a redox reaction with the Chl-AgNPs, causing a colour change from brown to colourless and a shift in the absorption peak. The method was tested with river water samples and showed good recovery values, making it suitable for environmental mercury detection, with polydispersity index 0.446, (Chl:AgNPs; 1:1) Scheme 2, shows the detection mechanism of mercury ions using Chl-AgNPs in the aqueous medium[37].



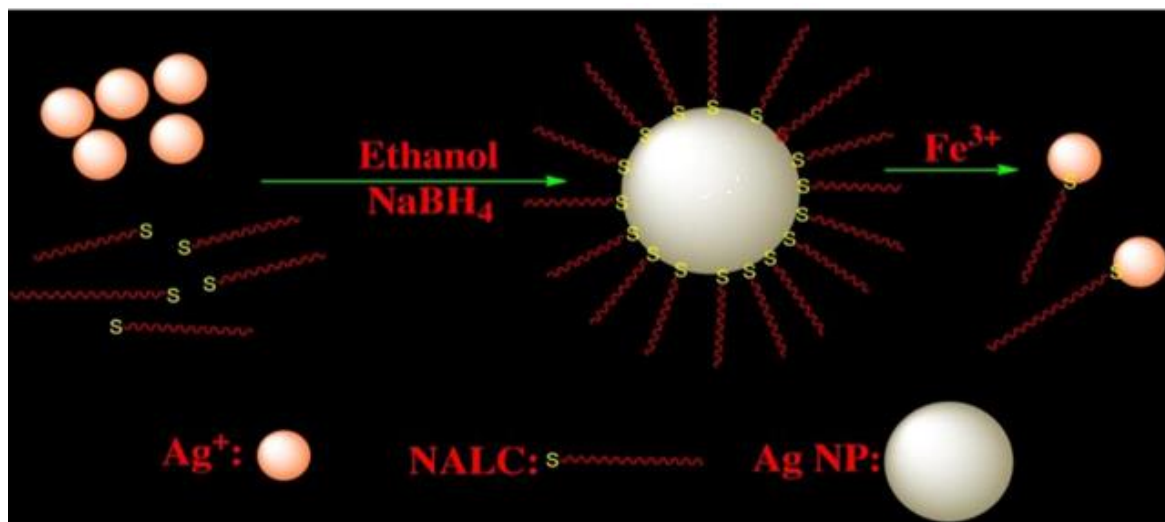
Scheme-2. Detection mechanism of $\text{Hg}(\text{II})$ ion using Chl-AgNPs

P. S. Sadalage and R.V. Patil et al. have been reported probe **5** almond skin extract mediated optimally biosynthesized antibacterial silver nanoparticles enable selective and sensitive colorimetric detection of Fe^{2+} ions. The synthesized AgNPs bind with Fe^{2+} ion through a colour change from brown to colorless. Thus, among the tested ratio, 4:1 (ASE: AgNO_3), pH 6, The recording of absorption spectra for the test of precision and accuracy of Fe^{2+} detection in the range of 70–105 ppm indicated that the increase in the concentrations of Fe^{2+} was directly proportional to the increase of A_{365}/A_{418} ratio. they were tested against gram-positive bacteria *B. Subtilis* and *S. aureus* and gram-negative bacteria *S. typhimurium*, *E. coli* and *P. vulgaris*. the zone of inhibition due to antibacterial activity of AgNPs against tested bacteria varied in the range of 7–17 mm. It was observed that *S. typhimurium* (ZOI 17 mm) was highly sensitive. The method was tested with distilled water sample and real water sample probe **5** [38].

Furthermore, P. Prema et al synthesized probe **6**, having the one step method of green tea mediated AgNPs applications in detection of Hg^{2+} having polyphenols, catechins. They observed that formation of AgNPs was confirmed by a change in colour from colourless to light yellow and eventually deep yellowish-brown. The presence of Hg^{2+} , caused a significant quenching of the AgNPs' surface plasmon resonance, resulting in a change in the solution dark brown to light brown showing selective detection of Hg^{2+} ions in the concentration range of 20 to 160 μM [39].

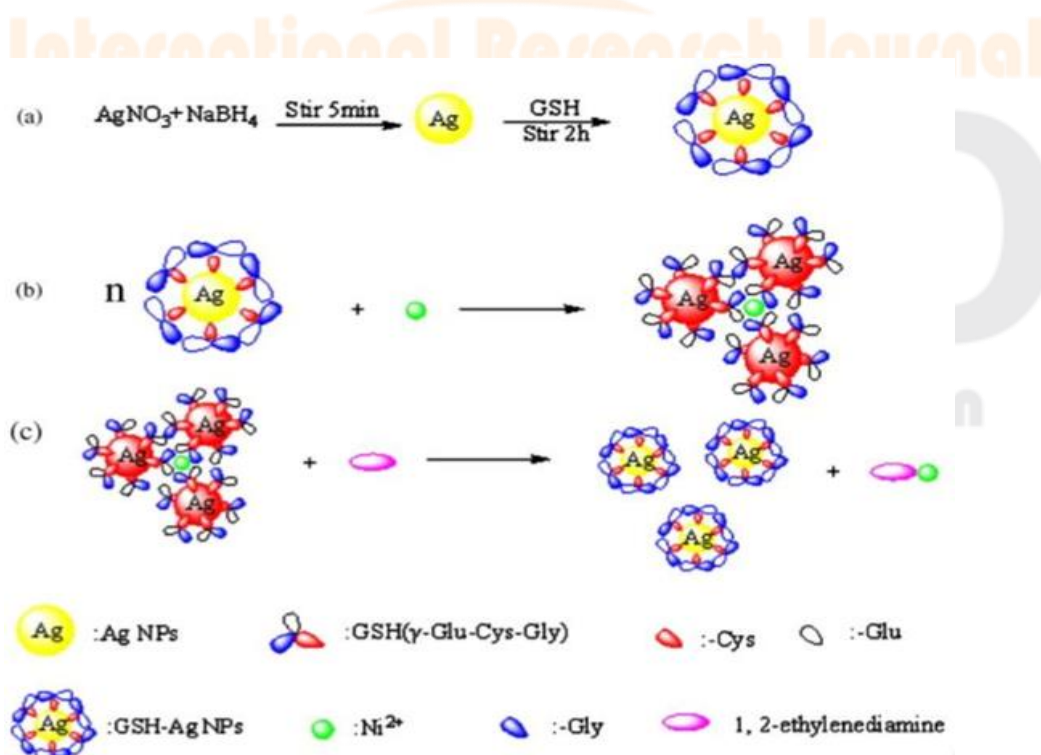
S.S. Harke et al. presented a probe **7** having a biogenic approach towards vitamin B_{12} functionalized AgNPs for the detection of iron in food samples. Researchers developed a method to detect Fe^{3+} ions using vitamin B_{12} functionalized silver nanoparticles (FAGNPs) and compared it with Atomic Absorption Spectrometry (AAS). They determined Fe^{3+} ion concentrations in a range of 1–10 mg/L using UV-Vis spectrophotometry. They used this method to analyze Fe^{3+} ion content in various food samples like pulses, legumes, cress products and found that probe **7** were more sensitive than bare AgNPs. The results from probe **7** -based detection were similar to those obtained by AAS. Probe **7** exhibit strong absorption in the UV-Vis region due to Surface Plasmon Resonance (SPR) properties, which are highly sensitive to environmental changes [40].

Furthermore, X. Gao, Y. Lu and co-workers reported probe **8** a highly sensitive plasmonic response of the N-acetyl-L-cysteine-stabilized [NALC] AgNPs for colorimetric sensing of Fe^{3+} ion. It is chemical reduction method with N-acetyl-L-cysteine (NALC) as a stabilizing ligand. The Ag NPs exhibited a strong absorption peak at around 400 nm, indicating their monodispersity and small size. The detection mechanism involves the oxidation-reduction process between the Ag NPs and Fe^{3+} ions. These measurements showed that no aggregated Ag nanoparticles were formed after Fe^{3+} ions were added. Based on these results, propose a new sensing mechanism of the as-synthesized NALC-Ag NPs for Fe^{3+} detection, (as shown in scheme 3) [41].



Scheme -3 The processes of formation of the NALC-protected Ag nanoparticles and the sensing mechanism for Fe^{3+} ions in probe **8**.

In another approach probe **9** H. Li and Co-workers reported Glutathione-stabilized AgNPs as colorimetric sensing of Ni^{2+} ion. Furthermore, an interesting phenomenon that 1,2-ethylenediamine can make the aggregated nanoparticles dispersed was observed and the possible mechanism is discussed. (Scheme 4)



Scheme 4 (a) Synthesis of GSH-Ag NPs. (b) Aggregating process of Ag NPs induced by adding Ni^{2+} . (c) The disassembly of aggregated GSH-Ag NPs

The NPs had showed some assembly features likely due to hydrogen bonding between GSH molecules. The stability of GSH-Ag NPs was assessed under varying pH>3, indicating good stability under certain conditions. The NPs exhibited a remarkable ability to recognize Ni²⁺ ions, resulting in a colour change from yellow to deep orange, with potential quantitative detection capabilities. From the structure of glutathione, both the terminal –COOH groups of glycine moiety and the free–NH₂ groups from glutamate moiety were believed to be responsible to bind to the Ni (II) centre and also participating in cross-linking. Here More interestingly, addition of 1,2-ethylenediamine can make the reaction reversible [42].

Lodeiro et al synthesized probe **10**, a novel fluorescein-functionalized gold and AgNPs were used as colorimetric probes to detect Hg⁺² ions through 1D-chain formation, primarily driven by chelation interactions between Hg⁺² and carboxylate groups on the nanoparticle's surfaces. In this study, AgNPs were prepared by mixing silver nitrate with a polyvinylpyrrolidone (PVP) solution and reducing it with sodium borohydride. The resulting AgNPs@PVP were characterized by their characteristic absorption spectrum. These AgNPs were then further modified by replacing PVP molecules with a chemosensor called **L**. Chemosensor **L** formula C₂₆H₂₄O₅S₂. At an acidic pH, **L** interacted with AgNPs, leading to the exchange of PVP for **L** and enhanced fluorescence due to increased excitation. Adjusting the pH 4.2 affected the stability and properties of AgNPs@L. Selective interaction of AgNPs@L with Hg⁺² resulted in the formation of 1D chains and a change in color from yellow to orange. This interaction was attributed to chelation between Hg²⁺ and carboxylate groups on **L** [43]. Probe **11** was reported by A. Ravindrana et al., it is based on selective colorimetric detection of nanomolar Cr (VI) in aqueous solutions using unmodified AgNPs. When exposed to Chromium (VI), the Ag NPs aggregate, causing a visible colour change. It also successfully detects Chromium (VI) in real water samples, demonstrating its practical applicability. The mechanism involves the reduction of Chromium (VI) to Chromium (III), which interacts with citrate-capped Ag NPs, leading to aggregation. This method offers a straightforward and efficient approach for on-site and real-time Chromium (VI) detection. When AgNPs interact with Cr⁺⁶ ion the change the colour solution from yellow to purple [44].

In probe **12** the researchers, P. Joshi et al., synthesized AgNPs by reducing Ag⁺ ions with L-dopa in an aqueous solution. The hydroxyl groups of L-dopa played a crucial role in the reduction process, forming dopaquinone, which further reduced Ag⁺ ions to AgNPs. Higher pH values resulting in stable and well-dispersed particles. The concentration of L-dopa also affected AgNP synthesis, with an optimized concentration of 0.001 M. The L-dopa-capped AgNPs demonstrated selective detection of Cr(VI) ions, undergoing aggregation and color change dark yellow to reddish brown in their presence [45]. Furthermore probes **13**, **14**, **15** are reported by R. Rajamanikandan et al., M. Ismalia and co-workers and Y. Sun et al it showed that the presence of ligands like β- cyclodextrin, flavonoids (D. Erecta ripe fruits), lysine perylene- lysine (LPL) respectively, it showed selectivity for Hg⁺², Cr⁺⁶, Cd⁺²[46][47] [48].

Smartphone and paper based colorimetric sensor:

Another development of smartphone based and paper-based colorimetric sensor for detection of heavy metal ions probes summarized in. Table 2

TABLE :2 Probes of Ag Nps based colorimetric sensor for detection of heavy metal ions

Probe	AgNPs ligand	Colour change	Detect metal	Limit of Detection	Phone application	Reference No.
16.	Tri Sodium Citrate	Brown to colourless	Hg ²⁺	0.86ppb	Mercury detector	[49]
17.	Tannic acid, EDTA	Colorless to bright yellow	Cr ³⁺	1.52ppm	PhotoMetrix	[50]
18.	Achillea Wilhelmsii Extract	Brown to colourless	Hg ²⁺	28x10 ⁻⁹ M	HSV spaces color picker	[51]

19.	Polyvinyl Pyrrolidone	Yellow to colourless	Hg ²⁺	10µL ⁻¹	RGB colour detector	[52]
20.	Starch	-	Hg ²⁺	0.12ppm	Adobe photoshop software, injected paper based	[53]
21.	Phenols, Sterols, Alkaloids	-	Hg ²⁺ , Cr ⁶⁺ , NH ₃	5ppb, 5-25ppm, 5ppm	Cellulose filter paper based sensors, UV-spectrometer	[54]

Nowadays, technology based on one of the novel smartphone and paper based colorimetric sensing of heavy metal ions have emerged more. In the work done on probe **16** digital images of samples in cuvettes and paper-based analytical devices (PADs) were taken using a smartphone and custom-made photo equipment (figure 2). The PADs were printed on Whatman filter paper with a modified inkjet printer. Digital image post-processing involved software like Photoshop, MATLAB, and Android Studio. To quantify the images, RGB color values were extracted from the smartphone photos and converted into a logarithmic scale. This conversion was directly related to mercury ion concentration and used for quantification. An Android app called “Mercury Detector” was created to capture and analyze images instantly, allowing for on-the-spot identification of mercury ion concentrations while minimizing errors. The study AgNPs for colorimetric mercury detection. AgNPs initially had a yellowish-brown color but became unstable after 45 days due to aggregation. The AgNPs changed color when exposed to mercury ions, making them selective for mercury detection. To improve sensitivity, a custom mini photo studio and flat cuvettes were used, meeting the EPA’s mercury threshold for drinking water. The method was also adapted for a paper-based analytical device (PAD) with consistent results. Recovery experiments in real river water samples showed accurate and precise results, with less than 2.4% error and better than 2.5% relative standard deviation.

The presence of high mercury levels in the river water indicated contamination from gold mining, posing risks to the environment and human health [49].

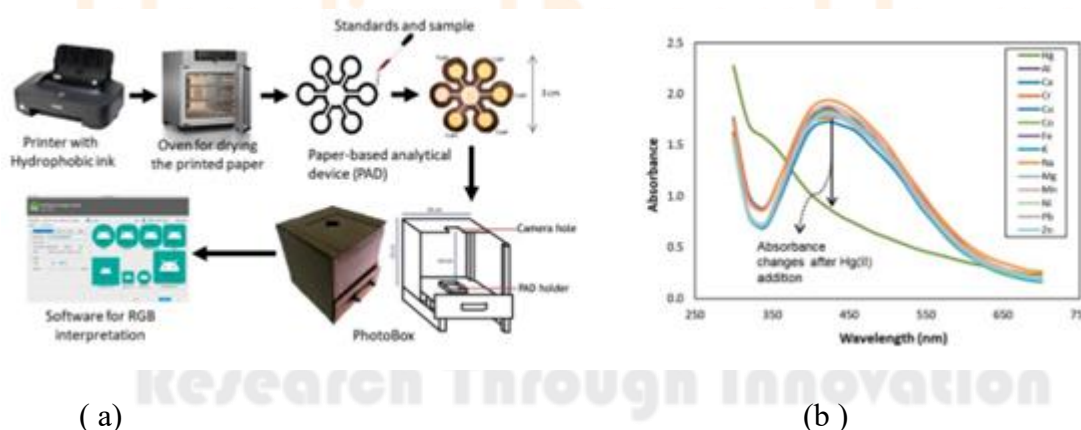
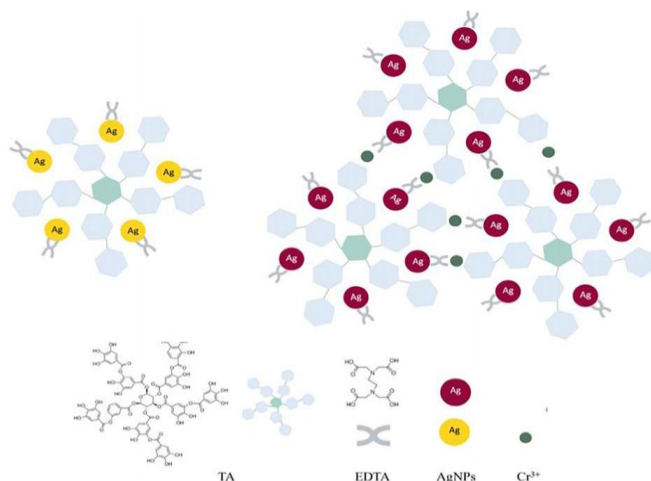


Figure 2. (a) Schematic of the paper-based analytical device (PAD) preparation and digital image acquisition for mercury ion determination. As with the colorimetric sensor, AgNPs were embedded on the PAD before sample addition. Each piece of PAD paper contains standards and a sample. (b) Change in absorbance of the AgNPs colloid in the presence of 60 ppm Hg (II) and other metal ion

S. Sangsin and co-workers another reported probe **17**. A new smartphone-based analytical tool by using EDTA-TA (Tannic acid)-AgNPs as a colorimetric sensor for determining Cr³⁺ was developed. The calibration curve for quantifying Cr³⁺ was generated using a smartphone app (Photo Metrix) by measuring RGB color intensities from digital images. Sample images were captured under controlled conditions with an iPhone 6 camera in a studio box with an LED lamp as the light source. The AgNPs exhibited a yellow color with a maximum

wavelength of 429 nm. Selective detection of Cr^{3+} was achieved, and only Cr^{3+} induced a color change in the AgNPs from yellow to wine red. UV-Visible spectra showed a new absorbance peak at 625 nm for Cr^{3+} .

Optimization included determining the best pH of 3.0 and reaction time of 10 minutes for Cr^{3+} detection. Interference studies showed good selectivity for Cr^{3+} over other species. The modified AgNPs demonstrated high sensitivity with a limit of detection (LOD) of 1.70 ppm for Cr^{3+} using a spectrophotometric method. The smartphone app (Photo Metrix) was used to measure color intensities and establish a calibration curve, with good linearity observed in the range of 2.0-5.0 ppm.



Scheme 5. Schematic illustration of possible Cr^{3+} induced-aggregation mechanism of EDTA-TA-AgNPs

Real dietary supplement samples were tested using the smartphone-based method, and the results were compared to the standard Flame Atomic Absorption Spectroscopy (FAAS) method, showing close agreement. The smartphone-based method provided accurate and precise results, making it suitable for on-site applications [50]. Another development of hand-drawn paper-based sensor for mercury; using green synthesized AgNPs and smartphone as a hand-held-device for colorimetric assay. The fabrication of a paper-based sensing platform for detecting Hg^{2+} ions using Aw (Achillea wilhelmsii extract)-AgNPs as a probe **18**. The process involves using crayon as a hydrophobic barrier on paper and stencil to create hydrophilic sensing regions.

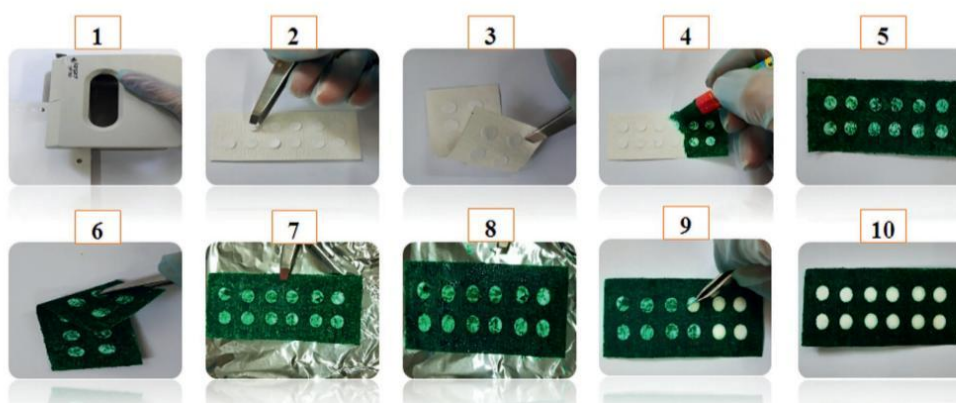


Figure 3 (a) Fabrication procedure of paper-based sensor

Optimization parameters are determined, including melting temperature $70\text{ }^{\circ}\text{C}$ and time 30 s, the amount of Aw-AgNPs and Hg^{2+} ions, and response time (figure3a). The sensing mechanism relies on the colour changes caused by the formation of Ag-Hg amalgam on the surface of AgNPs, resulting in a blue-shift of the LSPR peak. The stability of Aw-AgNPs in various environmental conditions is tested. The paper-based sensor demonstrated high selectivity for Hg^{2+} ions, even in the presence of various interfering ions. Only Hg^{2+} ions caused a color change in the Aw-AgNPs platform, brown turning it colourless sensor showed good sensitivity, with a linear relationship

between Hg^{2+} concentration and response signal. It also provided quantitative results for Hg^{2+} concentrations in the range of 100×10^{-9} to 100×10^{-6} m.

The sensors long-term stability was tested over 2 weeks, showing a slight decrease in effectiveness (8%) after this period. The paper assay was applied to real water samples from rivers, wells, and lakes, spiked with Hg^{2+} solutions. The sensor provided accurate and quantitative results with recovery rates ranging from 84% to 113.31% [51]. Another development of inkjet-printed paper-based colorimetric sensors with smartphones for selective detection of Hg^{2+} ions. Various metal ions were tested, but only Hg^{2+} caused a color change yellow to colourless in AgNPs on the paper. The color change was due to a specific reaction with Hg^{2+} . Optimization involved testing different capping agents for AgNPs, with PVP-stabilized AgNPs being the most effective for Hg^{2+} detection.

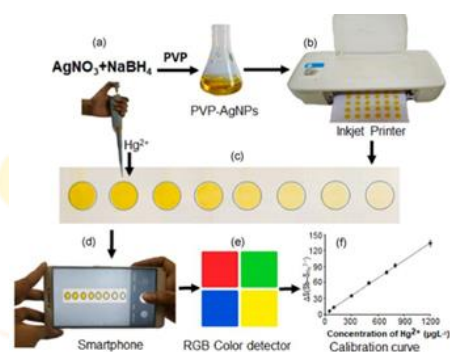


Figure 4. a) PVP-AgNPs, (b) inkjet printing of PVP-AgNPs on paper substrate, (c) deposition of Hg^{2+} ($500 \mu\text{g L}^{-1}$) on printed paper with different concentration of Hg^{2+} by micropipette, (d) recording of image using smartphone, (e) measurement of color intensity by RGB color detector app and (f) standard calibration curve.

Different concentrations of AgNPs were tested, with 0.45 mM yielding the best results. Different smartphones and paper substrates were also tested, showing that they didn't significantly affect the detection. The pH of the sample solution was found to be crucial, with a range of 3.0–6.0 pH being ideal for Hg^{2+} detection. The color intensity was measured with a smartphone, showing a significant decrease in the signal with Hg^{2+} (Figure 4a) were investigated. The mechanism involved the oxidation of AgNPs and reduction of Hg^{2+} ions. The method showed good linearity, with a linear range of 40–1200 $\mu\text{g/L}$. The sensor exhibited good precision, stability, and accuracy, making it suitable for the selective determination of Hg^{2+} ions in various water samples probe 19 [52]. A. Apilux and co-workers reported probe 20 simple and rapid colorimetric detection of $\text{Hg}(\text{II})$ by a paper-based device using AgNP probe 20 [53]

Furthermore, probe 21 experimental setup and findings is based on the optical sensing of ammonia (NH_3) and mercury (Hg^{+2}) was done using freshly prepared AgNPs and cellulose filter paper-based detection. The UV-visible spectrophotometer Shimadzu UV-1800 was used to detect AgNPs interactions with various metal ions, such as $\text{Pb}(\text{II})$, $\text{Cr}(\text{VI})$, $\text{Cr}(\text{III})$, $\text{Cd}(\text{II})$, $\text{Ni}(\text{II})$, $\text{Co}(\text{II})$, $\text{Hg}(\text{II})$, $\text{Zn}(\text{II})$, $\text{Fe}(\text{II})$, and aqueous ammonia. To sense $\text{Hg}(\text{II})$, a cuvette with 4 mL volume was used as a reaction vessel. 1.5 mL of freshly prepared AgNPs was mixed with 1.5 mL of different concentrations of $\text{Hg}(\text{II})$ solution. For sensing aqueous ammonia, different concentrations of ammonia were used. The decrease in intensity and changes in absorbance peaks at 452 nm of AgNPs after the addition of $\text{Hg}(\text{II})$ were monitored. (figure 5a). Detection of aqueous ammonia and Hg^{+2} was carried out using AgNPs-doped cellulose filter paper strips (CF-AgNPs). The strips were prepared by applying a colloidal solution of freshly prepared AgNPs to Whatman filter paper no. 1. Different concentrations of mercuric sulfate and ammonia were prepared for the detection. The time taken for decolorization of the strips when immersed in Hg^{+2} or ammonia solutions was noted. (figure 5, ab)

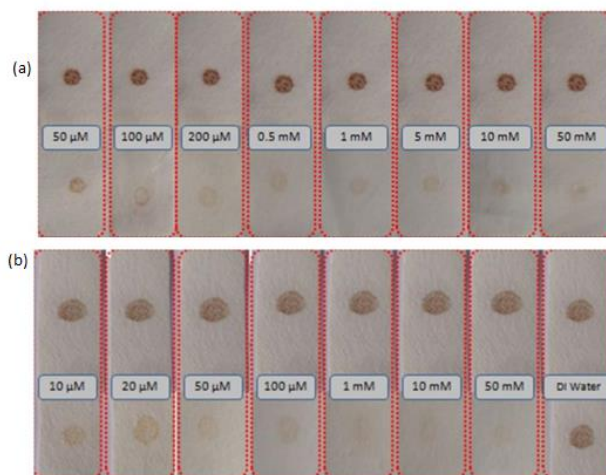


Figure 5 (a) Visual illustration of F-AgNPs based naked eye Hg(II) sensor, which show change in color after treating with different concentration of Hg (II) ion. (b) Visual illustration of F-AgNPs based naked eye ammonia sensor, which show, change in color after treating with different concentration of aqueous ammonia

The AgNPs showed selective colorimetric sensing properties for Hg⁺², Cr⁺⁶, and ammonia due to differences in their reduction potentials. Hg⁺² was detected by monitoring the shift in the SPR of AgNPs and a decrease in absorption intensity. A logarithmic curve of the ratio of absorption intensities (abs_{417}/abs_{452}) against various Hg⁺² concentrations exhibited a linear relationship. The color changes of AgNPs did not occur in response to other tested heavy metal ions (figure 5 c,d) The sensor showed visual detection of Hg⁺² down to 50 μ M concentration within an hour. For ammonia, it could detect concentrations of 10 μ M within a minute. The sensor showed promise for field deployment. The study also explored the detection of Cr⁺⁶, which is more toxic than Cr⁺³. The AgNPs underwent a color shift when exposed to dichromate anions, allowing the detection of Cr⁺⁶ in the range of 5-25 ppm, making it suitable for detecting Cr⁺⁶ in drinking water [54].

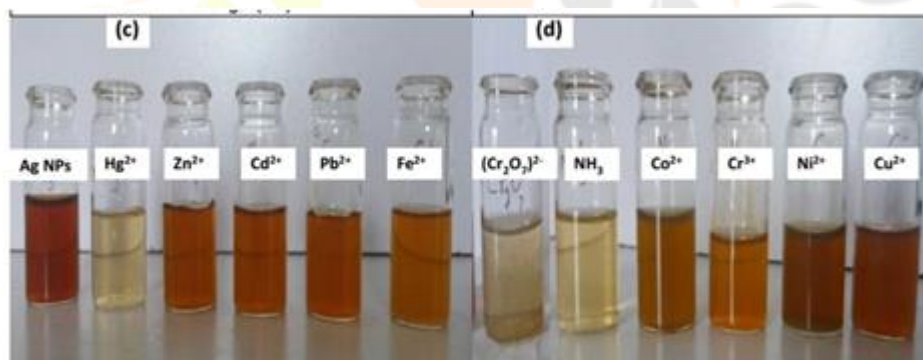


Figure 5 c,d . Visual representation of color change by different treated analyte

Fluorescent sensor

AgNPs -based fluorescent sensors for the detection of heavy metal ions operate on the principle of fluorescence quenching. The AgNPs surface is functionalized with specific ligands or molecules that have an affinity for the target heavy metal ions. These ligands can be selective to a particular metal ion. A fluorophore molecule is attached to the AgNPs or positioned in close proximity. This fluorophore emits fluorescence when excited by an appropriate wavelength of light. When heavy metal ions are introduced into the solution, they can bind to the functionalized AgNPs through chemical interactions. This binding causes a change in the electronic environment around the attached fluorophore. The presence of heavy metal ions leads to a reduction in the intensity of the fluorescence emitted by the fluorophore. This quenching effect is proportional to the concentration of the target metal ions. The quenching occurs because the energy transfer from the excited fluorophore to the metal ions depletes the fluorophore's energy state, reducing fluorescence. Many reported AgNPs based fluorescent sensors for detection of heavy metal ions probes summarized in Table 3.

TABLE 3: Fluorescent sensors based on detection of heavy metal

Probe	AgNPs ligand	Size	Functional group	Limit of Detection	Detect metal	Reference No.
22.	L- Tyrosine	5-10 nm	Carboxyl, amino	48 ppb 36ppb	Co ⁺² Cu ⁺²	[55]
23	Calix [4] Resorcinarene (-NH- NH ₂) Polyhydrazide (CPH)	7 ± 1	Hydrazide	0.1 µM	Fe ⁺³	[56]

A. Contino, G. Maccarrone and co-workers reported probe **22** for determination of copper(II) and cobalt(II) ions based on fluorescent sensors. The synthesis and characterization of L-Tyr capped AgNPs. The AgNPs were found to be highly dispersed, demonstrating stability over time. UV-Vis spectroscopy revealed a Surface Plasmon Resonance (SPR) peak at 400 nm, indicating their size. The AgNPs stability was attributed to the interaction of the amino acid tyrosine with their surface. These L-Tyr capped AgNPs were used for metal ion detection, specifically cobalt (II) and copper(II). Both metal ions caused a decrease in AgNPs absorption, and this change was attributed to the interaction with surface-bound tyrosine. The metal-induced quenching also affected the fluorescence of the tyrosine, making the AgNPs sensitive fluorescent chemosensors for metal ions. This method offers a faster and simpler alternative to traditional spectroscopic techniques for metal ion detection probe **22** [55].

Another probe **23** focused on the AgNPs as selective fluorescent sensor for Fe⁺³ ions and highly stable antibacterial activity. In this study, water-soluble calix[4] resorcinarene polyhydrazide (CPH) was used to create stable AgNPs (CPH-AgNPs) with good fluorescence properties. These nanoparticles were effective sensors for detecting Fe³⁺ ions in a specific concentration range(0.01 µM to 100 µM)pH 7. Additionally, they exhibited promising antimicrobial activity against various bacteria like Escherichia coli, Bacillus megaterium, Staphylococcus aureus, Bacillus subtilis, by Muller Hinton Agar (MHA) plates. [56]

Optical Sensor , Surface-Enhanced Raman Spectroscopy (SERS) Substrates, UV – Visible Spectroscopy

In the probe **24** cyclic peptide (CP) was used as a coating and reducing agent to synthesize CP-coated AgNPs (CP-AgNPs). The main application of CP-AgNPs was as a sensing probe for Hg²⁺ ions, showing selective binding to mercury ions compared to other metal ions. The effect of pH on the complex formation between CP-AgNPs and Hg²⁺ was investigated. Interference studies showed that CP-AgNPs were highly selective for Hg²⁺ even in the presence of other metal ions. FT-IR spectroscopy and AFM analysis confirmed the complex formation between CP-AgNPs and Hg²⁺.

A Jobs plot determined the binding stoichiometry, indicating a 1:2 ratio of CP-AgNPs to Hg²⁺. The research also demonstrated the applicability of CP-AgNPs for the detection of Hg²⁺ in tap water and human blood plasma samples, showing no interference from sample ingredients [57]. M.S. COUTINO et al., reported on the use of Ruthenium-modified AgNPs (RU-AgNPs) as chromogenic probes for detecting Fe³⁺ ions in aqueous solutions in the presence of ascorbic acid (AA) probe **25**. The addition of Fe³⁺ to RU-AgNPs results in a significant colorimetric change and is selectively detected compared to other metal cations. Ascorbic acid (AA) plays a crucial role in the selective detection of Fe³⁺. In the absence of AA, other cations induce stronger colorimetric changes. The mechanism of Fe³⁺ detection involves the growth and aggregation of AgNPs, with

strong alkaline conditions stabilizing the NPs. The interaction between RU-AgNPs and Fe^{3+} resulted in a combined growth and aggregation process given in Figure 6., leading to changes in the system 's color and UV-vis spectra. A calibration curve was constructed, demonstrating a good correlation between the absorbance at 396 nm and Fe^{3+} concentration. Furthermore, the paper introduced a practical approach using a conventional smartphone and chemometrics for the quantification of Fe^{3+} in aqueous solutions. The RGB-based values from smartphone images were correlated with Fe^{3+} concentrations, providing a simple and cost-effective method for Fe^{3+} quantification [58].

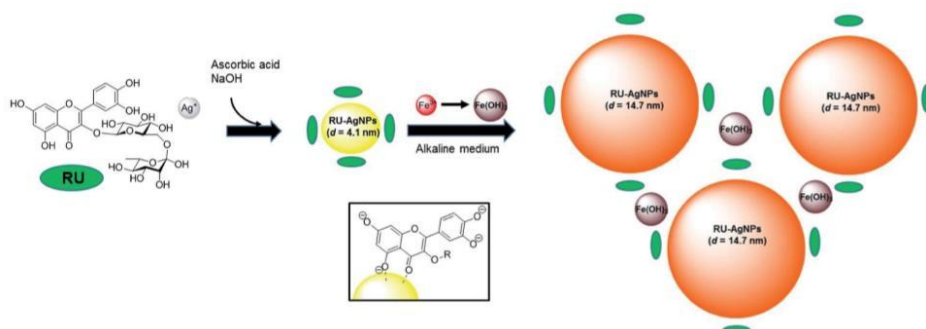


Figure 6. Mechanistic proposal for growth/aggregation of RU-AgNPs in the presence of Fe^{3+} . Insert: binding model for RU-AgNPs

Y. Luo and Co-workers reported probe **26** for detection for Hg^{2+} by silver nanoparticle in xylan solution via Tollens reaction. The formation of xylan-AgNPs composites was characterized. XRD patterns confirmed the presence of pure AgNPs with characteristic peaks, matching the face cubic center lattice of Ag metal. XPS analysis revealed the presence of Ag and a shift in Ag 3d peaks, indicating interactions between AgNPs and xylan. EDS confirmed the presence of oxygen from xylan, indicating capping. HRTEM and SAED revealed monocrystalline AgNPs. The structure of xylan before and after the reaction was analysed using FT-IR, ^1H NMR, ^{13}C NMR, and 2D-HSQC NMR. The results showed that the backbone structure of xylan remained after the reaction, but side chains, such as O-acetyl and 4-O-Me- α -D-GlcpA groups, dropped during the synthesis of AgNPs. New signals appeared, indicating the formation of oxidized reducing aldehyde groups. The study also explored the Hg^{2+} detection capacity of xylan-AgNPs composites. UV-vis spectra revealed a color change light yellow to colourless shift in absorption peak with increasing Hg^{2+} concentration. TEM images showed AgNPs aggregation. The selectivity of the xylan-AgNPs composites for Hg^{2+} was demonstrated, as other metal ions had minimal influence on Hg^{2+} detection [59].

Furthermore, S. Tamilselvan, R.M. Soniya et.al reported probe **27** sensing of eight metal ions in aqueous solutions by Silver nanoparticles based spectroscopic. When these metal ions were added to Ag NPs, it caused distinct color changes, and UV-Vis spectroscopy was employed to analyze the spectral shifts. The interaction between Ag NPs and metal ions was also examined using FT-IR analysis. As shown in figure 7 [60]

Research Through Innovation

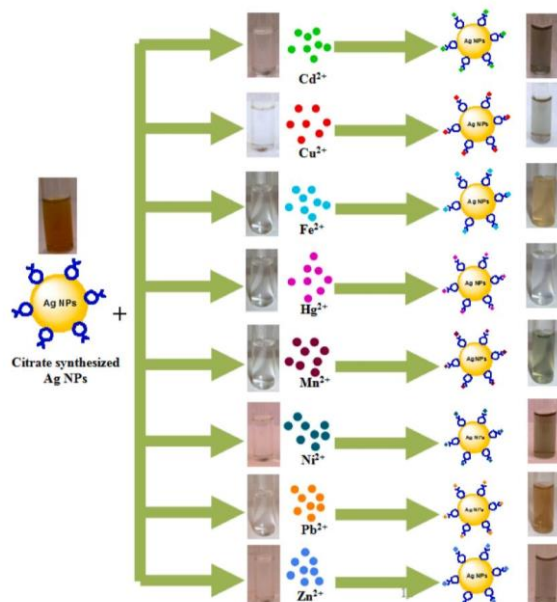


Figure 7. AgNPs interact with metal ions.

Probe **28** by C. Pattanaik et al showed glucose-capped stable AgNPs a cost-effective sensor for the selective detection of Hg^{2+} ions in aqueous solutions. This study outlines the synthesis of glucose-capped AgNPs using tulsi plant extract. However, remarkable colour change from brown to colourless was noticed when the AgNPs binding with Hg^{+2} ion [61]. Another probe **29** by I.O. Maralia et al., were synthesized AgNPs using citrus peel extracts from orange, tangerine, and lemon fruits. The citrus peel extracts contained various functional groups, which acted as reducing and capping agents for AgNPs. The AgNPs were self-assembled onto glass slides, creating suitable substrates for Surface-Enhanced Raman Spectroscopy (SERS) measurements. The AgNPs from orange and tangerine extracts were found to be more suitable for SERS due to their defined LSPR bands in the visible range, while the lemon extract-derived AgNPs also showed potential for SERS applications. The results indicated that the SERS intensities were particularly high for 4-aminobenzenethiol (4-ABT) as compared to the dye molecules. This eco-friendly and cost-effective method for AgNP synthesis and SERS substrate preparation holds potential for applications in SERS sensors and catalysis [62].

Bioactivities

AgNPs have gained significant attention in the field of medicine and material science due to their remarkable antimicrobial and antibacterial properties. These properties are attributed to their small size and high surface area, which enable them to interact effectively with microorganisms. Their small size allows them to penetrate the cell wall and disrupt its structure. This interference can lead to cell death. AgNPs release silver ions (Ag^+), which are highly toxic to bacteria. These ions can interfere with vital cellular processes, such as respiration and DNA replication, ultimately killing the microorganisms. [63-75]

CONCLUSION

This review comprises of information about the synthesized AgNPs, which are capped with various ligands and bioactive molecules. This ligand acting as capping agent and reducing agent. In this paper a large number of various approaches for the development of the fluorimetric and colorimetric sensor based on Silver Nanoparticles have been successfully applied for the detection of heavy metal ions. Researchers have created innovative colorimetric sensors for heavy metal-ions- detection using smartphones and paper-based platforms. The studies highlight the potential for on-site applications and environmental monitoring, showcasing advancements in colorimetric sensing technology. Here, various approaches on AgNPs antimicrobial/ antibacterial activity.

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