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Electrical Response of Chitosan Thin Films Toward Cu^{2+}, Cd^{2+} and Zn^{2+} Ions using Interdigitated Electrodes for Heavy Metal Ions Sensing Applications

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ABSTRACT

Real-time detection of heavy metal ions in water remains a challenge due to the complexity and cost of conventional analytical techniques. While chitosan exhibits strong affinity toward metal ions, there is limited understanding of how ion adsorption influences its electrical properties, hindering the development of low-cost electrical sensors for water quality monitoring. In this study, the electrical response of chitosan thin films toward Cu^{2+}, Cd^{2+} and Zn^{2+} ions were investigated using interdigitated electrodes (IDEs). Chitosan films were exposed to metal ion solutions and metal uptake was quantified using atomic absorption spectroscopy. Frequency-dependent resistance, capacitance, and impedance were measured to evaluate the influence of ion adsorption on electrical properties. The results show that increasing metal ion concentration leads to higher adsorption on the chitosan film, increased capacitance, and decreased in resistance and impedance. Cu^{2+} exhibited the strongest electrical response at higher concentrations, and the result is consistent with its higher adsorption affinity toward chitosan. Comparative analysis reveals that electrical parameters correlate directly with the amount of adsorbed ions. These findings demonstrate that adsorption-induced changes in electrical properties provide a reliable transduction mechanism for metal ion detection. The study suggests the potential of chitosan-based electrical sensing platforms as simple, low-cost candidates for portable water quality monitoring.

1. Introduction

Heavy metal contamination is a major environmental and public health concern due to the persistence, bioaccumulation, and toxicity of metallic pollutants in aquatic ecosystems. Contaminants like lead (Pb) cadmium (Cd), and chromium (Cr) are particularly challenging due to their toxicity and harmful effects even at very low concentrations. The sources of heavy metal contamination are diverse, such as industrial discharges and agricultural runoff. These metals are widely associated with severe health effects, including neurological disorders, kidney and liver damage, cardiovascular disease, reproductive toxicity, and increase risk of cancer [1,2]. While copper

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and zinc are essential trace elements involved in various biological processes, excessive exposure can lead to adverse health effects [3]. Cadmium, in contrast, is a highly toxic metal with no known physiological function and has been associated with kidney dysfunction, skeletal damage, and carcinogenicity [4]. Consequently, the development of rapid, sensitive, and cost-effective technologies for monitoring heavy metal ions has become increasingly important for environmental protection and public safety.

Unlike many organic pollutants, heavy metals are non-biodegradable and bioaccumulative. It means small concentrations and undetected heavy metal in water able to magnify along food chains and progressively increase human body burdens over time [5]. Meta-analyses and regional risk assessments consistently report metal concentrations in surface and drinking waters exceeding WHO and US EPA limits [6,7].

Contamination frequently occurs with no visible signs. The metals can exceed safe levels in drinking and surface waters without altering taste, color, or odor [8,9]. This situation makes managing heavy metal pollution a significant challenge. This dangerous especially for children because they experience higher risks than adults at comparable exposure levels [10]. The toxicity is harmful to aquatic organisms and ecosystem too [11]. It disrupts fundamental biological processes and poses hazards that reverberate through entire environments. It also interferes with cellular functions by inducing oxidative stress and impairing physiological and reproductive processes in aquatic organisms [12]. For example, exposure to cadmium can lead to less reproductivity among fish species, posing risks to their populations and ultimately influencing entire aquatic ecosystems [13].

Addressing heavy metal contamination requires integrated remediation strategies that combine various methods tailored to specific site conditions, contaminant speciation, and risk endpoints [14] [15]. Remediation can be broadly classified into three approaches: (i) physico-chemical methods, (ii) biological methods, and (iii) hybrid systems. Table 1 shows some studies on heavy metal remediation approaches.

Table 1
Integrated remediation methods for heavy metal-contaminated water

Method	Example of Remediation methods	Key principle	Reference(s)
Physico-chemical methods	• Chemical precipitation, • Adsorption (activated carbon, nanomaterials, MOFs, polymers), • Membrane filtration (RO, NF, electrodialysis), • Electrochemical methods (electrocoagulation, electrodeposition)	Removal of metal ions via phase transformation, surface binding, size/charge separation, or electrochemical reactions	[16] [17] [18] [19]
Biological methods	• Biosorption (agricultural waste, fungal biomass, biochar), • Phytoremediation, • Constructed wetlands, • Microbial-assisted remediation (e.g., sulfate-reducing bacteria)	Utilization of natural biological systems to bind, uptake, or transform metal ions	[20] [21] [22] [23]
Hybrid systems	• Integrated treatment trains (e.g., precipitation–adsorption–membrane, electrocoagulation–flotation, bio-physicochemical combinations) • Source control–based multi-barrier approaches	Sequential or synergistic coupling of multiple processes to overcome single-method limitations	[24] [25] [26]

Conventional analytical techniques such as atomic absorption spectroscopy (AAS) and inductively coupled plasma-based methods offer excellent sensitivity and selectivity for heavy metal detection. However, these approaches often require sophisticated instrumentation and extensive sample preparation, limiting their suitability for on-site and real-time monitoring applications. These limitations make electrochemical sensing platforms more favorable because of their portability, low cost, rapid response, and potential for miniaturization. A range of bioremediation and sensor technologies has been explored to detect or remove heavy metal ions in aquatic environment [27]. However, the effectiveness of treatment or remediation depends on sensitivity of a device to detect metal ions and their concentration [28]. Therefore, the success of heavy metal mitigation strategies also depends on detection limits, and robustness of the sensing systems that monitor and validate the remediation process.

As sensitive and early detection is an important factor in water safety management, on-site electrical sensing approaches are increasingly used in heavy metal detection. Examples of this technique are electrochemical and field-effect transistor (FET)-based platforms, that directly convert metal ion redox reactions or binding ions into measurable electrical signals. Researchers usually assess data such as current, potential, impedance, or conductance, using compact and portable instrumentation [29,30]. Compared with laboratory-bound spectroscopic techniques such as AAS, ICP-MS, and XRF, on-site electrical sensors enable real-time monitoring with minimal sample preparation [31]. Many studies show that the performance of electrical sensors can be further enhanced through integration with nanomaterials such as graphene and carbon nanotubes. These combinations increase surface area and conductivity, thereby improving sensitivity and selectivity [32,33].

Despite these innovations, on-site electrical sensing platforms still have constraints that reduce their use in the field. Their detection limits are often higher than those of lab equipment like Inductively Coupled Plasma Mass Spectrometry (ICP-MS) when testing complex samples [34]. Moreover, the selectivity can also be affected by other metals and organometallic compounds present in the sample [35]. Additionally, problems such as signal drift, and poor long-term stability can reduce measurement accuracy and reliability during continuous field operation.

Chitosan, a naturally derived biopolymer obtained from chitin deacetylation, has been integrated into biosensing technologies to establish rapid and effective detection systems for heavy metals in aquatic environment. The physicochemical behavior of chitosan is dictated by the degree of deacetylation (DD) and its average molecular weight (Mw). These qualities determine its crystallinity, degradation kinetics, and rheological properties. Chitosan possesses unique properties such as biodegradable, biocompatible, and ability to form stable films in various environments. It contains abundant amino ($–NH_2$) and hydroxyl (–OH) functional groups that strongly coordinate with a wide range of metal ions, including Hg^{2+}, Pb^{2+}, Cd^{2+}, Cu^{2+}, Cr, and As. These virtues enabling efficient metal binding, pre-concentration, and signal amplification [36] [37]. In addition, chitosan is inexpensive, abundant, biodegradable, and can be derived from waste seafood shells, making it attractive from both economic and environmental perspectives [38-40].

While numerous studies have demonstrated that chitosan is effective at adsorbing metal ions, most of them focus on how much metal can be removed rather than how the process can be used for sensing. Literature mostly focuses on thermodynamic isotherms, kinetic modeling, and maximum removal capacities. Existing studies also often emphasis on the detection of individual metal ions by varying experimental conditions, to learn their trends of sensitivity and selectivity. As a result, studies overlook the transduction potential of the polymer. The electrical response of chitosan thin films toward different heavy metal species and overall electrical conductivity of the solid-state chitosan film are not systematically understood.

Electrical sensing using interdigitated electrodes (IDEs) provides a simple and sensitive way to turn ions' physical and chemical interactions into measurable electrical signals. Likewise, comparison of the electrical response of chitosan films exposed to different heavy metal ions under the same conditions has not been well studied. This work addresses this gap by linking the adsorption of Cu^{2+}, Cd^{2+}, and Zn^{2+} ions to changes in resistance, capacitance, and impedance of chitosan films on IDEs. It seeks to address ion–polymer interaction mechanisms, factors governing sensor performance, and the extent to which chitosan-based IDE sensors can selectively discriminate among these metal ions.

2. Methodology

2.1 Materials

Medium molecular weight chitosan powder was purchased from Sigma-Aldrich (USA). Standard aqueous solutions (1000 mg/L) of copper (Cu^{2+}), cadmium (Cd^{2+}), and zinc (Zn^{2+}) ions were obtained from Fisher Scientific (UK). All reagents were of analytical grade and used without further purification. Deionized water was used throughout all experiments. Deionized water was used throughout the study. Gold interdigitated electrodes (IDE) fabricated on ceramic substrates were used for electrical measurements.

2.2 Preparation of Chitosan Thin Films

Chitosan thin films were prepared by dissolving chitosan flake in 100 ml of 1% (v/v) acetic acid at 55 ^{o}C under continuous stirring until a homogeneous solution was obtained. The resulting solution was cast onto clean substrates to form uniform thin films and left to dry at room temperature until solvent evaporated. The dried films were carefully peeled and cut into 8 mm × 8 mm specimens. This size is slightly bigger than the electrode area of the IDE to give full coverage of the sensing area (6 mm x 6 mm). The average thickness of the films was 0.132 mm. Prior to metal ion exposure, all films were stored in a desiccator to minimize moisture absorption.

2.3 Metal Ion Adsorption and Electrical Measurement

Chitosan films were immersed in aqueous solutions containing Cu^{2+}, Cd^{2+}, or Zn^{2+} ions at concentrations of 1, 2, 10, 30, and 50 mg/L. These concentration levels were selected to represent environmentally relevant contamination ranges commonly reported in aqueous systems.

After the films were immersed in ionic solutions for 20 minutes, they were gently removed and excess solution was allowed to drain. The remaining metal ion concentration in solution was quantified using atomic absorption spectroscopy to determine the amount of metal adsorbed onto the chitosan film. Calibration standards for atomic absorption spectrometry (AAS) were prepared separately for each metal ion to ensure analytical accuracy and linear response.

The treated chitosan films were then placed onto gold interdigitated electrodes connected to an LCR meter. The electrodes featured finger and gap widths of 200 µm deposited on a ceramic substrate. Gold electrodes were selected due to their stability and suitability for impedance-based sensing. Impedance (Z), resistance (R), and capacitance (C), were measured over a frequency range of 10 Hz to 200 kHz using an applied AC voltage of 1 V. Open- and short-circuit calibrations were conducted prior to measurements to minimize parasitic effects. All measurements were conducted under identical conditions to enable direct comparison among different metal ions and concentrations.

3. Results and Discussion

3.1 Adsorption of Metal Ions on Chitosan Films

The adsorption behavior of Cu^{2+}, Cd^{2+}, and Zn^{2+} ions on chitosan films, quantified using atomic absorption spectrometry, is summarized in Table 2. For all metal ions, the amount adsorbed increased with increasing initial solution concentration, indicating effective interaction between metal ions and the functional groups of the chitosan matrix. This trend confirms that the availability of binding sites and the driving force for adsorption increase with concentration.

At higher concentrations (30–50 mg/L), Cu^{2+} exhibited the highest adsorption followed by Zn^{2+} and Cd^{2+}. In contrast, at lower concentrations (1–2 mg/L), Cd^{2+} showed comparatively higher adsorption than Cu^{2+} and Zn^{2+}. These observations indicate that adsorption behaviour is not governed solely by concentration but also by the intrinsic affinity of different metal ions toward chitosan. The concentration-dependent adsorption trends provide the basis for interpreting the corresponding electrical responses discussed in subsequent sections.

Table 2
Adsorption behaviour of Cu^{2+}, Cd^{2+}, and Zn^{2+} ions on chitosan films

Film	Initial Concentration (mg/L)	Cu^{2+} Adsorption (mg/L)	Cd^{2+} Adsorption (mg/L)	Zn^{2+} Adsorption (mg/L)
A	1	0.1419	0.3918	0.1719
B	2	0.1938	0.2895	0.8600
C	10	1.68	0.5850	2.30
D	30	6.92	3.24	5.27
E	50	11.58	5.53	9.53

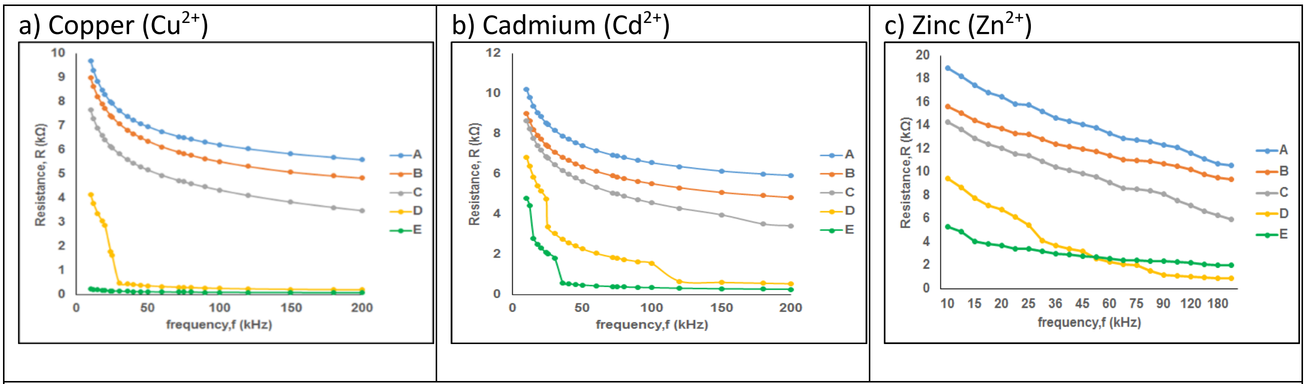
3.2 Frequency-Dependent Resistance Response

Figure 1 (a), (b) and (c) show the frequency-dependent resistance of chitosan films after adsorption of Cu^{2+}, Cd^{2+}, and Zn^{2+} ions, respectively, at different concentrations. For all metal ions, resistance decreased with increasing frequency and increased adsorption. For Cu^{2+} treated films, resistance decreased progressively from film A to film E, corresponding to increasing copper adsorption. A similar trend was observed for Cd^{2+} treated films, where higher cadmium adsorption resulted in lower resistance values. Zn^{2+} treated films also exhibited decreasing resistance with frequency.

Many studies consistently identify metal–amine/oxygen coordination and ion-exchange as the key mechanisms generating mobile charged species in chitosan matrices [41,42]. More adsorbed Cu^{2+}, Cd^{2+} and Zn^{2+} thus increase the density of charge carriers in the film [43].

At higher concentrations (30 and 50 mg/L), deviations in the resistance trend were observed. This kink reflecting non-homogeneous ion accumulation on the chitosan surface, which affects ion mobility and frequency response. It can be seen for Zn^{2+} there is an intersection between the resistance curves of films D and E at approximately 50 kHz indicates differences in ion mobility arising from variations in ion distribution.

Comparison at the highest concentration (50 mg/L) further highlights metal-dependent resistance behaviour. Zn^{2+} treated films exhibited the highest resistance, whereas Cu^{2+} treated films showed the lowest resistance, confirming stronger adsorption of Cu^{2+} ions by chitosan. Since resistance reflects the real component of impedance, lower resistance corresponds to increased mobility of charge carriers and enhanced conductivity [44]. The results demonstrate ion distribution influences frequency-dependent response, hence confirming resistance as a sensitive parameter for metal ion sensing.



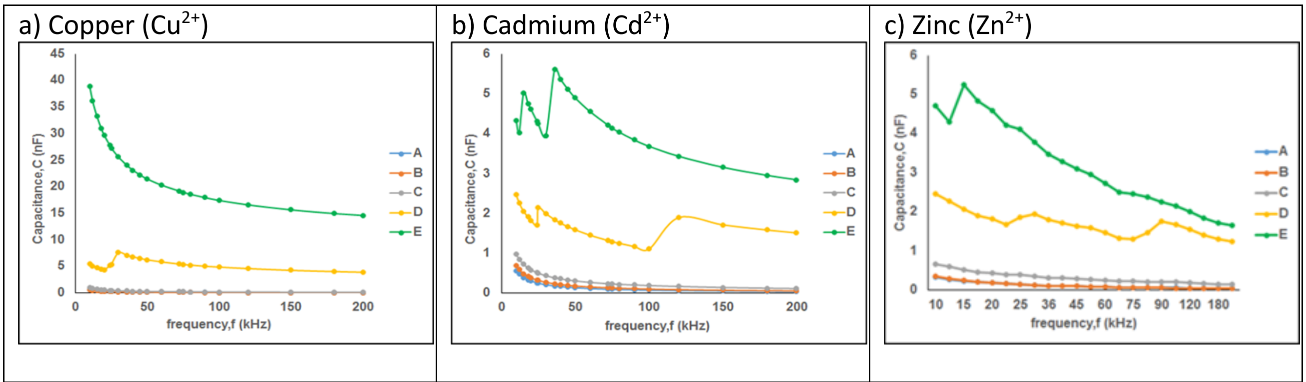
A=1 mg/L; B=2 mg/L; C=10 mg/L; D=30 mg/L; E=50 mg/L.

Fig. 1. Frequency-dependent resistance of chitosan films after adsorption at different concentrations for (a) Cu^{2+}, (b) Cd^{2+}, and (c) Zn^{2+} ions

3.3 Frequency-Dependent Capacitance Response

The capacitance response of chitosan films as a function of frequency and metal ion concentration is shown in Figure 2 (a)-(c). Similar with resistance, capacitance decreased with increasing frequency for all metal ions. Higher capacitance values were recorded at low frequencies, especially for higher metal-ion concentrations. This behavior is associated with increased mobile ion content and stronger interfacial polarization at the film–electrode interface, as commonly reported for chitosan-based metal-ion sensing and electrolyte systems [45-47].

Among the three metal ions, Cu^{2+} produced the highest capacitance values at elevated concentrations, in agreement with its higher adsorption affinity. It can also be seen that films A and B exhibited overlapping capacitance responses for all metal types. The observed trend reflects only small differences that occur in adsorption at low concentrations.



A=1 mg/L; B=2 mg/L; C=10 mg/L; D=30 mg/L; E=50 mg/L.

Fig. 2. Frequency-dependent capacitance of chitosan films after adsorption at different concentrations for (a) Cu^{2+}, (b) Cd^{2+}, and (c) Zn^{2+} ions

3.4 Frequency-Dependent Impedance Response

Figure 3 illustrates the frequency-dependent impedance behavior of chitosan films after metal ion adsorption. Impedance decreased with increasing frequency for all metal ions and concentrations, reflecting the combined effects of resistance and capacitance on the electrical response. Higher impedances at low frequencies are associated with space charge polarization. On the other hand, lower impedance at higher frequencies reflects increased charge mobility [48]. For Zn^{2+} treated films, impedance closely followed the resistance trend, as the capacitive contribution was relatively small. Distinct behavior in films D and E is attributed to inhomogeneous Zn^{2+} ion distribution [49,50].

Cu^{2+} treated films displayed the lowest impedance at higher concentrations, followed by Zn^{2+} and Cd^{2+}, consistent with the adsorption trends observed in AAS measurements. This sequence aligns with the Irving-Williams series, which predicts that the coordination affinity of transition metal cations for the nitrogen and oxygen donor atoms in chitosan peaks at Cu^{2+} [51]. These results indicate that impedance provides an integrated electrical response that reflects adsorption-induced changes in both resistive and capacitive components, making it a robust parameter for sensing applications.

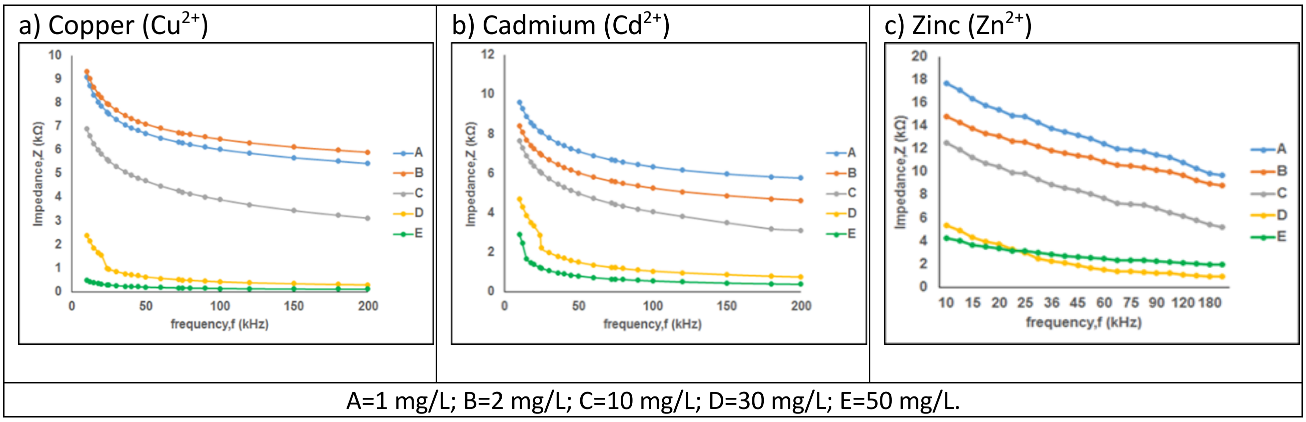


Fig. 3. Frequency-dependent impedance of chitosan films after adsorption at different concentrations for (a) Cu^{2+}, (b) Cd^{2+}, and (c) Zn^{2+} ions

Nyquist plot is a tool for diagnosing charge transport mechanisms in polymer electrolytes. By plotting the imaginary component of impedance (Z”) against the real component (Z’) across a broad frequency range, a Nyquist plot provides mapping for material's resistive and capacitive behavior. In traditional dielectric materials or mixed ionic-electronic conductors, the high-frequency region of a Nyquist plot typically displays a semicircle. However, as portrayed in in Nyquist plots for Cu^{2+}, Cd^{2+}, and Zn^{2+}, no semicircle is observed at high frequency and only an inclined spike appears at low frequency (Figure 4(a–c)). These patterns indicate that the resistive component of the polymer is dominated by rapid ionic mobility and the mobile ions accumulate at blocking electrodes under AC perturbation [52]. The effect is reproducible across various metal ion dopants such as Cu^{2+} [53], Zn^{2+} [41], and adjacent biopolymer systems [54].

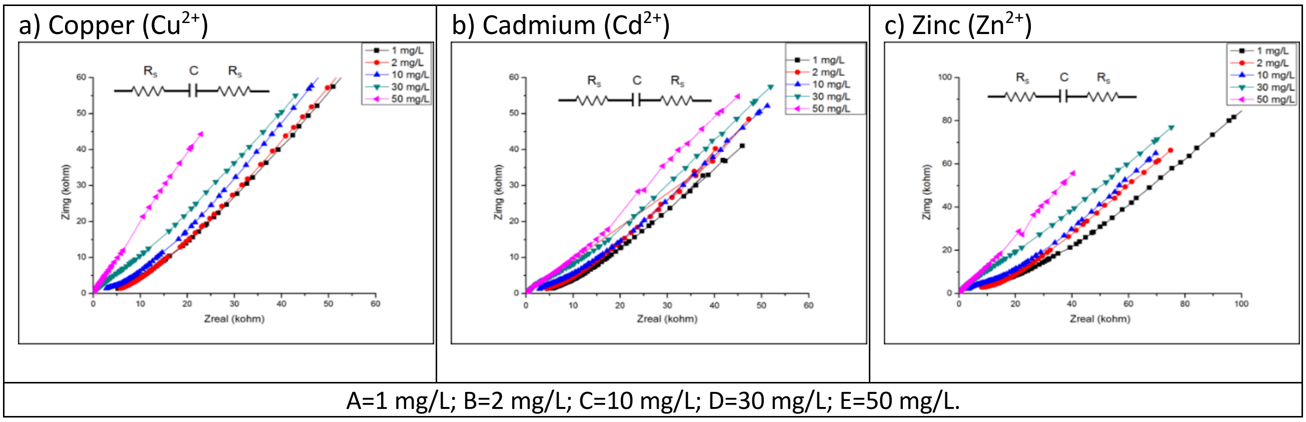


Fig. 4. Nyquist plot for (a) Cu^{2+}, (b) Cd^{2+}, and (c) Zn^{2+} ions

3.5 Correlation between Adsorption and Electrical Response

Figure 5 presents the adsorption behavior of Cu^{2+}, Cd^{2+}, and Zn^{2+} ions on chitosan films measured by AAS at different initial concentrations. At higher concentrations (>30 mg/L), Cu^{2+} exhibited the highest adsorption, followed by Zn^{2+} and Cd^{2+}.

This trend reflects competition among metal ions for a limited number of adsorption sites on the chitosan surface and is governed by differences in ionic potential and affinity. Previous studies have shown that ions with higher ionic potential are more strongly retained by adsorbent materials, leading to preferential adsorption [55] [56]. Consistent with this, chitosan is known to exhibit strong selectivity toward Cu^{2+} ions compared with Zn^{2+} and Cd^{2+} [57]. Even though many adsorption studies support that ions with higher ionic potential are more strongly retained and can outcompete others, especially on acidic or highly charged sites, there are several conditions that can reverse this trend. Factors like hydration, steric effects, and specific surface structure can overridden in ion adsorption selectivity [58] [56]. As such, ionic potential is a strong but not universal predictor of preferential adsorption.

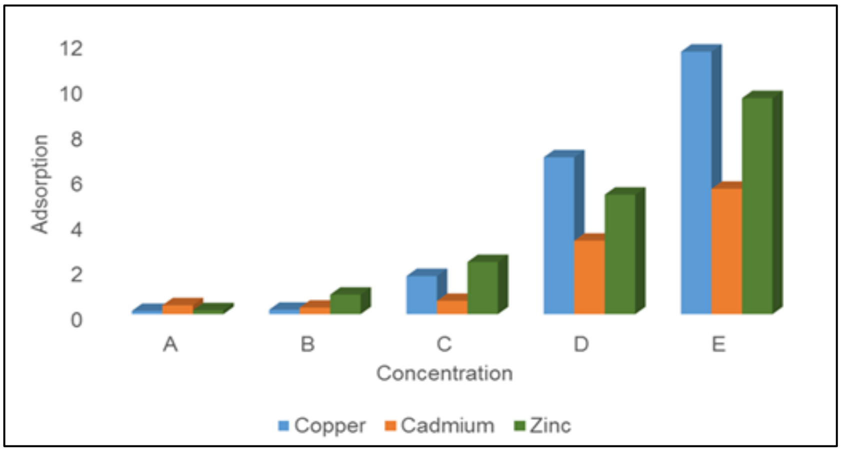


Fig. 5. Comparison amount of adsorption by chitosan at different concentration of the metal ion

The adsorption trends observed by AAS correlate directly with the electrical responses. Higher adsorption levels correspond to higher capacitance and lower resistance and impedance values, while lower adsorption results in reduced capacitance and increased resistance and impedance. At high concentrations, the stronger adsorption of Cu^{2+} leads to the most pronounced electrical response, followed by Cd^{2+} and Zn^{2+}. This finding is consistent with the work of Hu et al. on the adsorption characteristics of Cu(II) and Cd(II) using a magnetite–chitosan composite. Their results similarly indicate that Cu^{2+} exhibits a stronger affinity for chitosan-based sorbents compared to Cd^{2+} [59]. This consistent relationship confirms that adsorption governs the electrical behaviour of the chitosan film.

4. Conclusions

This study demonstrates that adsorption of Cu^{2+}, Cd^{2+}, and Zn^{2+} ions by chitosan films. Results show systematic changes in electrical properties can be exploited for sensing applications. AAS measurements confirmed metal-dependent adsorption behaviour, with Cu^{2+} showing the strongest affinity at higher concentrations, followed by Zn^{2+} and Cd^{2+}. The reversed trends were observed at lower concentrations due to adsorption site availability and solution conditions.

Electrical characterization revealed a consistent relationship between adsorption and electrical response. Increased metal ion uptake resulted in higher capacitance but lower resistance and impedance across the measured frequency range. These trends were observed for all metal ions. Cross-metal comparison at the highest concentration further confirmed that stronger adsorption produced more pronounced electrical responses.

The results show that meaningful sensing information can be obtained without complex impedance modelling, supporting the potential of simple electrical measurements for low-cost heavy metal detection. As such, this work highlights the feasibility of chitosan-based electrical sensing platforms for monitoring metal ion adsorption. It also provides a foundation for developing practical, adsorption-driven sensors for environmental applications.

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