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Research Article

Jovanovic and Sips Isotherm Parameters of Mango Seed Shell Cadmium Ion Sorption from Aqueous Solution

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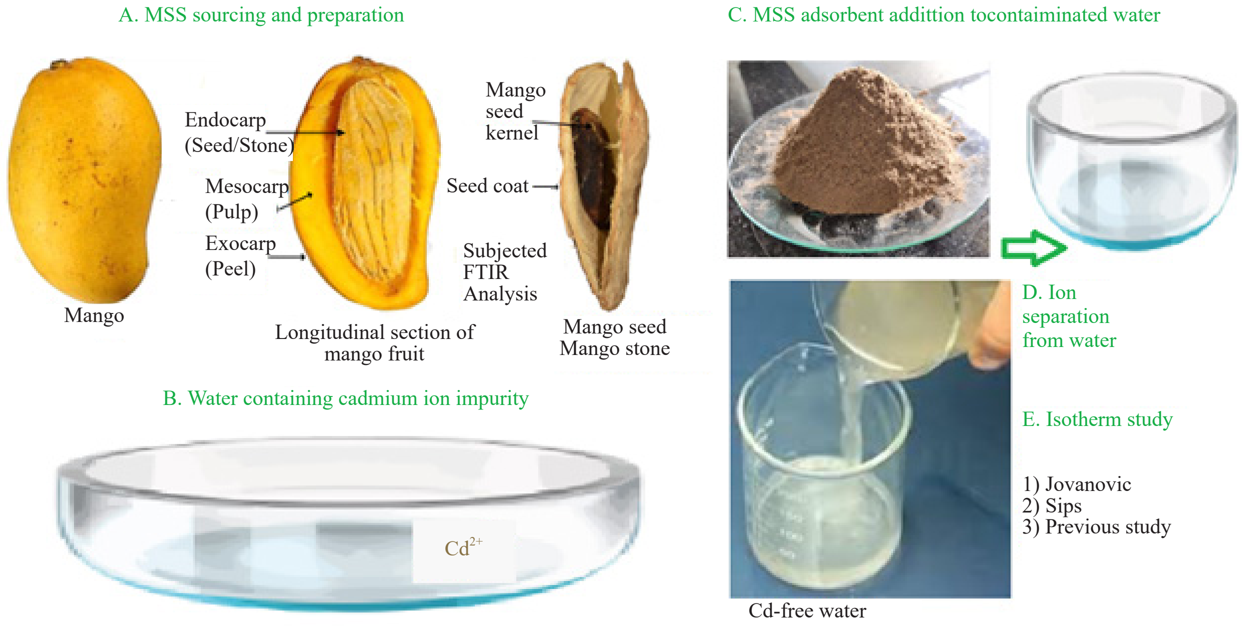
Graphical Abstract:



Abstract: Cadmium (Cd) is not crucial for animal and plant life, and its total elimination from irrigation or drinking water supplies will not deprive water consumers of any beneficial nutrient. Its toxicity to humans has been reiterated,

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and several mechanisms to get rid of it via numerous adsorbents have been researched over several decades, without capturing mango seed shell (MSS) adsorption data performance under the 2-parameter Jovanovic and the 3-parameter Sips isotherm models. The present study tries to address those gaps. As such, the parameter values, $q_{\max} = 97.51$ mg/g and $K_J = -0.0202$ L/mg for Jovanovic, and $K_s = 20.42$ L/g, $\beta_s = 1.16$, and $a_s = -0.046$ L/g for the Sips model, were computed and obtained. After a series of fine-tuning steps to improve model fitting and achieve greater convergence, it led to an R^2 value of 0.7231 and 0.9998, respectively. In addition to error functions, residuals and chi-squares were evaluated, it was found that the Jovanovic equation couldn't describe Cd(II) uptake by MSS, due to its larger return of statistical metric values compared to Sips. Therefore, an existing Cd^{2+}-MSS dataset favouring the Langmuir model as the best was re-evaluated—which then pinpoints the Sips isotherm as the best (in this study) at a low % difference of 0.04-20.59% for predicted vs. observed equilibrium Cd^{2+} uptake (q_e). The adsorption mechanism of Cd(II) onto MSS described by Sips is favorable and efficient adsorption, where the heterogeneous MSS surface has a high affinity for Cd^{2+} at low concentrations. A huge research gap still exists, as Jovanovic and Sips isotherm models are less commonly assessed during adsorption studies. For sustained water purification using MSS as an adsorbent, its abundance must be taken into consideration.

Keywords**:** cadmium, Sip isotherm, mango seed shell, Jovanovic, biosorption isotherm

1. Introduction

Cadmium (Cd, in the form of Cd^{2+} or Cd(II)) can enter water sources from industrial discharges (e.g., metal plating, alloy production, mining, electronics production and battery manufacturing), natural sources (e.g., volcanic activity, forest fires and some geological formations), urban runoff, Cd(II)-containing waste dump (e.g., pesticide, dye, textile, plastic, refining and mining operations, etc.) and agricultural runoff, especially phosphate-based fertilizers containing Cd^{2+}.[1-5] It can be absorbed by different types of aquatic organisms and subsequently enter the human body through biomagnification.[6] Exposure to heavy metals increases the risk of cancer, weakened bones, kidney damage and respiratory issues.[7] It can be removed from aqueous solution through biosorption,[8] solvent extraction, nanofiltration & ultrafiltration,[9,10] reverse osmosis, biological reactors, electrodialysis,[11] biomineralization,[12] membrane separation,[13] electro-sorption,[14,15] evaporation, ion exchange,[16] electro-deposition,[17] Fenton chemical oxidation,[18] chemical precipitation[19,20] and electrocoagulation.[21-24] At the moment, Cd(II) adsorption has been carried out in batch or using packed columns.[25] Biela et al.[26] reported that 0.15-0.2 mg/kg of Cd^{2+} is present in the Earth's crust and the amount supportive to be tolerable for consumption by WHO is 0.005 mg/L (ppm).[27] Usually, to free off synthetic wastewater[3], industrial wastewater, contaminated water, stormwater runoff, human blood plasma,[28] seawater[29] and other aqueous medium of Cd(II), sorption rates by studying their isotherm and kinetics were previously determined.[30] The Jovanovic and Sips biosorption isotherm models are both valuable tools for characterizing the adsorption behavior of pollutants onto adsorbent materials, but they differ in their mathematical formulations, significance, and underlying assumptions. Both models assume homogeneous adsorbent surfaces, monolayer adsorption, and reversible adsorption processes.[31] However, the Sips model allows for deviations from these assumptions, making it more versatile for describing complex adsorption phenomena;[32] whereas Jovanovic refuses to allow mechanical contact between the adsorbent and the adsorbate.[33-35] Because, at high adsorbate concentration and pressure, Sips isotherm predicts Langmuir and at low concentration and pressure, it reduces to Freundlich, as described by N'diaye & Kankou,[36] Gautam et al.[37] and Chilev et al.[38] Considerable amount of Cd(II) by-product removed via sorption can be re-channeled into several applications.

Biosorption is a prominent technique in environmental engineering that is employed in the removal of Cd(II) from different contaminated water sources. Favored by its effectiveness and minimal cost,[39-42] several materials called adsorbent/biosorbent have been employed to sorb Cd^{2+}. Namely, thermophilic and acidophilic algae *Galdieria sulphuraria*,[43] *Anabaena sphaerica*,[44] marine algae,[45] *Hizikia fusiformis*,[46] *Salicornia europaea*,[47] wheat straw/bran,[25,48] natural phosphate,[49] chicken bones,[50] potato peels,[51] *Tridax procumbens*,[52,53] fly ash,[54-56] natural clay,[57-64] nanomaterials/nanosorbents,[60,65-69] spent coffee grounds,[70] zeolite,[71-73] water hyacinth,[74-76] *Escherichia coli*,[77] *Cupriavidus necator*,[78] archaeal cells,[79] *Sargassum fluitans*,[80] chabazite,[81] NTA-silica gel,[82] *Saccharomyces cerevisiae* & *Leuconostoc mesenteroides*,[83,84] immobilized hydrophobic ionic liquids on nano-silica,[85] melon seed husk,[86] banana peels,[87] aluminum hydroxide on cation exchange resin,[88] hydroxyapatite,[89,90] waste shells of golden apple snail,[91] chitosan,[92-97] *Allium cepa*,[28] steel slag,[98] olive mill solid residue,[99] smart dry sludge,[100,101] calcite,[102] modified limestone,[103] silkworm excrement biochar,[104] eggshell,[105] metal organic framework,[106,107] gas,[108] decaying leaves,[109] sidr leaves,[110] cattail leaves,[111] montmorillonite,[112] activated carbon,[113,114] polyethylenimine-grafted gelatin sponge,[115] weathered sand of basalt,[116] pistachio hull waste,[117] pumpkin seed, soybean powder,[118] dragon fruit peel, passion fruit peel, rambutan peel,[119] *Samanea saman*,[120] *Delonix regia*,[121] *Syzygium cumini* seeds extract,[122] *Eucalyptus globulus*,[123] carp scales,[124] *Annona squamosa* shell,[125] agricultural residues bark,[126] *Albizia samanpod*,[127] lignocellulose,[128] bamboo charcoal,[129] sunflower waste,[130] green coconut waste, lemon peel,[131,132] corn residue,[133-135] cocoa pod waste,[136] biochar,[137,138] walnut,[139] avocado seed,[3] orange waste,[140] palm kernel shell,[141] yam,[142] onion skin,[143] and rice residues.[144,145]

Numerous studies employed mango seed shells (MSS) to adsorb diverse contaminants. The adsorption performance of Cd^{2+} detoxification utilizing many agricultural wastes was previously investigated.[146,147] Many other adsorbents notably used to remove Cd(II) were not reported in this study. Apart from Cd(II), MSS or associated waste parts acted as adsorbent in the removal of Fe,[148] Pb,[149-151] Cr,[152-154] Cu,[149,155-157] Zn,[149] Ni,[149] dye[158,159] and oil.[160,161] So far, the removal of Cd^{2+} using mango waste or derivatives were carried out by Parekh et al.,[162] Njuguna[163] and Zhang et al.[164] In those studies, none of the researchers analyze the performance of the MSS adsorbent using Jovanovic or Sips biosorption isotherm models for the removal of Cd^{2+}. Thus, Chu et al.[165] deeply studied the Jovanovic isotherm and pin point previously reported bogus versions. Conflicting versions of the Sips isotherm model is also employed by many researchers. Using appropriate Jovanovic and Sips isotherm models, this study focuses on removing Cd(II) from aqueous solution using MSS. The severity of Cd^{2+} pollutant given its higher solubility in water compared to other toxic metals,[166] necessitates its removal. Additionally, the study comprises of utilizing an existing Cd^{2+}-MSS sorption experimental data to estimate the parameters in Sips and Jovanovic models by nonlinear regression and compare with literature findings. Carrying out error analysis for observed and predicted data is significant for accuracy assessment, model improvement, decision making, uncertainty quantification, quality control, research validity and performance evaluation. As such, this study computes seven error functions for the predictions from both models to gauge the usefulness of the observed and predicted data. The findings will aid in optimizing the MSS sorbent and designing an effective Cd(II) sorption process, and open up avenues for further studies exploring the sorption capabilities of MSS for other heavy metals. Doing so, the study promotes sustainable practices, cost-effective and efficient water treatment, and an ecofriendly approach for waste utilization and environmental remediation.

2. Methodology

2.1 *Experimental adsorption procedure*

The current empirical study utilized fresh MSS endocarp, deionized water, distilled water, and cadmium sulphate stock solution. Equipment such as an oven, mortar and pestle, orbital shaker, atomic absorption spectrophotometer (AAS), and plastic bottles were employed. Fresh mango seeds were sourced from local fruit sellers near Modibbo Adama University (MAU), Yola, Nigeria. The seed shells were removed, washed with distilled and deionized water, and dried in an oven at 70 °C for 48-72 h until a constant weight was achieved. The dried biomaterial was ground and stored in airtight plastic bottles for use as a biosorbent. Stock solutions of Cd(II) were prepared using cadmium sulphate heptahydrate ($CdSO_4 \cdot 7H_2O$) and diluted with distilled water to various concentrations, resulting in a pH of 4.5. Time-dependent biosorption studies were conducted with intervals of 10, 30, 60, and 120 min, using a metal concentration of 5 mg/L, an orbital shaker speed of 200 rpm, and a temperature of 32 °C. Dose-dependent and metal concentration-dependent biosorption studies were also performed, varying biosorbent doses and metal concentrations while maintaining optimal conditions. Cd ion concentrations were determined using AAS, and all experiments were performed in quadruplicate.

Fourier Transform Infrared (FTIR) analysis was also carried out before and after sorption of the Cd^{2+} by MSS at 10 min contact time, to identify and confirm the functional groups present on the surface of the adsorbent material. It is believed that FTIR conducted at shorter contact times may show fewer interactions, with less noticeable spectral changes. Because, prolonged contact time may lead to more thorough adsorption, potentially saturating functional groups on the adsorbent surface. For a meaningful comparison, FTIR was performed at a consistent, representative condition (e.g., the optimal condition identified in the isotherm study: 4.5 pH, 10 min, 5 mg/L & 0.5 g dose) to provide a clearer view of the functional groups involved in the adsorption process.

2.2 *Jovanovic and Sips parameter determination*

Existing experimental mango seed shell adsorption data of Cd(II) sorption from a previous article issued by Luka et al.[167] or the current study author, was used, in continuation of harnessing the suitability of several isotherm models in describing the adsorption behavior. A numerical study by the author establishes the Langmuir model as the most fitting model for the mango seed shell adsorption process. Hence, the Jovanovic and Sips isotherm equations described by Equations 1-2[168,169] and 3-4,[65,170,171] respectively, were used to determine their parameters, in furtherance of finding models that appropriately describe the Cd(II) sorption mechanism by the adsorbent.

$$q_\mathrm{e} = q_\mathrm{max} e^{-K_\mathrm{J} C_\mathrm{e}} \tag{1}$$

$$\ln q_\mathrm{e} = \ln q_\mathrm{max} - K_\mathrm{J} C_\mathrm{e} \tag{2}$$

$$q_\mathrm{e} = \frac{K_\mathrm{s} C_\mathrm{e}^{\beta_\mathrm{s}}}{1 - a_\mathrm{s} C_\mathrm{e}^{\beta_\mathrm{s}}} \tag{3}$$

$$\beta_\mathrm{s} \ln C_\mathrm{e} = -\ln\left(\frac{K_\mathrm{s}}{q_\mathrm{e}}\right) + \ln a_\mathrm{s} \tag{4}$$

Where, q_e = amount of adsorbate in the adsorbent at equilibrium (mg/g), q_max = maximum uptake of adsorbate obtained from the plot of $\ln q_\mathrm{e}$ versus C_e, K_J = Jovanovic constant (L/mg), K_s = Sips isotherm model constant ($\mathrm{Lg^{-1}}$), β_s = Sips isotherm exponent/index describing the homogeneity or heterogeneity of the biosorption process and a_s = Sips model constant ($\mathrm{Lg^{-1}}$). The parameters defined were determined using Origin Pro 2018 via nonlinear regression analysis. In addition, the % difference between the observed and calculated q_e of $\mathrm{Cd^{2+}}$ sorption by MSS, was determined using Equation 5.

$$\%\ \text{Difference} = \left|\frac{q_{\mathrm{e}_{(\mathrm{Expt.})}} - q_{\mathrm{e}_{(\mathrm{Prdct.})}}}{q_{\mathrm{e}_{(\mathrm{Expt.})}}}\right| \times 100 \tag{5}$$

At this point, the predicted responses were determined by substituting the estimated isotherm parameters into their respective models using C_e.

2.3 *Error analysis*

Error functions listed in Ilaboya and Okpoko,[172] Bajic et al.,[124] Nworie et al.,[173] Sampranpiboon et al.,[174] Abdel Hafez et al.,[175] as tabulated in Table 1 (Equation 6-12), were evaluated for the two models q_e data.

Table 1. Selected error function definition

Error function	Mathematical expression	Equation no.
SSE	$\sum_{i=1}^{N}\left(q_{\mathrm{e}_\mathrm{expt}} - q_{\mathrm{e}_\mathrm{prdct}}\right)_i^2$	(6)

Table 1. (cont.)

Error function	Mathematical expression	Equation no.
ARE (%)	$\frac{100}{N}\sum_{i=1}^{N}\left\|\frac{q_{e_{\text{prdct}}}-q_{e_{\text{expt}}}}{q_{e_{\text{expt}}}}\right\|$	(7)
RMSE	$\sqrt{\frac{1}{N-1}\sum_{i=1}^{N}\left(q_{e_{\text{prdct}}}-q_{e_{\text{expt}}}\right)^{2}}$	(8)
HYBRID	$\frac{100}{N-p}\sum_{i=1}^{N}\left[\frac{\left(q_{e_{\text{expt}}}-q_{e_{\text{prdct}}}\right)_{i}^{2}}{q_{e_{\text{expt}}}}\right]_{i}$	(9)
MPSD	$100\times\sqrt{\frac{1}{N-p}\sum_{i=1}^{N}\left[\frac{\left(q_{e_{\text{expt}}}-q_{e_{\text{prdct}}}\right)_{i}^{2}}{q_{e_{\text{expt}}}}\right]_{i}}$	(10)
NSD	$100\times\sqrt{\frac{1}{N-1}\sum_{i=1}^{N}\left[\frac{\left(q_{e_{\text{expt}}}-q_{e_{\text{prdct}}}\right)_{i}^{2}}{q_{e_{\text{expt}}}}\right]_{i}}$	(11)
SEE	$\left[\frac{\sum_{i=1}^{N}\left(q_{e_{\text{prdct}}}-q_{e_{\text{expt}}}\right)^{2}}{N-p}\right]^{0.5}$	(12)

Where, N = total number of data points, p = number of parameters in the model being evaluated, SSE = Sum of squared error, ARE = Average relative error, RMSE = Root mean square error, HYBRID = Hybrid fractional error function, MPSD = Marquardt's percent standard deviation, NSD = Normalized standard deviation and SEE = Standard error of estimate.

2.4 *Analysis of previous findings*

In tabular form, previous findings as regards the obtained result in this isotherm study, were investigated and compared.

3. Results and discussion

3.1 *Predicted Cd(II) uptake*

Except at C_e = 70.92 mg/L where Jovanovic % difference is low (5.35%), lower q_e of Cd^{2+} estimated using the model are very high. Unlike % differences in Jovanovic, Sips isotherm actual and predicted q_e differences, returns very low differences, with approximately 21% as the highest. High % difference infer poor correlation or fit between the empirical and estimated q_e, as shown in Table 2 for Jovanovic.

While it is expected that the low % differences observed in Sips' model prediction is consistent with a better fit. Estimated parameters from Sips isotherm is more likely to describe the sorption of Cd^{2+} using MSS than Jovanovic model.

Table 2. Cd^{2+} Observed and calculated adsorption capacities from sips and Jovanovic models

		Jovanovic		Sips	
	Experiment	Predicted		Predicted	
C_e (mg/L)	q_e (mg/g)	q_e (mg/g)	% Difference	q_e (mg/g)	% Difference
1.12	18.35	99.74087	443.547	22.12898	20.59388
1.88	41.48695	101.2808	144.127	38.7236	6.660775
27.25	302.509	168.9084	44.16418	302.7635	0.084121
70.92	386.6797	407.3788	5.353056	386.5141	0.042821

3.2 *FTIR analysis before and after adsorption*

O-H stretching vibrations typically observed around 3,200-3,500 cm^{-1} are broad peaks indicative of hydroxyl groups' presence. These groups are known for their strong interaction with metal ions, including Cd^{2+}, which suggests potential sites for sorption. Also, in Figure 1a, there is the C=O stretching in carboxyl groups. It is a peak around 1,700-1,750 cm^{-1} associated with the carbonyl (C=O) group in carboxylic acids. The presence of this peak suggests that carboxyl groups may play a significant role in binding Cd^{2+}.[176] Figure 1(a) additionally contains C-O stretching found in the region of 1,000-1,300 cm^{-1}. It corresponds to the stretching vibrations of C-O bonds in alcohols, esters, or ethers, which can also contribute to metal ion sorption. There is also a peak usually present in MSS but missing in Figure 1(a). If C-H stretching in alkanes (typically around 2,850-2,950 cm^{-1}) is missing in the FTIR spectrum, it could indicate structural changes in the MSS after preparation or modification for Cd(II) adsorption, possibly due to the removal of certain organic components. This is in spite of not initiating the sorption experiment yet, which is shown in Figure 1(b).

In comparison, there is a shift in O-H stretching peak before sorption (Figure 1(a)). The broad O-H peak, typically around 3,200-3,500 cm^{-1}, might shift slightly to a lower wavenumber after Cd^{2+} sorption. A shift occurring after sorption (Figure 1(b)) indicates interaction between the hydroxyl groups and Cd^{2+} and suggests the formation of a bond between Cd^{2+} and the O-H groups. Due to the introduction of Cd^{2+}, it might lead to new peaks or an intensification of existing peaks in regions associated with metal-oxygen bonds, typically around 500-700 cm^{-1}. These peaks are indicative of Cd(II) binding to the functional groups on the MSS surface. Specifically, intensification of C=O stretching around 1,700-1,750 cm^{-1} might become more pronounced or shift, indicating stronger interaction with the metal ions. The new or shifted peaks, particularly those related to the O-H or C=O stretching, are crucial as they demonstrate the binding of Cd(II) ions to specific functional groups on the MSS. The presence and intensification of these peaks provide evidence that the adsorption process is successful and that these functional groups are actively participating in the sorption mechanism. Introducing more functional groups such as carboxyl (-COOH), hydroxyl (-OH), or amine (-NH$_2$) groups via chemical treatments (e.g., acid or base modification) can enhance the adsorption capacity by providing more binding sites for Cd^{2+}.

3.3 *Correlation*

As expected, the graphical correlation of the data in Table 2, as shown in Figure 2, that Sips is above Jovanovic's isotherm model in describing the MSS performance. The calculated q_e fits the experimental values in Sips but failed in Jovanovic, based on q_e vs. C_e plot conducted. This poor fit could be due to the oversimplification of the adsorption process, where the assumption of a uniform surface and monolayer adsorption does not hold true for MSS, especially in the presence of Cd^{2+} which may interact with a variety of functional groups on the MSS surface.

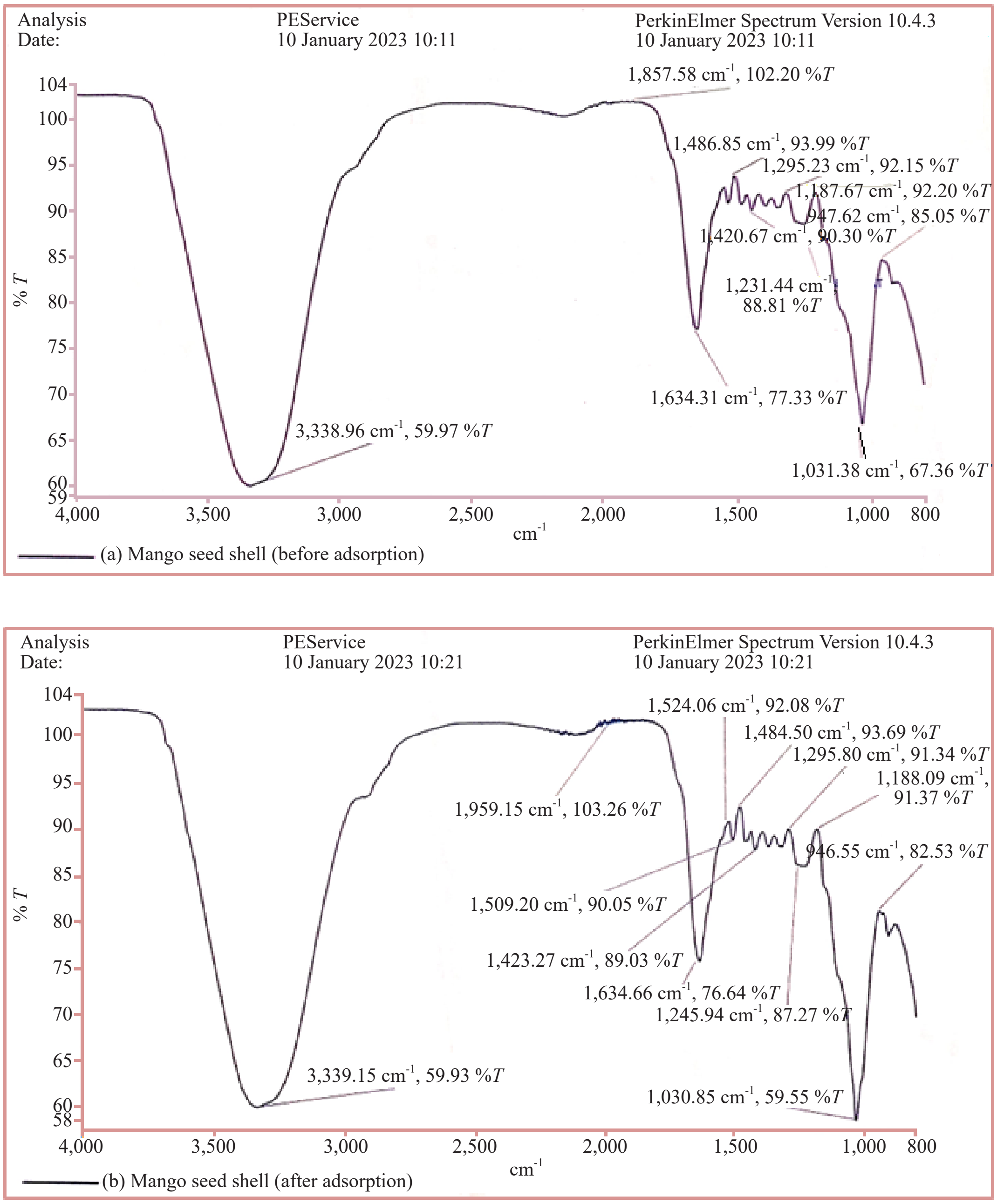

Figure 1. MSS FTIR (a) Pre-sorption and (b) Post-sorption of Cd^{2+}

A q_e vs. C_e plot in Figure 2(a & b) is expected to result in a curve rise to a peak or equilibrium point. Figure 2(b) shows a much better alignment of the data points along the empirical line, reflecting a closer match between the predicted and experimental q_e values. Sips isotherm's ability to account for surface heterogeneity and variable adsorption energies likely makes it a better model for describing the adsorption of Cd^{2+} on MSS. Binding energies for Cd^{2+} on MSS might not be uniform, as the Jovanovic isotherm assumes. The Sips model accommodates this by combining the Langmuir (uniform binding energy assumption) with Freundlich (which accounts for varying affinities) isotherms, leading to a more accurate prediction of q_e across different concentrations. At higher Cd^{2+} concentrations, the assumption of no interaction between adsorbed molecules (central to the Jovanovic model) becomes less valid. However, the Sips isotherm better captures these interactions, especially at higher loadings, leading to a more accurate fit.

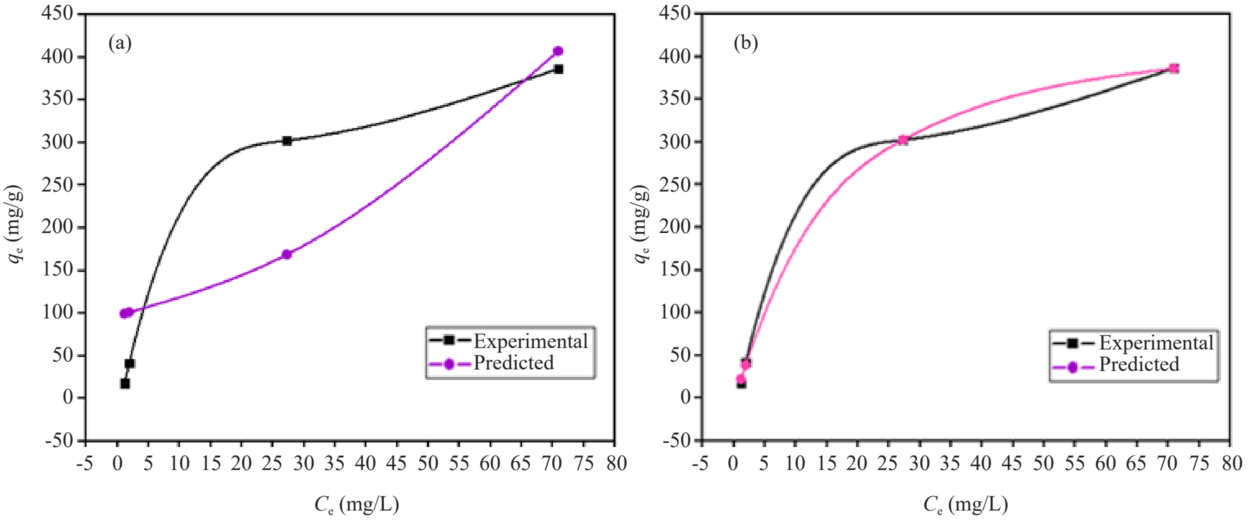

Figure 2. Actual vs. predicted equilibrium Cd(II) uptake by MSS from (a) Jovanovic isotherm and (b) Sips model

3.4 *Isotherm parameter interpretation*

Parameters, q_{max} and K_J, determined after the nonlinear regression using the Jovanovic model and K_s, β_s and a_s from Sips model, are displayed in Table 3. Magnitude and sign of K_J depends on factors such as the properties of the adsorbent and adsorbate, solution chemistry, temperature, and the presence of competing species. In the Jovanovic plot of Figure 2(a), q_e continues to increase as the C_e increases without reaching a flat peak as does the experimental data. It suggests that the adsorption process does not follow a typical Langmuir-type behavior where saturation occurs.[177] Several scenarios may be hindering the process inclination to efficient adsorption, as a result of the negative K_J value (-0.02016 L/mg) obtained in Table 3. It might be multilayer adsorption, heterogeneous surface,[178] chemisorption, competitive adsorption, poor diffusion and/or some kinetic limitations.

The strength of Cd^{2+} interaction with the MSS is described by K_s, which is ideally positive. In this case, the obtained K_s value of 20.41855 L/g suggests a relatively high affinity and efficient adsorption between Cd(II) and MSS adsorbent at lower Cd(II) concentration. In Sips, β_s (ranging from 0 → 1) influence the shape of the adsorption isotherm curve, where a value closer to unity implies a strong and favorable cooperative adsorption behavior as well as a more pronounced curvature in the isotherm.[31,179] Thus, $\beta_s = 1.15715$ obtained herein, suggests a moderate curvature in the isotherm, where multiple layers of Cd^{2+} are being adsorbed on the MSS surface. The negativity of a_s (i.e., -0.04561 L/g) indicates that the adsorption process might experience some unfavorable conditions, possibly due to steric hindrance or repulsive interactions at certain sites on the adsorption surface. Essentially, the parameter accounts for heterogeneity in the MSS surface[38] and provides insights into the distribution of adsorption energies.

Complementary derivations from the isotherm parameters can be found in the fit statistics reported in the same table. The Jovanovic model has a higher Reduced Chi-Squared value (14,238.67603) compared to the Sips model (22.00851), indicating that the Sips model provides a better fit to the experimental data. An R^2 value for the Sips model of 0.99979 is significantly higher than that of the Jovanovic model (0.72307), suggesting that the Sips model explains a larger proportion of the variance in the data. The adjusted R^2 value for the Sips model (0.99936) is also higher than that of the Jovanovic model (0.5846), further supporting the superior fit of the Sips model. Both models converged during the regression process, indicating that the optimization algorithm successfully found parameter values that minimized the difference between the model predictions and the experimental data. Generally, the fit statistics in Table 3 demonstrate that the Sips model provides a more accurate and precise description of the sorption behavior of Cd^{2+} by MSS compared to the Jovanovic model.

Table 3. Fit statistics and isotherm model parameters obtained

	Jovanovic	Sips
Parameter	Value	Value
q_{max} (mg/g)	97.51404	-
K_J (L/mg)	-0.02016	-
K_s (L/g)	-	20.41855
β_s	-	1.15715
a_s (L/g)	-	-0.04561
Statistics		
Number of points	4	4
Degrees of freedom	2	1
Iterations performed	15	9
Reduced Chi-Sqr.	14,238.67603	22.00851
Residual sum of squares (RSS)	28,477.35207	22.00851
R^2	0.72307	0.99979
Adj. R^2	0.5846	0.99936
Fit status	Converged	Converged

3.5 *Statistical error comparison*

Seven error functions were used to calculate their corresponding values for Jovanovic and Sips' data, as shown in Table 4.

Table 4. Calculated errors based on capacities of MSS adsorbents in both models

Error function	Value	
	Jovanovic	Sips
SSE	28,477.37	22.00895
ARE	159.2978	6.845399
RMSE	97.42923	2.708564
HYBRID	25,364.88	96.25835
MPSD	1,592.635	98.11134
NSD	1,300.381	56.64461
SEE	119.326	4.69137

Normally, lower SSE, ARE, RMSE, HYBRID, MPSD, NSD and SEE values point to better fits and consistency in prediction performance. Based on the estimates provided in Table 4, it is evident that the Sips biosorption isotherm model consistently outperforms the Jovanovic model across all error functions. Therefore, the Sips model can be considered the better-performing model for describing the adsorption behavior in this particular study.

3.6 *Current study juxtaposed with previous findings*

Looking at Table 5, only a few studies have investigated the Jovanovic isotherm parameters for the adsorption of Cd(II). Biosorbents used in those studies were nanoclay, sidr leaves, *Cyprinus carpio scale*, *Allium cepa* and coal fly ash.

Table 5. Adsorbents, adsorbates and Jovanovic parameters from previous study

Research	Adsorbate	Adsorbent	q_{max} (mg/g)	K_J (L/mg) or (L/g)
180	Curcumin	Zinc imidazole framework-8	0.8632	0.3105
181	Sudan-IV	Lipophilic activated carbon	214.7	0.025
110	Cd	Sidr leaves powder	56.73	-0.0094
35	Dye in curcumin solution	MSS	5.9316	0.1286
34	L-lysine	Imprinted polymer	0.18588	0.31237
28	Cd	Dried *Allium cepa*	1.6242	0.637
174	Zn	Pulp waste	1.048-1.155	-0.030 to -0.017
124	Cd, Pb & As	*Cyprinus carpio* scale	NR	NR
169	Curcumin solution	Carbon from peanut shell	1.508	0.3331
169	Curcumin solution	Carbon from rice husk	3.502	0.139
169	Curcumin solution	Silica from rice husk	3.294	0.1727
169	Curcumin solution	Tungsten oxide	1.614	0.3755
54	Cd	Coal fly ash	539	0.024
54	Rhodamine B	Coal fly ash	501	0.016
182	Ni	*Pandanus amaryllifolius* stem ash	NR	NR
59	Cd	Nanoclay	17.544-18.143	-0.0672 to -0.0748
183	Mono azo dye methyl orange	Pinecone	297.77	0.025
184	Malachite green	Desert date seed shell	26.59	0.06
173	Methylene blue	Activated rice husk biochar	-93.645	6.917
Current study	Cd	MSS	97.51404	-0.02016

NR-Not reported

Table 6. Adsorbates, adsorbents and Sips parameters from preceding research

Study	Adsorbate	Adsorbent	K_s (L/g)	β_s	a_s (L/g)	q_{max} (mg/g)
95	Cd	Nanochitosan	5.837	0.729	-	2.036
185	Cd	Nanochitosan	0.0412	1.37	-	1.96
94	Victoria blue	Chitosan nanocomposite	0.11-0.30	0.49-0.74	-	601-683
134	Cd	Corn silk	0.534	2.12	-	21.95
186	Synthetic dye	Clay	0.16-1.88	0.36-0.75	-	15.69-31.97
181	Sudan-IV	Lipophilic activated carbon	0.301	0.552	-	205.7
67	Cd	Magnetite nanocomposite	0.016	0.64	-	76.67
179	Cu	Groundnut shell	12.8689	1.25346	0.47347	-
179	Pb	Groundnut shell	6.13959	1.54742	0.24345	-
179	Hg	Groundnut shell	3.7113	1.6536	0.1544	-
100	Cd	Activated sludge immobilized onto chitosan beads	0.039	0.58	-	216
100	Zn	Activated sludge immobilized onto chitosan beads	0.014	0.57	-	188.3
137	Cu	Pinewood biochar	0.46-1.12	1.52-2.71	-	86.2-112
140	Cd	Orange waste	1.3	0.889	-	0.573
140	Zn	Orange waste	2.31	0.575	-	0.453
172	Mn	Acid activated shale	0.8649	0.8862	-	1.7561
172	Pb	Acid activated shale	0.9219	0.9631	-	1.8180
60	Cd	SiO_2-$Mg(OH)_2$ nanocomposite	0.0035-0.0152	1.5806-1.2674	-	121.23-141.49
66	Cd	Poly (acrylamide-co Na acrylate)	7.58×10^{-4}	1.1295	-	1,893.09
65	Cd	Polyamido- amine functionalized silica	29.096	29.0846	NR	-
52	Cd	*Tridax procumbens*	NR	NR	-	NR
93	Cd	Functionalized chitosan	0.171	9.44	-	228.1
187	Cu	Granular activated carbon	394	1.11	-	0.095
187	Pb	Granular activated carbon	164	1.32	-	0.146
187	Zn	Granular activated carbon	792	0.94	-	0.058
92	Cd	Chitosan-based hydrogel	0.5432-1.5050	0.6854-0.9435	-	175.01-234.83
92	Cd	Polyacrylic acid-based hydrogel	0.0622-2.2294	0.7146-0.7277	-	158.26-197.91
71	Pb	Zeolite	0.41	0.7	-	245.75
71	Cd	Zeolite	1.62	2.81	-	4.43
71	Hg	Zeolite	2.23×10^{-4}	1.83	-	0.22
188	Mn	Black carbon from rice straw	0.38	2.09	-	9.56
Current study	Cd	MSS	20.41855	1.15715	-0.04561	-

NR-Not reported

Findings show that apart from MSS adsorbent used in this study to sorb Cd(II) and those previously reported in the literature (Table 5), an extensive investigation is still missing to properly gauge the performance of numerous other sorbents for Cd(II) sorption on the same level. Furthermore, it shows that a negative K_J value is not alien and q_{max} may differ depending on the experimental condition set. If the Jovanovic constant K_J is positive, it would indicate a positive correlation between the adsorbate concentration and q_e. In other words, as the concentration of the adsorbate increases, q_e also increases, which aligns with typical adsorption behavior. Before now, > 80% of studies reported a positive K_J as against this study's findings. Even as the study herein resulted in unfitted Jovanovic predictions, the model is valid for several other adsorbate-adsorbent correlations (apart from Cd^{2+}-MSS or Cd^{2+}-others in Table 5); showing that it is one of the most least employed adsorption isotherm models. Reports show that Nandiyanto et al.[169] used MSS to adsorb dye from curcumin solution, studying its Jovanovic parameters in contrast with the present study.

The Sips model is a 3-parameter isotherm equation that has been extensively used to describe Cd(II) biosorption using different adsorbents, as shown in Table 6. However, only the present study utilizes the model to describe MSS sorption behaviour, based on the extent of the literature search conducted.

A few other heavy metals engaged by researchers in the study of Sips parameters are Cu, Pb, Hg, Zn and Mn. Due to varying versions of the Sips isotherm model employed, the parameters often showcased are K_s, β_s, q_{max} and a_s. There is a need to identify and use the correct Sips model when carrying out such an investigation. There is technically only one Sips isotherm equation, but the interpretation and characteristics of the isotherm can vary based on the value of the exponent. During Cd(II) sorption, $\beta_s > 1$ in polyamido-amine functionalized silica, functionalized chitosan and zeolite utilization (Table 6), will exhibit a convex isotherm and a cooperative multilayer adsorption. But, when β_s is not close to 1, as in activated sludge immobilized onto chitosan beads and magnetite nanocomposite-Cd^{2+} sorption reported in Table 6, it implied a departure from the typical sigmoidal curve shape (concave downwards), where antagonistic adsorption occurred. MSS-Cd sorption herein, is indicative of a favorable adsorption demonstrated by a concave upward sigmoid shape. Compared to Jovanovic, Sips isotherm is more studied in the literature, where it performed absolutely well in most cases.

4. Conclusion

Nonlinear regression analysis was fruitfully run to determine the isotherm parameters of Jovanovic and Sips biosorption of Cd(II) from water using MSS adsorbent. It was discovered that high R^2 of 0.99979, low RSS, low % difference of < 20% and lower error function values realized for the Sips model, indicated a strong correlation between the predicted and experimental data, thereby validating the suitability of the isotherm for describing the adsorption process. With q_{max} = 97.51 mg/g, the Jovanovic model couldn't describe the Cd(II) sorption from aqueous solution using MSS. The favorable adsorption performance of Sips isotherm in this study adds to the Langmuir model previously ascertain to be suitable for the same MSS data by Luka et al.[167] Previous studies analyzed show that the two models could fit Cd^{2+} sorption data using various adsorbents and the MSS biomass employed herein adds to this list. FTIR analysis carried out already rated MSS high as regards the sorption of Cd^{2+}. However, Thermal activation of MSS at controlled temperatures could increase the surface area and pore volume, enhancing the accessibility of adsorption sites for Cd^{2+}. Further investigation and analysis, possibly using more adsorption models or experimental techniques, may be needed to understand the underlying mechanisms driving the observed behavior of Cd(II) sorption using MSS. A host of isotherm models that have not been run for similar adsorbate and adsorbent used in this study are Baudu, Fritz-Schlunder, Weber-Van Vliet, Radke-Pravsniiz and Koble-Carrigan isotherm models.

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Supporting information

This material is available free of charge via the Internet at SCE. The work further advances the previous study entitled: "Isotherm and Batch System Kinetics of Cadmium Ion Sorption Using Mango Seed Shell", published in Research Journal of Chemical Sciences (RJCS). This, and the forgone article are thus, Part One and Part Two. There is possibility, readers will find more versions published after this one, as not all models were exhausted in the two.

Conflicts of interest

The authors declare that there are no conflicts of interest.

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