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Antimicrobial activity of *Mukia maderasapatna* stem extract of jujube seeds activated carbon against gram-positive/gram-negative bacteria and fungi strains: Application in heavy metal removal

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ABSTRACT

Bacterial and fungal infections are the major threat to the human and other living organism. In the present research, the modified jujube seeds have been synthesized and checked its antimicrobial behavior for different types of bacteria and fungi strains. The antimicrobial activities of *Mukia maderasapatna* stem extract from three forms of jujube seeds such as raw jujube seeds (RJS), sulphuric acid assisted jujube seeds (SAJS) and ultrasonic assisted jujube seeds (UAJS) have been investigated. The preliminary phytochemical screening of the stem extracts from *Mukia maderasapatna* was performed by the standard phytochemical methods. The conduction band and functional groups of modified jujube seeds were evaluated by using UV-vis spectrophotometer and Fourier Transform Infrared Spectrometer, respectively. The activated jujube seeds were explored for their antimicrobial activity against multidrug-resistant of two gram-positive (*Staphylococcus aureus, Proteus mirabilis*), three gram negative bacteria (*Escherichia coli, Pseudomonas aeruginosa, Klebsiella pneumoniae*) and two fungal strains (*Aspergillus niger, Candida albicans*). The different zone of inhibition was captured for different forms of jujube seeds which indicates that UAJS has higher zone of inhibition for all clinical pathogens when compared to RJS and SAJS. Additionally, the prepared UAJS has been effectively utilized for the removal of heavy metal ions from aqueous solution. Freundlich model provided the best results for the removal of heavy metal ions by UAJS. Finally, it can be concluded that *Mukia maderasapatna* stem extract of modified jujube seeds has an excellent antimicrobial activities against bacterial and fungal strains.

Keywords: Activated carbon; Adsorption; *Mukia maderasapatna;* Antimicrobial activity; Fungi; Microbes

1. Introduction

In the present scenario, the water contamination is the most important problem for all living organisms. Water source is the most significant raw material on the earth's surface. Nowadays, the surface and ground water were easily contaminated by several harmful microbial pathogens [1–3]. The microbial contamination of water is a public health concern because it causes several toxic health effects to the humans such as aesthetic problems, gastrointestinal, typhoid, dysentery, cholera and other harmful illness [4,5]. The poor water quality causes 3.7 million deaths a year world-wide. World health organization has reported that 80% of the disease was due to the contaminated drinking water [1]. In addition to that, the microbial pathogens have an important influence on the food borne illness. The food borne illness includes are a growing health problem throughout the world which are responsible for generating harmful health effects such as immunological and neuro-

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logical disorders [6–8]. The different microorganisms such as *Staphylococcus aureus* [9], *Shigella spp.* [10], *Proteus mirabilis* [11], *Escherichia coli* [12], *Pseudomonas aeruginosa* [13], *Salmonella spp.* [14], *Klebsiella pneumonia* [15], *Aspergillus niger* [16], *Candida albicans* [17], *Vibrio spp.* [18], *Staphylococcus epidermis* [19], *Fusarium spp.* [20], *Enterobacter spp.* [21] are the major accountable for food borne illness and water contamination. Therefore, in order to remove or inactive the harmful microorganisms, the different disinfection methodologies have been employed. Unfortunately, these methods are impeded by restrictions such as capital and maintenance cost for large scale adoption. So, there is a need to seek alternative technique, hence in economic and environmental factors concern, activated carbon (AC) from agricultural waste biomass plays an important role in antimicrobial study [22]. AC are now considered to be a feasible alternative to antibiotics, it seems to be have a high potential to remove or inactive the harmful pathogens in water and food environment [23]. AC has several advantages to solve the problem of the emergence of microbial multidrug resistance.

In the present study, the AC (UAJS) has some specific characteristics such as large surface area, enhanced porosity, easily availability, low cost, suitable for both acidic and basic environments. Hence, the newly synthesized UAJS has been utilized as an effective adsorbent for the removal of heavy metals from aqueous solution. The main objective of the present research is to prepare the novel material from agricultural waste and evaluates its antimicrobial activity against different bacterial and fungi strains. The newly synthesized AC was extracted by using the solvent of crude stem extract of *M. maderasapatna*. The *M. maderasapatna* stem extraction of AC was characterized by FTIR and UV-vis spectra analyses. The antimicrobial activity was studied by using in vitro against two forms of gram-positive (*Staphylococcus aureus, Proteus mirabilis*), three forms of gram negative bacteria (*Escherichia coli, Pseudomonas aeruginosa, Klebsiella pneumoniae*) and two fungal strains (*Aspergillus niger, Candida albicans*) using well diffusion assay method. The results from these studies reported that AC of jujube seeds has a possible inhibitory activity against different strains. The application of this material was extended to remove the different heavy metal ions such as copper, cadmium and nickel from its aqueous solutions. The different adsorption isotherm models such as Langmuir, Freundlich, Temkin and Dubinin Radushkevich models have been tested.

2. Materials and methods

2.1. Chemical and reagents

The laboratory chemicals such as ethanol, acetone, ethyl acetate and hexane were purchased from Ranbaxy Fine Chemicals Ltd, Mumbai, India. Nutrient agar medium and antibiotic disc (Ampicillin and Streptomycin) was procured from Himedia Laboratories, Mumbai, India. Copper sulphate pentahydrate salt, Cadmium sulfate and Nickel(II) sulfate heptahydrate was procured from E. Merck, India. All the chemicals were used for analytical grade. Fresh deionized water was used during the experimental works.

2.2. Equipment

The pH of the solution was measured by using pH meter (HI 98107; Hanna Equipment Private Limited, Mumbai, India). The ultrasonication process was performed by using sonicator (VCX750, Sonics and Materials Inc, Newtown, USA). The sterilization process was carried out in the Manish autoclave (Manish Scientific Instruments, Chennai, India) equipment. The drying process was carried out by using hot air oven (Hasthas Scientific Instruments India (P) Ltd, Chennai, India). The inoculation and sub-culturing process was done under specified Laminar Air Flow Chamber (Model No. D-0284, Dolphin Instruments and Equipments, Chennai, India). The incubation shaking was performed in Orbital Incubation Shaker (Royal Testing Equipment, Chennai, India). The concentration of the metals ions before and after the adsorption process was measured by using Atomic Absorption Spectrophotometer (AAS, SL176 Model, Elico Limited, Chennai, India).

2.3. Extraction of Mukia maderasapatna (Stem)

The plant material of *M. maderasapatna* (Tamil name: Musumuskkai; English name: Rough bryony) were collected from Kolli Hills, Namakkal District. The collected plant material was compared with voucher specimen (Specimen No. HPRKVK 2013-097) deposited at Herbarium Division of Botany, Presidency College, Chennai, India. The stem of the collected plant material was cut into small pieces and stored in a plastic container to prevent from the decay of some bioactive compounds. The stem was washed with deionized water to remove the dust particles and other impurities. The dried material was kept in a hot air oven at 60°C for about 24 h to remove the moisture content. The dried stem was crushed and grinded into small finer particles. The crushed and powdered stem (100 g) was extracted with methanol (500 mL) for 2 d at room temperature. The collected extract was filtered using Whatman 42 filter paper and then concentrated at 40°C for about 30 min to get the residues which constituted the crude extract. Finally, all the extracts were kept at 4°C for future use.

2.4. Phytochemical analysis

The presence of plant secondary metabolites such as alkaloids, tannins, triterpenes, flavonoids, carbohydrate, polyphenols, proteins, anthraquinones, sterols, coumarins, saponins, fixed oil and fats in the stem extract of *M. maderasapatna* was identified by phytochemical analysis. The screening of the phytochemical analysis was performed according to the common described phytochemical methods.

2.5. Collection and preparation of jujube seeds activated carbon

The agricultural waste biomass of jujube seeds was collected from SSN College of Engineering, Chennai, India. The collected jujube seeds were washed using distilled water to remove the dust particles and other impurities. The washed jujube seeds were placed in the hot air oven at 60°C for about 2 h to remove the moisture content. Then, the dried seeds were grounded and sieved in a particle size of 0.354 mm. This grounded seed powder is called as raw

jujube seeds (RJS). This material was further used for surface modification process by chemical treatment. The prepared RJS was mixed with concentrated sulphuric acid in the ration of 1:3 (one part of RJS with three part by weight of Conc. H_2SO_4) for about 24 h. After this, the mixture was washed thoroughly with distilled water, until the pH of supernatants remains neutral. The washed material was kept in a hot air oven for about 3 h at 80°C. This prepared material is called as sulphuric acid treated jujube seeds (SAJS). This SAJS was further undergoes ultrasonic treatment. 4 g of SAJS was mixed with 50 mL of deionized water and kept in a ultrasonicator with the working frequency of 24 kHz and agitated at the speed of 500 rpm for about 20 min. After that, the mixture was filtered using Whatman 42 filter paper. The filtered slurry was kept in hot air oven for about 4 h at 80°C. This newly synthesized material is called as ultrasonic assisted jujube seeds (UAJS).

2.6. Characterization studies

The characterization of AC was performed by using FTIR analyses. The different functional groups were present on the surface of the RJS, SAJS and UAJS (before and after crude stem extract of *M. maderasapatna*) was identified by FTIR spectrometer (Perkin Elmer FTIR Spectrometer, UK). The samples were scanned at the attenuated range of 400 to 4000 cm^{-1}. The surface plasma resonance band of RJS, SAJS and UAJS (after crude stem extract of *M. maderasapatna*) was characterized by UV-vis spectrophotometer (Jasco V – 630, spectrophotometer, Japan). The absorbance maxima were scanned at the wavelength of 200 to 700 nm.

2.7. Microorganism

In the present study, two gram-positive (*Staphylococcus aureus, Proteus mirabilis*), three gram negative bacteria (*Escherichia coli, Pseudomonas aeruginosa, Klebsiella pneumoniae*) and two fungal strains (*Aspergillus niger, Candida albicans*) were used for the antimicrobial study. These different microorganisms *Staphylococcus aureus* (MTCC 902), *Proteus mirabilis* (MTCC 3310), *Escherichia coli* (MTCC 443), *Pseudomonas aeruginosa* (MTCC 741), *Klebsiella pneumoniae* (MTCC 3040), *Aspergillus niger* (MTCC 872) and *Candida albicans* (MTCC 227) were purchased from Microbial Type Culture Collection, Chandigarh, India.

2.8. Culture media and medium inoculation

500 mL of Nutrient agar medium was prepared by dissolving the required amount of nutrient agar in 500 mL of deionized water. The prepared media was autoclaved (Manish Scientific Instruments, Chennai, India) for about 15 min at 15 psi pressure at 121°C. The sterilized petri plates were poured with 20 mL of nutrient agar medium. In that, 0.1% of inoculum suspension was swabbed uniformly in the petri plates and then inoculum was allowed to drying for about 24 h.

2.9. Antimicrobial study

Agar well diffusion method is the commonly used technique to determine the antimicrobial activity of the prepared material. The well was made by using sterile cork borer in microbial culture grown nutrient agar plates. The *M. maderasapatna* stem extract of RJS, SMJS, UAJS were loaded in a well diffusion of different plates. Ampicillin and streptomycin was used as a control for antibacterial and antifungal activity. The well loaded plates were incubated at 37°C for about 24 h for bacteria and 72 h for fungi. After the incubation period, the diameter of zone of inhibitory activity of the present jujube seeds were measured in mm and compared with control.

2.10. Preparation of metal ion solution

A stock solution of different metal ion solution was prepared by dissolving appropriate amount of salt in 1 L of distilled water. The desired concentration of the working solution was prepared by diluting the stock solution in suitable proportion using distilled water.

2.11. Batch adsorption study

Batch adsorption studies were performed for the removal of metal ions from aqueous solution by using UAJS at optimum conditions. The required concentrations of 100 mL working solutions were taken in the series of the 100 mL conical flasks. In that, the known quantity of UAJS was added to the working solutions. The adsorption process was carried out in a temperature controlled incubation shaker at the constant speed of 80 rpm and at the specified time interval. After the prescribed time interval, the flask was withdrawn from the shaking incubator and this mixture were filtered by using Whatman 42 filter paper. The concentration of the metal ions in the supernatant was measured by using AAS. The percentage removal of metal ions was calculated by using the following formula:

$$\%\ \text{removal of metal ions} = \frac{(C_o - C_e)}{C_o} \times 100 \quad (1)$$

where C_o and C_e are the initial and final metal ion concentration in the solution (mg/L), respectively.

2.12. Isotherm study

The 100 mL of working solution of different metal ion concentration was taken along with an optimum adsorbent dose in a series of 100 mL conical flasks. Batch adsorption studies were conducted at the equilibrium conditions with different metal ion concentration for the purpose of checking the applicability of different adsorption isotherm models. After the system was attained the equilibrium condition, the adsorption mixtures was separated by using Whatman 42 filter paper. The concentration of metal ions in the supernatant solution was measured by using AAS. The quantity of metal ion adsorbed over the adsorbent (UAJS) at equilibrium, $q_{e,}$ (mg/g) was calculated by using the following formula:

$$q_e = \frac{(C_o - C_e)V}{m} \quad (2)$$

where q_e is the amount of metal ions adsorbed per g of adsorbent at an equilibrium (mg/g), V is the volume

of the metal ion solution (V) and m is the mass of the adsorbent (g).

The nonlinear forms of different isotherm models are listed as follows:

Langmuir isotherm model [24]:

$$q_e = \frac{q_m K_L C_e}{1 + (K_L C_e)} \tag{3}$$

Freundlich isotherm model [25]:

$$q_e = K_F C_e^{1/n} \tag{4}$$

Temkin isotherm model [26]:

$$q_e = B \ln(AC_e) \tag{5}$$

Dubinin-Radushkevich isotherm model [27]:

$$q_e = q_{m,D} \exp\left[-\beta\left(RT \ln\left(1 + \frac{1}{C_e}\right)\right)^2\right] \tag{6}$$

where q_m is the Langmuir maximum monolayer adsorption capacity (mg/g), K_L is the Langmuir constant related to the affinity of metal ions to the UAJS (L/mg), K_F is the Freundlich constant [(mg/g)(L/mg)$^{(1/n)}$] relating to the bonding energy, n is the measure of the deviation from the linearity of adsorption (g/L), B is the constant related to the heat of adsorption (B = RT/b), b is the heat of adsorption (J/mol), $q_{m,D}$ is the Dubinin-Radushkevich adsorption capacity, β is the constant related to the adsorption energy, R is the gas constant (8.314 KJ/mol) and T is the temperature (K).

3. Results and discussion

3.1. M. maderasapatna stem extraction

The prepared activated biomass (RJS, SAJS and UAJS, 100 g) were extracted with the solvent of crude stem extract of *M. maderasapatna* (250 mL) for about 48 h using Soxhlet apparatus. The extracts were filtered using Whatman 42 filter paper. The filtered extract was stored in a deep freezer at –7°C. Then, the extracts were finally loaded in a well diffusion of different plates for antimicrobial test.

3.2. Phytochemical analysis of M. maderasapatna stem extract

The potential bioactive compounds in the plant material were identified by qualitative phytochemical analysis method. The qualitative phytochemical analysis of *M. maderasapatna* stem extract showed that the presence and absence of important secondary metabolites (Table 1). The results from Table 1 stated that the different crude extract of screening test confirms the presence of alkaloids, saponins, glycosides, carbohydrates and steroids in *M. maderasapatna* stem extract. These phytochemical compounds were used as drugs and medicine to inactivate the growth of microbes.

3.3. Characterization studies

The characterization of activated carbon before and after the crude stem extract of *M. maderasapatna* was performed using Fourier transform infrared spectrometer and UV-visible spectrophotometer.

3.3.1. FTIR studies

The FTIR study is the most widely used technique to identify the different functional groups on the AC. Figs. 1–3 show the FTIR images of RJS, SAJS and UAJS before and after the crude stem extract of *M. maderasapatna*. The powdered AC material was analyzed in FTIR spectroscopy with the attenuated range of 4000 to 400 cm^{-1}. It can be seen from Table 2, the peak value at 3287.32 cm^{-1} was indicated the presence of Polymeric OH stretch and the peak at 1634.71 cm^{-1} shows the presence of Alkenyl C=C stretch. A strong stretching vibration at 1015.33 cm^{-1} corresponds the presence of aliphatic fluoro compound (C-F stretch). The peak area at 549.94 and 510.20 cm^{-1} shows the presence of aliphatic iodo compound (C-I stretch). After the stem extract of *M. maderasapatna*, the functional groups were present in RJS such as polymeric OH stretch, alkenyl C=C stretch and aliphatic fluoro compound, C-F stretch gets converted into alkyne C-H stretch (3326.86 cm^{-1}), aromatic ethers, aryl-O

Table 1
Preliminary phytochemical screening of *Mukia maderasapatna* stem extract

S. No	Phytochemicals	Different extracts of Mukia maderasapatna				
		Ethanol	Acetone	Ethyl acetate	Hexane	Water
1	Alkaloids	–	+	–	–	+
2	Saponins	+	+	–	–	–
3	Carbohydrates	+	+	+	+	–
4	Phenols/Tannins	–	–	–	–	–
5	Flavonoids	–	–	–	–	+
6	Glycosides	–	+	–	–	–
7	Steroids	+	–	+	+	–
8	Proteins	–	–	–	–	–

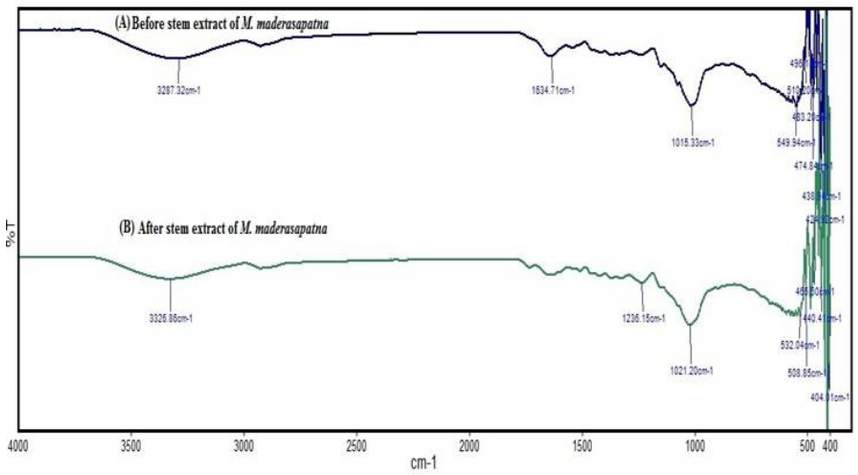

Fig. 1. FTIR images of RJS (A) before stem extract of *M. maderasapatna* (B) after stem extract of *M. maderasapatna.*

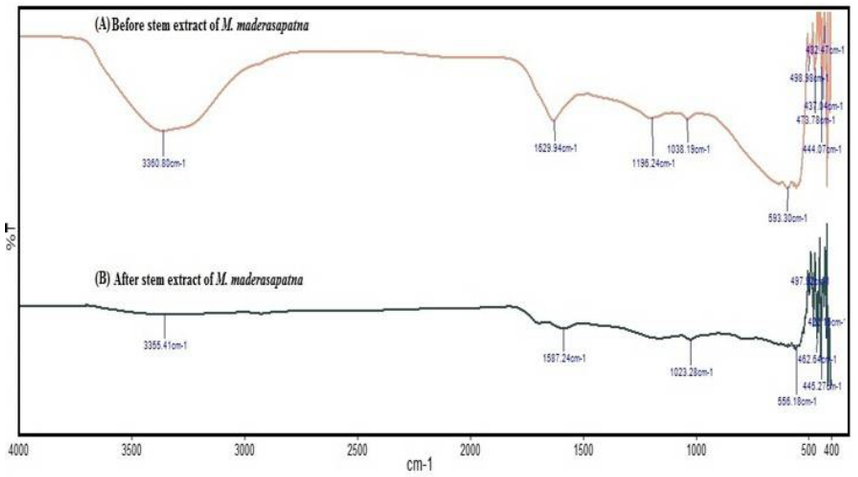

Fig. 2. FTIR images of SAJS (A) before stem extract of *M. maderasapatna* (B) after stem extract of *M. maderasapatna.*

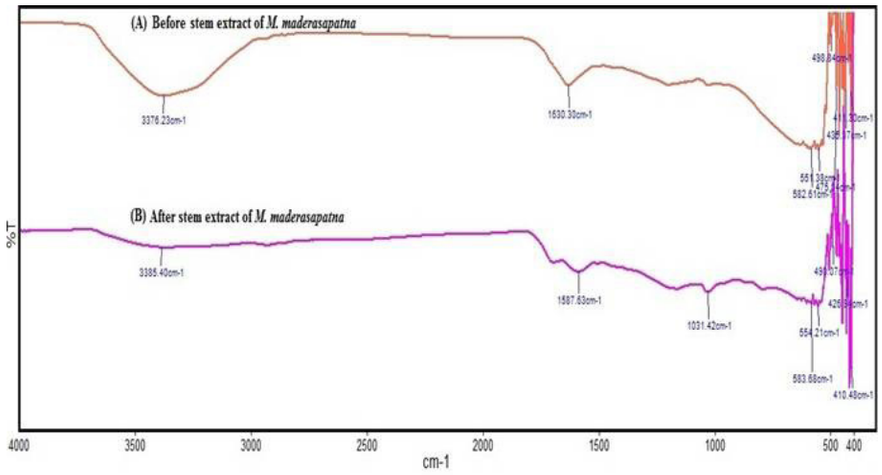

Fig. 3. FTIR images of UAJS (A) before stem extract of *M. maderasapatna* (B) after stem extract of *M. maderasapatna.*

Table 2
FTIR peak values and functional groups present on RJS

S. No	Before stem extract of *M. maderasapatna*		After stem extract of *M. maderasapatna*	
	Peak value cm^{-1}	Functional groups	Peak value cm^{-1}	Functional groups
1	3287.32	Polymeric OH stretch	3326.86	Alkyne C-H stretch
2	1634.71	Alkenyl C=C stretch	1236.15	Aromatic ethers, Aryl-O stretch
3	1015.33	Aliphatic fluoro compound, C-F stretch	1021.20	Aromatic C-H in-plane bend
4	549.94	Aliphatic Iodo compound, C-I stretch	532.04	Aliphatic Iodo compound, C-I stretch
5	510.20		508.85	
6	496.18	Polysulfide S-S stretch	465.50	Polysulfide S-S stretch
7	474.84			

Table 3
FTIR peak values and functional groups present on SAJS

S. No	Before stem extract of *M. maderasapatna*		After stem extract of *M. maderasapatna*	
	Peak value cm^{-1}	Functional groups	Peak value cm^{-1}	Functional groups
1	3350.80	Aliphatic primary amine N-H stretch	3355.41	H-bonded OH stretch
2	1629.94	Aryl substituted C=C stretch	1587.24	Aromatic ring stretch C=C-C[a]
3	1196.24	Terminal amine C-N stretch	1023.28	Aromatic C-H in-plane bend
4	1038.19	Organic siloxane (Si-O-Si)	556.18	Aliphatic Iodo compound, C-I stretch
5	593.30	Alcohol, OH out-of plane bend	497.92	Polysulfide S-S stretch
6	498.98	Polysulfide S-S stretch	462.64	
7	473.78			

Table 4
FTIR peak values and functional groups present on UAJS

S. No	Before stem extract of *M. maderasapatna*		After stem extract of *M. maderasapatna*	
	Peak value cm^{-1}	Functional groups	Peak value cm^{-1}	Functional groups
1	3376.23	Aliphatic secondary amine N-H stretch	3385.40	H-bonded OH stretch
2	1630.30	Aryl substituted C=C stretch	1587.63	Aromatic ring stretch C=C-C[a]
3	582.61	Disulfides (C-S stretch)	583.68	Disulfides (C-S stretch)
4	551.38		554.21	
5	498.84	Aliphatic iodo compounds,C-I stretch	498.07	Aliphatic iodo compounds,C-I stretch

stretch (1236.15 cm^{-1}) and aromatic C-H in-plane bend (1021.20 cm^{-1}), respectively. It is observed from Table 3, the stretch vibrations at 3350.80 cm^{-1} shows the presence of aliphatic primary amine N-H stretch. The peaks at 1629.94, 1196.24 and 1038.19 cm^{-1} was corresponds the presence of aryl substituted C=C stretch, terminal amine C-N stretch and organic siloxane (Si-O-Si), respectively. In SAJS, after the stem extract of *M. maderasapatna*, the functional groups of aliphatic primary amine N-H stretch, aryl substituted C=C stretch, terminal amine C-N stretch and organic siloxane (Si-O-Si) in SAJS were converted into H-bonded OH stretch, aromatic ring stretch C=C-C[a] (1587.24 cm^{-1}), aromatic C-H in-plane bend (1023.28 cm^{-1}) and aliphatic iodo compound, C-I stretch (556.18 cm^{-1}), respectively. It could be seen from Table 4, the peak values at 3376.23 cm^{-1} and 1630.30 cm^{-1} represents the presence of aliphatic secondary amine N-H stretch and aryl substituted C=C stretch, respectively. The strong peaks at 582.61 and 551.38 cm^{-1} was indicated the presence of disulfides (C-S stretch). After the stem extract of *M. maderasapatna*, the functional groups present in UAJS such as aliphatic secondary amine N-H stretch and aryl substituted C=C stretch gets converted into H-bonded OH stretch (3385.40 cm^{-1}) and aromatic ring stretch C=C-C[a] (1587.63 cm^{-1}). Among the different func-

tional groups observed from the FTIR studies, OH group was uniformly present in all the crude stem extract of *M. maderasapatna* of jujube seeds (RJS, SAJS and UAJS). Normally, OH groups has the ability of forming hydrogen bonding capacity (H-bonded OH stretch) which probably indicates that higher potential of jujube seeds towards inhibitory activity against the bacterial and fungal strains. The crude stem extract of *M. maderasapatna* of RJS, SAJS and UAJS suggested that presence of several important functional groups such as alkyne, ether, hydroxyl and aromatic compound which will enhance the antimicrobial characteristics of native and surface modified jujube seeds.

3.3.2. UV-vis spectrophotometer studies

The conduction band of the present AC was analysed by using UV-vis spectrophotometer. The strong absorption band for the RJS, SAJS and UAJS was identified by the spectra analysis and it was showed in the visible region. This specific absorption band is called as surface plasma resonance band (SPR), which is due to the collective dipole oscillations of free electrons. Figs. 4–6 show the UV-vis spectra of *M. maderasapatna* stem extract of RJS, SAJS and UAJS, respectively. The wide intensive band for the *M. maderasapatna* stem extract of RJS, SAJS and UAJS were listed in Table 5. The tabulation report shows that increased absorbance of *M. maderasapatna* stem extract of jujube seeds with different time intervals. The UV-vis spectrophotometer studies stated that an increase in the absorbance peak with time indicated the continuous reduction of analyte in the *M. maderasapatna* stem extract of jujube seeds which means analyte were easily separated from the carbon matrix in the AC while doing the extraction.

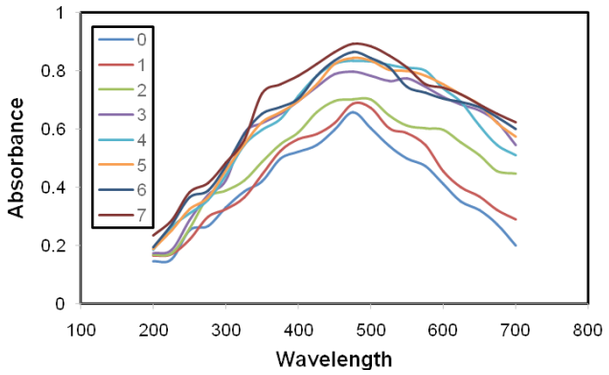

Fig. 4. UV-vis spectra of *M. maderasapatna* stem extract of RJS.

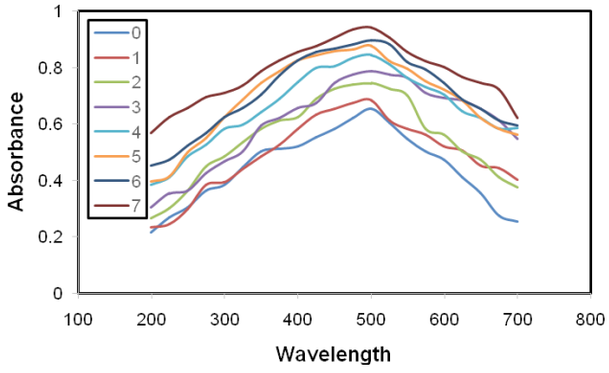

Fig. 5. UV-vis spectra of *M. maderasapatna* stem extract of SAJS.

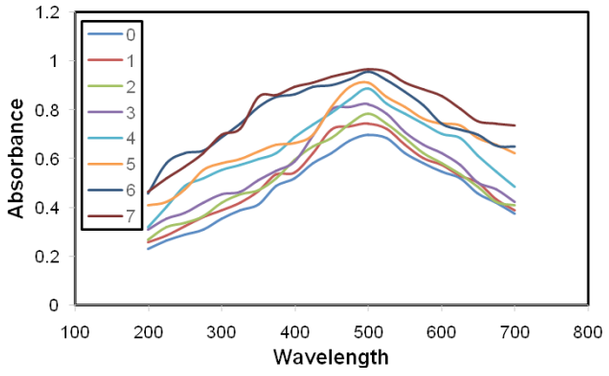

Fig. 6. UV-vis spectra of *M. maderasapatna* stem extract of UAJS.

3.4. Antimicrobial study

In the present study, in vitro antimicrobial activity of *M. maderasapatna* stem extract of raw jujube seeds and surface modified jujube seeds were evaluated. The result of the present studies was shown in Table 6. The experimental results showed that AC has finer inhibitory activity against different bacterial (*Staphylococcus aureus, Proteus mirabilis, Escherichia coli, Pseudomonas aeruginosa, Klebsiella pneumoniae*) and fungal strains (*Aspergillus niger, Candida albicans*). This antimicrobial activity might be due to the various hydrophilic functional groups were present on the AC. The aforementioned functional groups in the FTIR studies were oxygenated due to the tendency of the carbons to oxidize even at room temperature. As a result, these functional groups enable to show antimicrobial activity in an effective manner. Additionally, the *M. maderasapatna* stem extraction was carried out to separate the analyte from the carbon matrix. The complex nature of carbon surface may require solvents to interrupt ionic interactions between the analyte and the carbon. The analyte (chemical constituents) from the AC has superior inhibitory activity against different bacterial and fungal strains.

3.5. Adsorption isotherms

Adsorption isotherm plays a significant role in the adsorption process which can be used to determine the rela-

Table 5
Intensive band of *M. maderasapatna* stem extract of jujube seeds

S. No	Jujube seeds	Wavelength (nm)
1.	Raw jujube seeds	480
2.	Sulphuric acid treated jujube seeds	500
3.	Ultrasonic assisted jujube seeds	510

tionship between the distribution of metal ions in the liquid solution and on the solid adsorbent surface at an equilibrium condition. The adsorption isotherm parameters, correlation coefficient values (R^2) and error values [SSE (sum of squared error), RMSE (root mean squared error)] were calculated by plotting the graph between C_e and q_e [Fig. 7a–c]. The calculated values were listed in Table 7. The best fitted isotherm model was identified based on higher correlation coefficient values and low error values. It can be seen from Table 7, Freundlich isotherm model has higher correlation coefficient (0.9982, 0.9977 and 0.9972 for Cu(II), Cd(II) and Ni(II), respectively) and low error values as compared with other isotherm models. Hence, Freundlich isotherm model was best fitted with the present adsorption system which suggests that adsorption of metal ions [Cu(II), Cd(II) and Ni(II)] onto UAJS is multilayer adsorption and heterogeneous in nature. This indicates the adsorbent surface was made of heterogeneous patches which was favourable for adsorption process. The strength of adsorption in the adsorption process was decided based on n value: (i) if $n > 1$, which indicates that adsorption intensity good for whole range of metal ion concentration (II) if $n < 1$, which indicates that adsorption intensity is good only for higher concentrations but much less for lower concentrations. In the present study, n value was found to be greater than 1 for all the metal ions which indicates that

Table 6
Antimicrobial susceptibility of synthesized activated carbon against bacterial and fungal strains

Organism	Zone of inhibition (mm)		
	RJS	SAJS	UAJS
Staphylococcus aureus	2.4 ± 0.1	2.7 ± 0.16	3.4 ± 0.12
Proteus mirabilis	2.9 ± 0.19	3.3 ± 0.12	3.8 ± 0.10
Escherichia coli	2.0 ± 0.20	3.2 ± 0.25	3.4 ± 0.31
Pseudomonas aeruginosa	3.0 ± 0.20	4.3 ± 0.00	4.0 ± 0.18
Klebsiella pneumoniae	3.3 ± 0.09	4.5 ± 0.00	5.1 ± 0.00
Aspergillus niger	3.2 ± 0.09	3.9 ± 0.19	4.5 ± 0.23
Candida albicans	4.1 ± 0.15	4.6 ± 0.20	5.3 ± 0.30

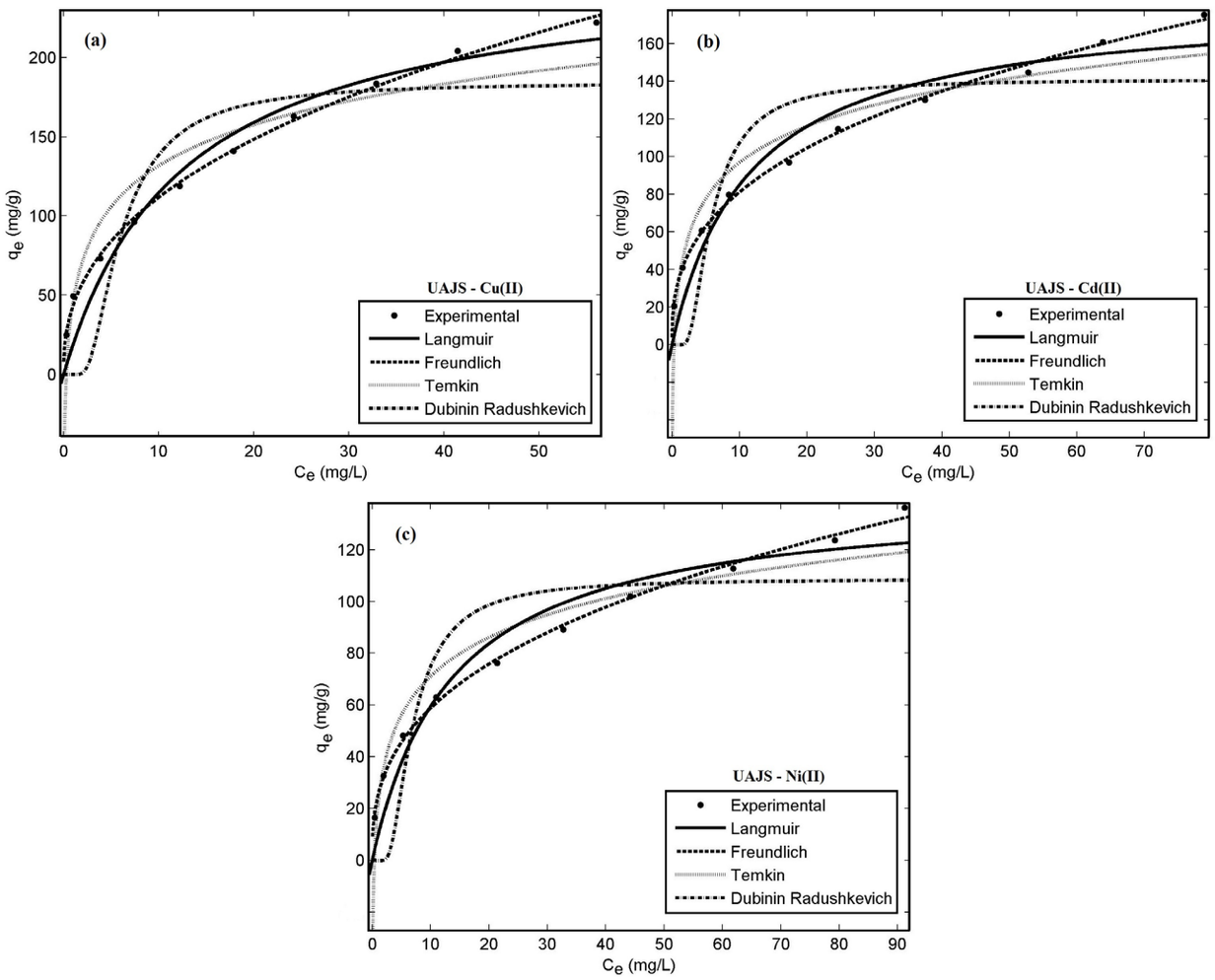

Fig. 7 (a)–(c). The non linear adsorption isotherm for Cu(II), Cd(II) and Ni(II) onto UAJS.

Table 7
Adsorption isotherm fit for the adsorption of metal ions onto UAJS

Isotherm model	Parameters	Cu(II)	Cd(II)	Ni(II)
Langmuir	q_m (mg/g)	259	182.5	140.9
	K_L (L/mg)	0.07955	0.0872	0.07305
	R^2	0.9666	0.9421	0.9359
	SSE	1834	1401	914.5
	RMSE	14.28	13.23	10.69
Freundlich	K_F((mg/g)(L/mg)$^{(1/n)}$)	43.55	34.89	25.27
	n	2.443	2.729	2.726
	R^2	0.9982	0.9977	0.9972
	SSE	99.45	56.79	40.66
	RMSE	3.324	2.664	2.254
Temkin	A (L/mg)	3.425	3.27	2.667
	B	16.18	12.06	9.4
	b (kJ/mol)	0.15	0.21	0.26
	R^2	0.9022	0.9306	0.9325
	SSE	3479	1679	963.1
	RMSE	20.85	14.49	10.97
Dubinin-Radushkevich	$q_{m,D}$ (mg/g)	184.5	141.1	108.7
	β_D ((mol.K/kJ)^2)	1.783	1.688	2.32
	R^2	0.7791	0.7541	0.7368
	SSE	8842	5950	3758
	RMSE	31.34	25.71	20.43

good for whole range of metal ion concentration and highly favourable adsorption process. The maximum monolayer adsorption capacity for Cu(II), Cd(II) and Ni(II) ions was found to be 259, 182.5 and 140.9 mg/g, respectively.

3.6. Sticking probability

In the adsorption process, the capacity of metal ion to remain in adsorbed indeterminately was measured by using sticking probability (S*). Sticking probability of metal ions can be determined by using the following equation

$$S^* = (1-\theta)\exp\left(-\frac{E_a}{RT}\right) \tag{7}$$

where θ represents surface coverage, E_a represents activation energy, R represents gas constant and T as temperature. The sticking probability in the surface area was calculated by the following formula:

$$\theta = 1-\frac{C_e}{C_o} \tag{8}$$

The values of activation energy (E_a) and sticking probability (S*) was shown in Table 8. It can be seen from tabulation report, the values of S* was noted that below 1 for Cu(II) ions upto 250 mg/L, Cd(II) ion upto 200 mg/L and Ni(II) ion upto 100 mg/L of concentration. While increasing the metal ion concentration the sticking probability value has also increased. Hence, the sticking probability of metal ions onto the UAJS are very low at higher concentrations and also negative value of E_a corresponds that adsorption of metal ions onto UAJS was more favourable at lower solution temperature. Therefore, the present adsorption system was exothermic in nature.

Table 8
Sticking probability of Cu(II), Cd(II) and Ni(II) ions onto UAJS

S. No	Conc of metal ion Solution (mg/L)	Sticking Probability (S*)		
		Cu(II)	Cd(II)	Ni(II)
1	50	0.116	0.157	0.272
2	100	0.191	0.374	0.597
3	150	0.459	0.682	1.084
4	200	0.644	0.952	1.651
5	250	0.855	1.581	2.599
6	300	1.028	1.800	3.265
7	350	1.194	2.363	3.792
8	400	1.383	2.938	4.732
9	450	1.556	3.159	5.360
10	500	1.929	3.435	5.408

4. Conclusion

In this research, the novel surface modified adsorbent material (SAJS and UAJS) was prepared and the prepared material has good antimicrobial activities against different types of bacteria and fungi strains. The dual modified agricultural waste biomass was prepared by using sulphuric acid treated followed by ultrasonic-assisted treatment. The characterization studies of SAJS and UAJS were evaluated by using UV-vis spectrophotometer and FTIR. The *M. maderasapatna* stem extract of SAJS and UAJS were explored for their antimicrobial activity against two gram-positive (*Staphylococcus aureus, Proteus mirabilis*), three gram negative bacteria (*Escherichia coli, Pseudomonas aeruginosa, Klebsiella pneumoniae*) and two fungal strains (*Aspergillus niger, Candida albicans*). The results were very promising and well diffusion assay confirming that RJS, SAJS and UAJS materials have finer antimicrobial characteristics on selected clinical pathogens. The newly synthesized UAJS has been tested as an adsorbent material for the effective removal of metal ions from aqueous solution. In the adsorption study, the data obtained from the effect of different metal ion concentration was applied with different adsorption isotherm models. The result showed that present adsorption system was fitted well with Freundlich isotherm model which suggests that adsorption of metal ions onto UAJS is heterogeneous in nature. The sticking probability study reported that the sticking capacities of metal ions onto the UAJS are very low at higher concentrations. The results demonstrated that UAJS has shown the higher zone of inhibition on the selected clinical pathogens when compared to RJS and SAJS and also has good adsorption capacity for the removal of heavy metal ions from aqueous solution.

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