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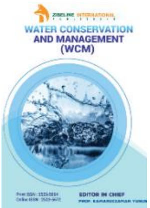

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RESEARCH ARTICLE

REMOVAL OF MANGANESE CATIONS AS A TYPE OF HEAVY METALS FROM CONTAMINATED AQUEOUS SOLUTIONS USING AGRICULTURAL WASTE: WATERMELON RINDS AS A CASE STUDY

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ABSTRACT

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The study investigated the removal of divalent manganese (Mn^{2+}) cations using watermelon rinds, as a low-cost adsorbent, in a batch-type adsorption system under different ranges of the most important operating conditions affecting the adsorption process, which included pH, contact time, initial concentration of divalent manganese ions (Mn^{2+}), agitation rate, temperature, and adsorbent dosage. The operating factor values varied between 1-10, 5-180 minutes, 1-40 ppm, 100-500 rpm, 25-50°C, and 0.5-7 g, respectively. The results obtained indicated that a dose of 6.5 g of Iraqi virgin watermelon rinds successfully treated solutions contaminated with divalent manganese (Mn^{2+}) cations by 81.4% in 150 minutes, at the highest temperature, with an agitation speed of 400 rpm, and a pH of 8.

KEYWORDS

Agricultural waste, Batch adsorption, Manganese Cations, Removal, Watermelon Rinds.

1. INTRODUCTION

Adsorption is defined as a basic physicochemical phenomenon that occurs at the interface between two phases, one solid and the other fluid (either liquid or gas), where particles (atoms, ions, molecules) from the fluid phase accumulate on the surface of the solid phase, as a result of electrostatic attraction forces, weak physical bonds, or chemical interaction between the adsorbent surface particles and the adsorbed particles (Hasan et al., 2025). Adsorption depends on fundamental properties of the adsorbent material such as surface area, functional groups, energy distribution, porosity, and permeability (Alsarayreh et al., 2025b). It is also affected by several operational factors such as pH, concentration, temperature, zero charge point, and process time (Alalwan et al., 2021). In adsorption technology, the set of dynamic and sequential steps that particles follow when transitioning from the liquid phase to the solid phase is referred to as the adsorption mechanism (Abbas and Ibrahim, 2020). This mechanism generally involves three steps, the first is the movement of particles within the gaseous or liquid phase until they are close to the solid surface; this step is called mass transfer (Alwan et al., 2021). This step is subject to several factors, including diffusion rate, solution viscosity, substance concentration, and fluid kinetics (Alsarayreh et al., 2024). The particles then move to the outer surface of the adsorption medium, having passed through the film boundary layer and overcome the forces resisting their advance (Al-Hermizy et al., 2025). In this step, the actual bonding between the adsorbent and adsorbate particles occurs at the active sites within the large and medium-sized pores. The diffusion step within the pores is often the slowest in the adsorption mechanism (Alhamd et al., 2024a). While the final step is diffusion within the microporous particles, where the structure of the adsorbed material plays a crucial role in determining the speed and efficiency of the process (Alalwan et al., 2018). This step may be accompanied by reactions such as ion exchange or the formation of complexes and precipitates, as well as the possibility of secondary rearrangement of the adsorbed particles on the surface (Abbas et al., 2021). Adsorption can be classified, according to the interaction forces controlling the bond between the adsorbent and adsorbed particles, into two main types: physical adsorption and chemical adsorption (Hasan et al., 2021). Physical adsorption can be defined as the adsorption resulting from weak physical bonding forces such as van der Waals forces and hydrogen bonds (Khaleel et al., 2022). This type is characterized by being reversible, occurring rapidly, and often multilayered, where particles can be arranged in layers on top of each other (Mahmood et al., 2025). Furthermore, this type does not require high activation energy; therefore, the adsorption enthalpy does not exceed 40 kJ/mol (Hameed and Abbas, 2024). This type of adsorption is clearly affected by changes in concentration, applied pressure, and system temperature (Alsarayreh et al., 2024). Chemosorption, on the other hand, results from the formation of stronger and more specific bonds, such as strong chemical bonds resembling covalent or ionic bonds, between the adsorbent and specific active sites on the surface of the adsorbent (Akhtar et al., 2024). It is characterized as irreversible because it requires a large amount of energy to reverse, therefore it occurs more slowly than the previous type (Agboola and Benson, 2021). This type requires high activation energy, as the adsorption enthalpy is higher than 80 kJ/mol. It is often monolayer because each adsorbed particle occupies a specific site, and it is selective towards specific types of pollutants (Alsarayreh et al.,

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2025a). In addition to the two types mentioned above, there are other forms such as ion exchange, where an adsorbed particle is replaced by a particle bound to the adsorption surface; electrostatic adsorption, which occurs as a result of the effect of opposite charges; and coordinate adsorption, where metal ions are formed that are capable of forming complexes with functional groups on the surface of the adsorbent medium (Alsharif, 2025). Furthermore, a distinction can be made between single-layer and multi-layer adsorption, and between adsorption on homogeneous and heterogeneous surfaces (Tenea et al., 2024). One of the most widely used adsorbents is activated carbon, the most popular adsorbent among all other materials (Maddodi et al., 2020). It is known for its high ability to adsorb various types of materials under a wide range of operating conditions due to its unique characteristics, such as its diverse functional groups, very high surface area, and advanced porous structure (Scholz, 2023). Zeolite is also considered a well-known adsorption material (Khudair et al., 2024). It is a composition of aluminum silicates characterized by its unique crystalline structure, which has channels and pores of a specific molecular size. Therefore, it is also known as (molecular sieves) (Zulfiqar et al., 2025). It has long been used in material separation processes and the efficient treatment of contaminated media (water and air), and at the same time it can be used as a catalyst in many petroleum processes and chemical applications (Mancinelli and Martucci, 2025). One of the most well-known and popular absorbent materials is silica gel, a porous form of silicon dioxide, known for its high ability to absorb moisture (drying) and used in product preservation and packaging (Chen and Lo, 2022). In addition to its use in drying gases and purifying hydrocarbons, alumina or aluminum oxide is also used as a catalyst in many important chemical reactions (Shadhan et al., 2024). Clays such as bentonite have also been used as effective adsorption media, particularly in water treatment, due to their availability and reasonable cost (Reyes-Bozo et al., 2025). In the last two decades, other materials have emerged that have been used as highly efficient adsorbents, such as nano-oxides like iron oxide and titanium oxide, metal-organic frameworks (MOFs), and others (Abbas et al., 2020). Despite the efficiency of these materials and their ability to treat many pollutants, especially in aquatic environments, their use faces many challenges and obstacles. The most prominent of these limitations is their high production cost and exorbitant price, especially for activated carbon, advanced nanomaterials, and metal-organic frameworks (MOFs) (Kidanemariam and Cho, 2025). Producing these materials requires specialized manufacturing techniques that are costly, resulting in their high price. This makes their use economically limited and unsuitable, restricting their application in large-scale settings (Alhamadani et al., 2026). Furthermore, these materials require continuous regeneration, as restoring the adsorbent capacity after saturation requires costly and energy-intensive chemical or thermal processes (such as solvent washing or burning) and often results in a loss of part of the material's mass as well as a gradual decrease in its efficiency after each regeneration cycle (Alminshid et al., 2021). On the other hand, the byproducts of the recycling process itself may be liquid or gaseous waste that requires treatment (Ali et al., 2023). This material is characterized by its selectivity towards certain pollutants, and while this is an advantage, it has another drawback that becomes evident in highly polluted environments: it may adsorb beneficial substances or fail to adsorb harmful substances, which then remain in the medium untreated (Alminshid et al., 2025). Adsorption technology has attracted the attention of many environmental researchers due to its numerous advantages, including its simple design, ease of operation, lack of need for complex equipment or large spaces, low energy consumption, and remarkable ability to treat many pollutants (Ali et al., 2021). Adsorption has the ability to treat polluted environments even at extreme concentrations, and is therefore a promising and sustainable solution, especially if suitable alternatives are used for materials that face challenges and limitations in operation (Abbas and Abbas, 2013a). The most prominent alternatives that have attracted specialists are agricultural and industrial waste, as its use offers numerous benefits. This waste is considered one of the most significant contemporary environmental challenges due to the substantial expansion of agricultural and industrial activities (Ali et al., 2020a). The accumulation of these wastes or their disposal in unplanned ways leads to multiple environmental problems, including air, water, and soil pollution (Abbas et al., 2019a). But converting them into useful materials will make them a sustainable economic and environmental resource instead of disposing of them, thus reducing their negative environmental impacts and transforming them into materials of added value. Among the most prominent waste materials investigated for their suitability in water treatment are sawdust, pomegranate peels, lemon peels, tangerine peels, almond shells, watermelon rinds, rice husks, mandarin peels, spent tea leaves, mango peels, banana peels, orange peels, plastic waste, aluminum foil, sunflower seed husks, pistachio shells, pineapple peels, and tree leaves such as *Eucalyptus*, buckthorn, tangerine, alga, Water Hyacinth (*Eichhornia Crassipes*), and others, eggshell (Alhamadani et al., 2026; Ali et al., 2024a; Al-Hermizy et al., 2022; Gadooa et al., 2025; Hammed et al., 2025; Abbas and Nussrat, 2020; Abbas and Abbas, 2013b; Alhamd et al., 2024b; Al-Ali et al., 2023; Abed et al., 2025a; Abdullah et al., 2023; Hasan et al., 2021; Abed et al., 2025b; Ghulam et al., 2020; Abdulkareem et al., 2023; Ali et al., 2025; Ibrahim et al., 2025a; Ali et al., 2020b; Alhamadani et al., 2025; Ibrahim et al., 2025b; Mahmood et al., 2025; Abbas et al., 2019b; Hashem et al., 2021). Due to their unique properties, such as their suitable structure, surface area, and multiple functional groups, these materials have proven their ability to remove many pollutants, including dyes, pesticides, heavy metals, hardness, acids, inorganic toxins, pharmaceutical residues, sulfur, and medications (Hameed and Abbas, 2024; Abd Al-Latif et al., 2023; Abbas and Abbas, 2013c; Ibrahim et al., 2021; Abbas and Abbas, 2014; Alalwan et al., 2020; Ibrahim et al., 2020a; Abbas and Alalwan, 2019; Ibrahim et al., 2020b). Although these materials accumulate in large quantities after the treatment process is complete, the application of the zero-residue level concept has provided a sustainable solution to this problem (Rajaa et al., 2023). Toxic residues can be converted into useful materials for several applications, including their use as rodenticides for mice) and rabbits, conversion into environmentally friendly organic fertilizer, use as an additive for reinforcement the concrete mixture, or as a raw material in the preparation of acetone or promoted ethanol (Abd ali et al., 2018; Abd Ali et al., 2024; Abbas, 2015; Abbas et al., 2022a; Abbas et al., 2022b; Hamdi, 2024). Watermelon rind is considered an important type of agricultural waste due to its huge amount produced in different countries in the world. Each fruit of watermelon dispose between 10-20% of its weight as a waste rind. Thus, there is a very big amount of rinds waste that must be used in an eco-friendly way. Table 1 show the annual production of watermelon rinds from 2000-2025.

Table 1: Annual Global Production of Watermelon Harvest

Year	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**	**2006**	**2007**
Production (Million Tons)	76.7	78.2	80.9	82.0	84.1	87.3	89.8	91.1
Year	**2008**	**2009**	**2010**	**2011**	**2012**	**2013**	**2014**	**2015**
Production (Million Tons)	92.2	93.0	93.5	95.1	96.7	97.8	98.9	100.5
Year	**2016**	**2017**	**2018**	**2019**	**2020**	**2021**	**2022**	**2023**
Production (Million Tons)	100.7	101.2	101.0	101.84	101.92	101.64	99.96	104.93

Therefore, and because the literature lacks studies specifically targeting it compared to other similar minerals, the current research aims to investigate the performance of watermelon rinds as a low-cost adsorbent for recovering one of the most important heavy metals, manganese, in a batch-type adsorption unit. It also aims to determine the optimal operating conditions, including pH, contact time, initial concentration, agitation speed, temperature, and adsorption dose, to achieve the highest efficiency of treatment.

2. Experimental Procedure

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This part of the research covers all the details related to the experimental procedure of the study, from the materials used to explaining how the practical part is carried out in terms of equipment, operating conditions and their ranges, in addition to explaining the method of measuring heavy metals automatically along with the method of preparing the calibration curve.

2.1 Stock solution

To prepare manganese stock solution, 5.35 g of 98% purity manganese (II) nitrate hexahydrate of chemical formula ($Mn(NO_3)_2 \cdot 6H_2O$) supplied from Sigma-Aldrich, Germany, was dissolved in a sufficient quantity of double-distilled water. After that the solution was carefully transferred to a clean and sterile 1000 ml glass volumetric flask, and then the volume was completed to a 1-liter solution. The stock solution prepared according to the above method had a concentration of 1000 ppm of divalent manganese (Mn^{2+}) cations and stored in a dry, dark place until use.

2.2 Calibration curve

According to the method described in (Ali et al., 2020c), the divalent manganese (Mn^{+2}) cations in the solutions after treatment were measured using an AAS apparatus, and a calibration curve was prepared by measuring the absorbance of concentrations at a wavelength of 403.1 nm. Each reading was measured triplicate to conduct the accuracy, and the average values were recorded. By plotting the relationship between the measured concentrations of divalent manganese (Mn^{+2}) cations on the x-axis and the corresponding absorbance on the y-axis, the calibration curve required to detect the concentration of the target (Mn^{2+}) cations in the treated solutions was performed. Figure 1 shows the calibration curve used in the current study.

2.3 Adsorbent medium

Iraqi watermelon rinds were collected from grocery stores selling vegetables and agricultural products in Baghdad. After peeling, the peels were washed several times with tap water until clean, and then rinsed with double-distilled water. The clean peels were dried by spreading them in an open air then drying using an electric oven at a temperature of 45°C until the weight is constant. The dried peels were ground using a laboratory grinder and the powder passing through 841 μm sieve was selected. Iraqi virgin watermelon rind powder was stored in a cool, dry place at ≤ 4°C until use. Figure 2 shows the watermelon rinds used in this study.

2.4 Adsorption unit

To determine the performance of Iraqi virgin watermelon rinds as a low-cost adsorbent for recovering divalent manganese (Mn^{2+}) cations, a batch-type adsorption unit was employed. The first step was adjusting the acidity, shaking speed, temperature, and initial concentration of adsorbate. A determined amount of adsorbent, i.e. Iraqi virgin watermelon rind powder, is mixed with the polluted solution in the experimental container. The experimental parameters investigated were, temperature (25-50°C), contact time (5-180 minutes), dose of Iraqi virgin watermelon rinds (0.5-7 g), initial concentration of divalent manganese (Mn^{2+}) cations (1-40 ppm), agitation speed (100-500 rpm), and pH (1-10). The unit continues to operate until it automatically shuts down.

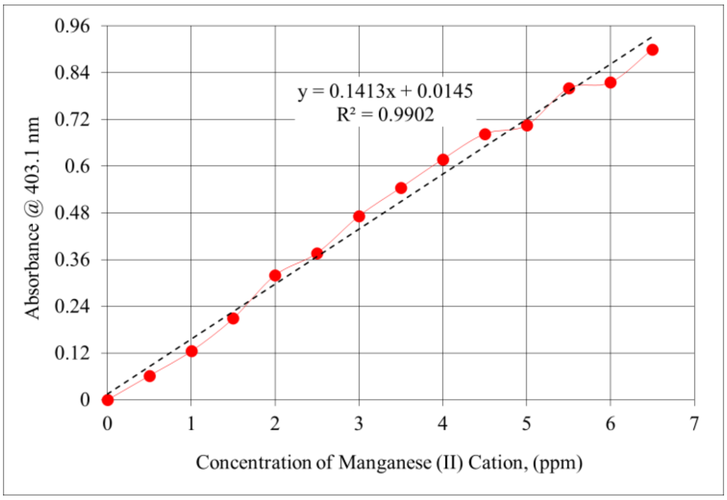

Figure 1: AAS Calibration Curve of Divalent Manganese Cation used in this Study

Figure 2: Iraqi Watermelon Rinds used in this Study

The experimental flaks were then painstakingly extracted and filtered using filter paper. The residual concentration of divalent manganese (Mn^{+2}) cations is detected by the AAS device, and calculated through equations (1) and (2) (Gadooa et al., 2025):

$$\%R = 1 - \frac{C_{out}}{C_{in}} \tag{1}$$

$$q = \frac{V}{1000}\frac{(C_{in} - C_{out})}{w} \tag{2}$$

Where: $\%R$: Efficiency of divalent manganese (Mn^{2+}) cations recovery during the process (-); C_{in}, C_{out}: Initial and final concentrations of divalent manganese (Mn^{2+}) cations before and after treatment, (ppm); q: Adsorption capacity of Iraqi virgin watermelon rinds, (mg/g); V: Volume of aqueous solution utilized in the experiment (100 ml); w: weight of adsorbent added to the experimental flask (g).

3. RESULTS AND DISCUSSION:

3.1 Influence of pH factor

The results obtained from varying pH, as shown in Figure 3, indicate the percentage removal increases from 2.476% to 22.339% with increasing pH between 1-8, then constant at upper values. It is observed that the percentage removal is low at high acid values. As a result, the hydrostatic repulsion between the surface of the watermelon rinds and the divalent manganese cations increases, and consequently, the available active sites for trapping the target ions are insufficient, resulting in minimal removal efficiency (Alsarayreh et al., 2024). Thus, increasing their binding to the functional groups responsible for adsorption, and consequently, the removal efficiency begins to rise between pH 2 and 8. After exceeding a pH value of 8, the removal efficiency remains almost constant and is close to 22.339%. The adsorption mechanism within this pH range has shifted from electrostatic forces to physical adsorption or stable surface coordination (Alhamd et al., 2024a).

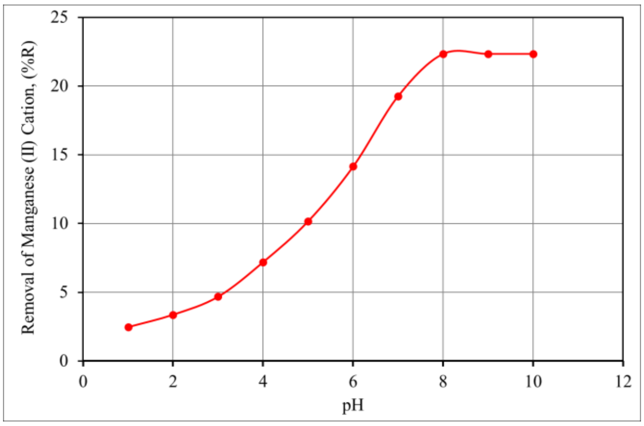

Figure 3: Effect of pH on the removal of divalent manganese (Mn^{+2}) cations using Iraqi watermelon rinds @250 rpm, 1g, 1 ppm, 50°C

3.2 Agitation speed

The results obtained from varying agitation speed, as shown in Figure 4, indicate the percentage removal increases from 4.276% to 63.956% with increasing agitation speed from 100-400 rpm, respectively. This improvement in efficiency can be explained by the fact that increasing the agitation speed of the adsorption unit leads to improved mixing and contact efficiency between the contaminating solution and the adsorbent in the experimental flask (Abdullah et al., 2023). After exceeding the agitation speed of 400 rpm, it is observed from Figure 4 that the removal efficiency remains constant and unchanged. This result can be attributed to the dynamic equilibrium between the adsorbent and adsorbate material, such that the number of adsorbed ions equals the number of ions released from the surface of medium (Abbas et al., 2019b).

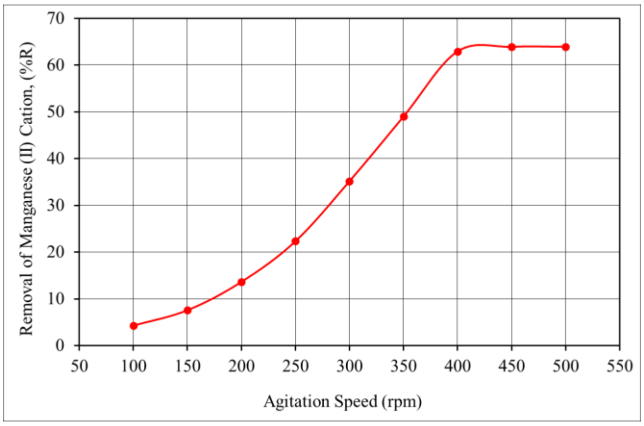

Figure 4: Effect of agitation speed on the removal of divalent manganese (Mn^{+2}) cations using Iraqi watermelon rinds @50°C, 1 g, 8, 180 mins, and 1 ppm

3.3 Initial concentration

The results obtained from varying initial concentration, as shown in Figure 5, show that the relationship between percentage removal and initial concentration decreases from 63.956% to 7.431% as the initial concentration of divalent manganese (Mn^{2+}) cations increases from 1 to 38 ppm, respectively. This decrease is attributed to the fixed number of active sites in the adsorbent material, which is able to accommodate a specific quantity of the target (Mn^{2+}) cations. If the initial concentration is low, the adsorbent will be able to efficiently retain a greater number of divalent manganese cations (Alhamadani et al., 2025). At the same time, Iraqi virgin watermelon rinds can only adsorb a limited number of (Mn^{2+}) cations, and this makes the number of unadsorbed (Mn^{2+}) cations greater as the initial concentration increases, and thus the treatment efficiency decreases according to equation (1) (Hameed and Abbas, 2024).

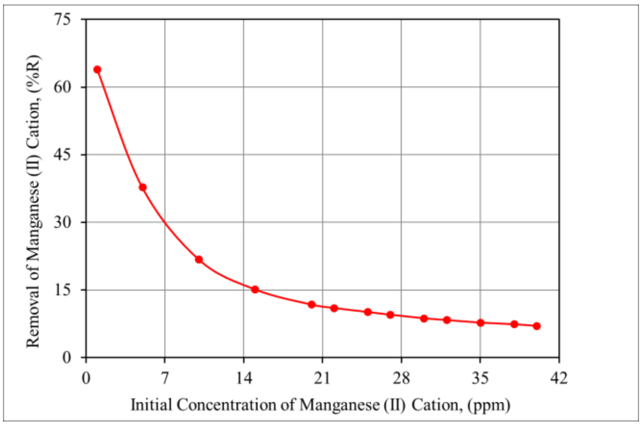

Figure 5: Effect of initial concentration on the removal of divalent manganese (Mn^{+2}) cations using Iraqi watermelon rinds @50°C, 1 g, 8, 180 min, and 400 rpm

3.4 Adsorbent dosage

The results obtained from varying adsorbent dosage, as shown in Figure 6, show that the relationship between the percentage removal and the dose of the adsorbent is a direct relationship as the percentage removal increase from 3% to 81.36% with an increase in the adsorbent dose from 0.5 to 6.5 g. The increase in the mass of the adsorbent means an increase in surface area and therefore an increase in the number of active sites that can participate in the capture of divalent manganese (Mn^{2+}) cations, given the constant concentration (Ibrahim et al., 2021). Consequently, the concentration of target (Mn^{2+}) cations in the solution will decrease. Thus providing a greater opportunity for target ions to reach the adsorption surface or vice versa, leading to higher treatment efficiency (Mahmood et al., 2025).

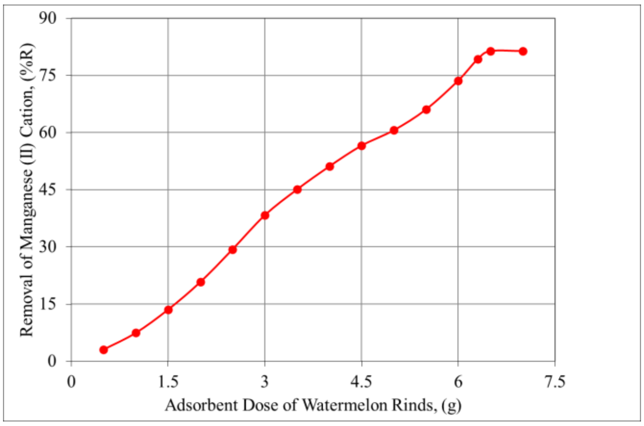

Figure 6: Influence of adsorbent dosage on the removal of divalent manganese (Mn^{+2}) cations using Iraqi watermelon rinds @50°C, 38 ppm, 8, 180 min, and 400 rpm

3.5 Contact time

The results obtained from varying contact time, as shown in Figure 7, indicate that the percentage removal increases from 2.75% to 81.36% with an increase in contact time from 5 to 150 minutes, respectively. This classic result can be explained by the fact that increasing the time increases the chance of divalent manganese (Mn^{+2}) cations reaching the active sites on the surface of Iraqi virgin watermelon rinds (Alwan et al., 2021). Upon reaching a time of 165 minutes, the treatment efficiency reaches its maximum value, and no significant improvement in the adsorption of pollutant (Mn^{2+}) cations occurs thereafter as a result of reaching a state of saturation, such that Iraqi virgin watermelon rinds cannot adsorb any additional amount of divalent manganese (Mn^{2+}) cations (Alhamd et al., 2024b).

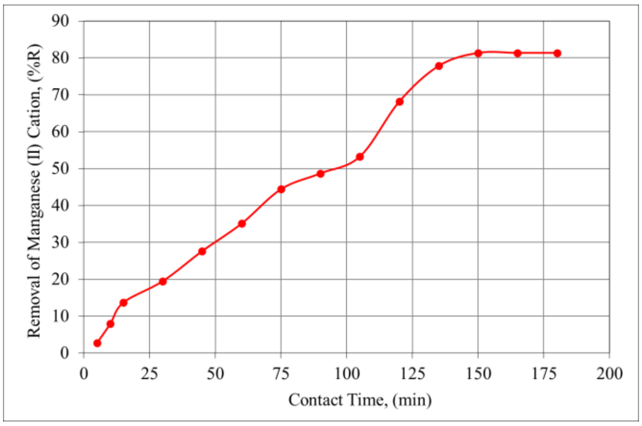

Figure 7: Influence of adsorbent dosage on the removal of divalent manganese (Mn^{+2}) cations using Iraqi watermelon rinds @50°C, 38 ppm, 8, 6.5 g, and 400 rpm

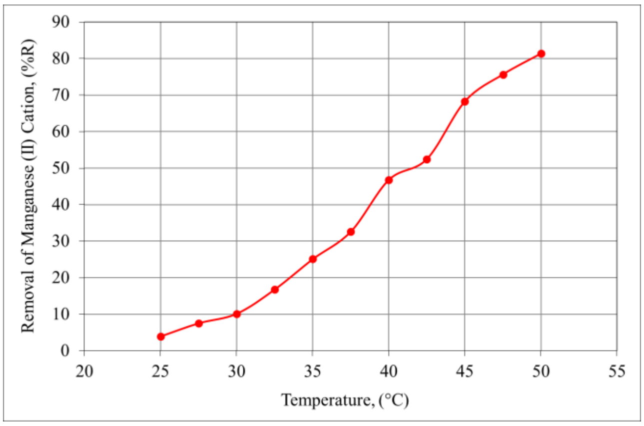

Figure 8: Influence of temperature on the removal of divalent manganese (Mn^{+2}) cations using Iraqi watermelon rinds @150 minutes, 38 ppm, 8, 6.5 g, and 400 rpm

3.6 Temperature

The results obtained from varying temperature, as shown in Figure 8, indicate that percentage removal increases from 4% to 81.36% with an increase in temperature from 25-50 °C, respectively. This behavior can be explained by the fact that increasing the temperature leads to an increase in the kinetic energy of the pollutant (Mn^{2+}) cations, thus increasing their chances of reaching the active sites on the adsorption surface (Abed et al., 2025a). The above results also indicate no decrease in processing efficiency with increasing temperature, ruling out the possibility of structural decomposition in the adsorbent material and demonstrating its high capacity for recovering target (Mn^{2+}) cations at elevated temperatures (Ali et al., 2024b).

3.7 Literature survey

Many researchers used raw and modified watermelon rind as a low-cost adsorbent to treat contaminated water. Each study obtained a different result. Table 2 show a literature review of most important of these investigations.

Table 2: Some studies used watermelon rinds as an adsorbent

Type of Watermelon rind	Modification method	Pollutant	Adsorption unit	Maximum Efficiency	Reference
Treated	Chemically by HNO_3, and Thermally	Methyl Parathion Pesticide	Batch	99%	Memon et al., 2008
Raw	-	Cu(II)	Batch	84%	Banerjee et al., 2012
Raw	-	Cd(II)	Batch	88.6%	Lakshmipathy et al., 2013
Treated	Chemically by HCl	Methylene blue, Crystal Violet, and rhodamine B dyes	Batch	489.8 mg/g, 104.76 mg/g, and 86.6 mg/g	Lakshmipathy and Sarada, 2014
Raw and treated	Chemically by H_2SO_4	Brilliant Green	Batch	92.6 mg/g and 188.6 mg/g	Lakshmipathy et al., 2015
Treated	Chemically by HCl	Pb(II), and Cu(II)	Batch	116.2 and 39.2 mg/g	Lakshmipathy and Sarada, 2016
Raw and treated	Microwave-Irradiated in H_2O_2, NaOH and deionized water	Cd(II)	Batch	55.80%, 69.39%, 66.22%, and 71.77%	Husein et al., 2017
Raw	-	methylene blue	Batch	188.68 mg/g	Jawad et al., 2018
Raw	-	Fe(II) and Pb(II)	Batch	89.03%, and 95.74%	Augustine et al., 2019
Raw	-	Ca(II)	Continuous	67 mg/g	Abbas and Nussrat, 2020
Treated	Microwave	Zn(II)	Batch	36.90%	Yılmaz and Tugrul, 2021
Raw	-	Ce(III), and Pr(III)	Batch	90%	Devanathan et al., 2022
Raw and treated	Chemically by citric acid	As	Batch	81% and 99.99%	Letechipia et al., 2023
Raw	-	Cr(VI)	Batch	83.41%	El-Tawabty et al., 2024
Treated	Chemically by triethylamine	F^-	Batch	85.97%	Ndung'u et al., 2025

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4. CONCLUSIONS

The current study reached several conclusions, the most important of which is that agricultural waste no longer represents an environmental problem, but has become an important resource for addressing environmental problems resulting from human activities. Therefore, factories and activities that generate industrial waste can be established at a suitable distance from agricultural fields to take advantage of this untapped resource. The results of this study proved that a dose of 63 g of Iraqi virgin watermelon rind, is capable of treating one liter of wastewater contaminated by divalent manganese (Mn^{+2}) cations with a removal percentage close to 81.5%, at a basic pH, 38 ppm initial concentration of manganese, and a 50 °C, 400 rpm agitation speed, and 165 minutes.

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