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Original Research Article

Efficiency of aptenia cordifolia mucilage in removal of anion dyes from aqueous solution

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ABSTRACT

Dye-containing wastewater is one of the main environmental challenges that can cause health issues. It is crucial to remove the dyes from wastewater before its discharge into the environment. This study aims to evaluate the efficiency of aptenia cordifolia mucilage in removal of anionic dyes from textile wastewater via coagulation and flocculation process. To this goal, the effect of different parameters including pH, initial dye concentration, concentration of plant mucilage and temperature on the efficiency of dye removal were investigated. The effect of application of poly(propylene imine) dendrimer (PPI) with the plant mucilage on the efficiency of dye removal process was evaluated.The results showed that the maximum dye removal with aptenia cordifolia (85.41 %) was obtained when using 5 mg of the plant mucilage at 25 °C and pH=4. Moreover, it was found that increasing the concentration of the plant mucilage and the initial dye concentration increased the dye removal efficiency. The application of 1 mg poly(propylene imine) dendrimer increased the efficiency of plant mucilage in the removal of anionic dye from aqueous solution from 85.41% to 92.18%. According to the results of this study, the application of aptenita cordifolia mucilage can be a beneficial, affordable and environmentally-friendly approach in the removal of textile anionic dyes from aqueous solutions.

Keywords: Wastewater, Phytoremediation, Flocculation and coagulation, Dendrimer

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Introduction

One of the main challenges of environmental issues is the quality of water resources [1]. Due to industrialization, a considerable amount of wastewater discharging to water bodies has threatened the environmental health [2]. Textile industry is one of the key industries in many countries, which consumes large quantities of water and is responsible for the pollution of large amounts of water [3].Textile industry wastewater contains suspended solid materials, aromatic hydrocarbons, surfactants, heavy metals[4], BOD, COD, and high amounts of dyes[5] that make it one of the most important problems of the environmental pollution. Dyes are complex organic materials[6] which enter the environment through the different stages of textile industry including dyeing and printing[7]. Some dyes are potentially toxic[8] with complicated molecular structure and low biodegradability with long-time stability in the environment[9]. Nowadays, there are around 100000 types of dyes in the world[10] with annual production of about 700000 to 1 million tons[6]. The annual consumption of dyes in textile is more than 36000 tons all over the world[11], 15% of which are wasted during the process of dyeing and finishing and are discharged into the environment as sewage and wastewater[6]. The discharge of textile wastewater into the ecosystems not only is aesthetically displeasing, but also disturbs the photosynthesis process due to the low light penetration[12]. Dyes are among the most dangerous chemical compounds found in textile wastewater that are the sources of dangerous by-products made by oxidation and hydrolysis[13]. Furthermore, most dyes cause allergies, skin diseases, cancer and genetic mutation in human kind[14,15]. Consequently, the discharge of textile industries effluent into the water resources and ecosystem affects the aquatic animals and food chain and causes irreparable damage to the environment[16]. Therefore, the treatment of colored textile effluent before its discharge into the environment is crucial[4]. Today, different treatment methods of industrial wastewaters are being studied in order to decrease its toxicity and meet the standards. Recently, special focus has been made on innovative methods for dye removal from textile effluent like adsorption by activated carbon, adsorption onto the new adsorbents, biomass adsorption, membrane filtration[17,18], chemical oxidation processes[8], electrolysis[19], photocatalysis[20], catalytic removal[21]and electrochemical treatment processes[22]. Besides being complicated[10] and costly, most of these methods produce a large quantity of sludge and other dangerous secondary products[23,24].

Nowadays, the coagulation and flocculation are among the most applicable techniques that are used extensively in treatment of the industrial wastewaters[2]. In these processes, chemical coagulants and natural or synthetic polymers are added to the wastewater. This process sticks the small pollutant particles together to make bigger particles to be removed more easily[5]. Although the processes of coagulation and flocculation by using inorganic substances like alum, ferric chloride, ferric sulfate, and poly aluminum chloride are used efficiently in textile wastewater treatment[25], but they suffer the disadvantages of being ineffective in low temperature[26], demanding to adjust pH before and after the treatment, producing a large amount of sludge, and introducing unwanted chemicals like aluminum, iron, sulfate and chloride into the environment[27]. Therefore, coagulation and flocculation by natural polysaccharides is among the most efficient, economic, and environmentally friendly methods of dye removal from textile wastewaters. Natural polysaccharides originate from biomass feedstocks, marine resources, and microorganisms which are considered as appropriate and environmentally-friendly substitutions for the synthesis of polymeric products and harmful synthetic coagulants in wastewater treatment[28]. The advantages of using these biopolymers are their availability, environmentally-friendly properties, biodegradability, non-toxic and unique molecular structure[29-31]. In recent years, many attempts have been made to improve the production and performance of polysaccharide-based bio-flocculants in the removal of dyes from textile wastewater. Feritas et al (2015) optimized the process of coagulation-flocculation by using Okra (A. esculentus) mucilage as a natural coagulant. The maximum percent removal of dyes by using okra was 93.57% at pH=6 and the coagulant dose of 3.20 mg/L[5]. Dalvand et al (2016) studied the use of Moringa stenopetala seed extract as a clean coagulant and compared it with Moringa stenopetala-Alum hybrid coagulant for textile wastewater treatment. It was found that increasing the coagulant dose and initial dye concentration improved the dye removal performance. In addition, the modification of Moringa stenopetala seed extract through hybridization with Alum increased the dye removal efficiency[27]. Basically, pectin drives from the plant cell walls[32]. It has coagulative activities and can be used as a coagulant in various suspensions in order to remove the dyes and other pollutants[31]. Aptenia Cordifolia is a fleshy short-lived species of succulent plants that is known as a flowering, covering herb resistant to heat and drought. Its mucilage can be used as a natural coagulant in coagulation and flocculation of anionic dyes because of having polysaccharides like glucose, fructose, glucosamine, xylose, lignin, and pectin[33-38]. Considering the fact that polysaccharide bio-based coagulants show high capability in coagulation of dyes and anionic compounds[31], in this paper, the efficiency of aptenia cordifolia in coagulation and flocculation of anionic dyes and their removal from textile wastewater is being examined.

Experimental

The anionic dye, Acid red 252 (polar red BL), was purchased from Ex-Ciba (Switzerland). Aptenia cordifolia was supplied from greenhouse of University of Birjand. Poly(propylene imine) dendrimer (3^{rd} generation) was obtained from SyMO-Chem (The Netherland). All other chemicals used in this study were provided from Merck (Germany). For optimizing the effect of process on coagulation and flocculation processes, different parameters like pH (4, 7, 10), initial dye concentration (6/25, 12/5, 25, 50 ppm), plant mucilage concentration (2, 4, 6 mg/L), temperature (20, 40, 60 °C) were varied. Additionally, PPI dendrimer (0, 0.2 and 1 mg/L) was employed in order to improve the efficiency of the dye removal.

Preparation the plant mucilage

To extract the mucilage, firstly, leaves were washed by distilled water twice to remove the dirt and dust. Secondly, they were placed in the room temperature for two hours. Thirdly, leaves were inserted in a polyethylene pocket and were compressed to take their extract. Finally, mucilage was placed in a cold and still place for 48 hours and after settlement, a sample of supernatant were collected and used as a coagulant.

Experimental method

First, 50 ml of Acid red 252 solution with concentration of 50 ppm was provided. Sodium carbonate 0.1 M and sulfuric acid 0.1 M were used to adjust the pH (4, 7, and 10) by pH meter HANNA, model pH211. The effect of pH was examined in 3 beakers containing 50 ml of dye liquid with 50 mg/L concentration and 6 mg of plant mucilage.

To assess the effect of initial dye concentration on coagulation-flocculation process, different concentrations of the dye under study (50, 25, 12.5, and 6.25 mg/L) were provided.

The optimization of the plant mucilage amount in optimum pH (acidic) in 50 ml of dye solution with concentration of 50 mg/L (optimum dye liquid concentration), and increasing the concentration of plant mucilage from 1 ml to 3 and 5 ml were studied. To examine the influence of temperature in the removal of dye by coagulation-flocculation process, 3 beakers holding 50 mg of dye solution with 50 mg/L concentration, optimum pH and optimum plant mucilage concentration were tested in different temperatures of 25, 40, and 60°C. To study the efficiency of dendrimer polypropylene imine 3rd generation on this process, dendrimer amount increased from 0 to 0,2 and 1 mg in optimum conditions of dye concentration, plant mucilage concentration, pH and temperature. At each stage of optimization, after adding the plant mucilage, magnetic stirrer (MS-300HS) was applied to stir and blend the solution at fast speed of 100 rpm for 2 minutes followed by slow speed of 40 rpm for 10 minutes. After 30 minutes of settlement, the residual dye concentration was measured at the wavelength of 473 nm by Jenway UV/Visible spectrophotometer model 6305.

Results and discussion

Investigating the effect of pH on coagulation-flocculation process

pH is an important controller parameter in dye removal process. According to the results, by reducing pH from alkaline to acidic, the efficiency of dye removal increased from 0.63 to 72.36 %. the mucilage of aptenia cordifolia in acidic condition is more effective in removing the anionic dye and this characteristic may be due to the below facts:

1. When pH of the dye solution is such that dye molecules exist in ionized form, the adjacent molecules on the flocculant surface will repel each other significantly because of their same electrical charge. Therefore, the dye molecules cannot pack together densely on the surface of the flocculant. In contrast, when the dye molecules are not in the ionized form, no electrical repulsion exists; thus the packing density of the dye molecules by the flocculant will be at its maximum level. It can be said that dye molecules in acidic pH are not ionized and this property increases the flocculation of dye molecules at this pH[39].
2. The maximum removal of anionic dyes was expected at acidic pH as these dyes (anionic dyes) give anions to the aqueous solution that are neutralized in the presence of H+ ions[39].
3. At acidic pH, the cationic strength of lignin increases; therefore, its flocculation ability and consequently its dye removal intensify as well. (At acidic pH, the ionic charge of

polysaccharide becomes positive, ending in more flocculation and adsorption of ionic dye) [40].

The results of this research is in line with the study done in 2010 by Run Fang and others. In their study, the removal of azo anionic dye by cationic flocculants was investigated. It was found that by decreasing pH, the amount of flocculants needed for optimum dye removal decreased. Therefore, the maximum dye removal occurred in acidic pH such that under low pH conditions, the amine groups existed in dye became protonated by chitosan cations. Low pH improved the cationic strength of chitosan and enhanced the flocculation efficiency[40].

Table 1. the effect of pH in coagulation-flocculation of anionic dye.

pH	Plant mucilage (mg)	Dye concentration (mg/L)	Contact time (min)	Temperature (centigrade)	Percent removal
4	5	50	30	25	73.36
7	5	50	30	25	27.06
10	5	50	30	25	0.63

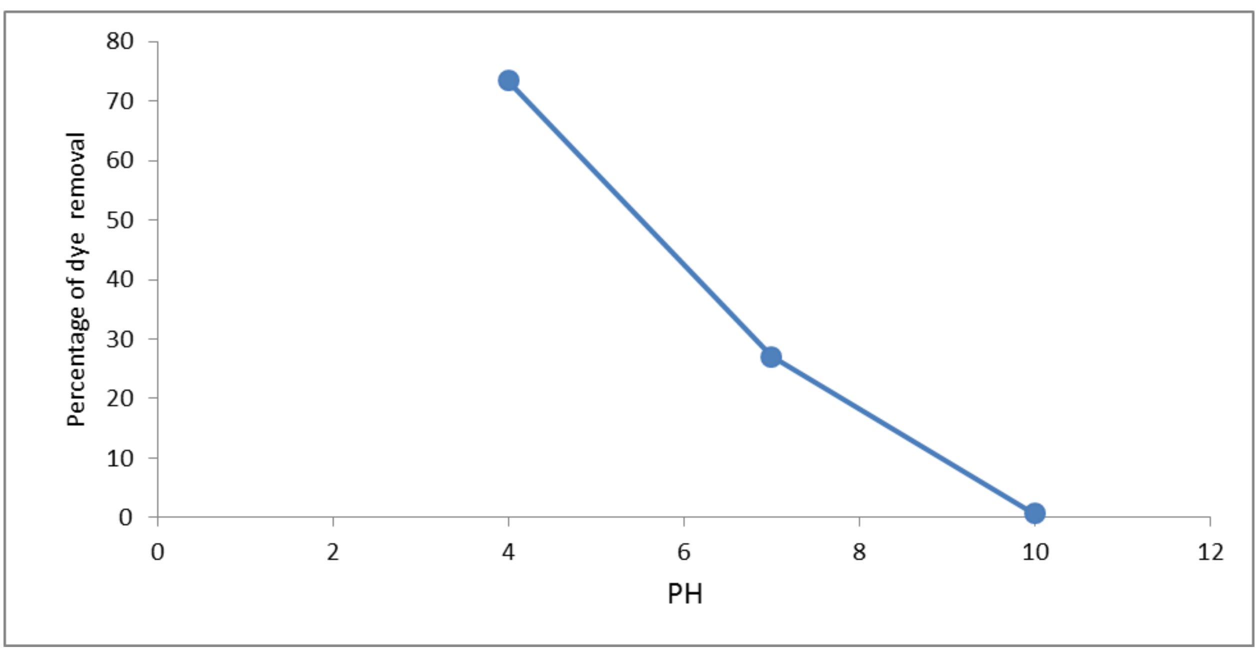

Fig 1. the effect of pH on the removal of anionic dye.

Investigating the effect of plant mucilage concentration on coagulation-flocculation process

Plant mucilage concentration plays an important role in the process of coagulation-flocculation. It was seen that by increasing the amount of plant mucilage in acidic pH, the percent removal of dye improved from 73.36% to 85.41%.This may happen because of the presence of polysaccharides like glucose, fructose, glucosamine, xylose, lignin and pectin in aptenia cordifolia mucilage which enhance the coagulation-flocculation of dye. Colloidal particle aggregation by polysaccharide flocculants is considered to be based on two main mechanisms: 1. Neutralization of charge, and 2. Attachment of neutralized particles (bridging) [28]. When the polysaccharide molecular weight is low, the neutralization mechanism is the effective mechanism for coagulation-flocculation. On the contrary, the bridging mechanism is the effective mechanism in case the polysaccharide molecular weight is high[28]. Pectin is basically driven from plant cell walls and has flocculative activities and can be used as a flocculant in various suspensions for removing dyes and other pollutants. Pectin has bridging property which leads to flocculation and settling of dye particles. Lignin is cationic and has charge neutralization characteristic which causes the flocculation and coagulation of dyes. With an increase in flocculant dose, the efficiency of charge neutralization and dye removal increase as well. Thus the higher the plant mucilage concentration and the higher the concentration of the polysaccharide flocculant, the better the performance of dye coagulation-flocculation and dye percent removal.

Mishra and others (2005) studied the performance of Tamarindus mucilage in relation to removal of direct and vat dyes. They investigated the dye removal performance by testing various doses of flocculants and found that increasing the flocculant dose ended in the enhancement of percent dye removal. Regarding the fact that the effective mechanism for flocculation is bridging between particles, increasing the flocculant concentration dosage led to more aggregation and density of dye particles and better dye removal[41].

Table 2. the effect of plant mucilage in coagulation-flocculation of anionic dye.

Optimization of plact extract concentration (mg)	Plant extract (mg)	pH	Contact time (min)	Temperature (centigrade)	Percent removal
1	50	4	30	25	73.36
3	50	4	30	25	79.28
5	50	4	30	25	85.41

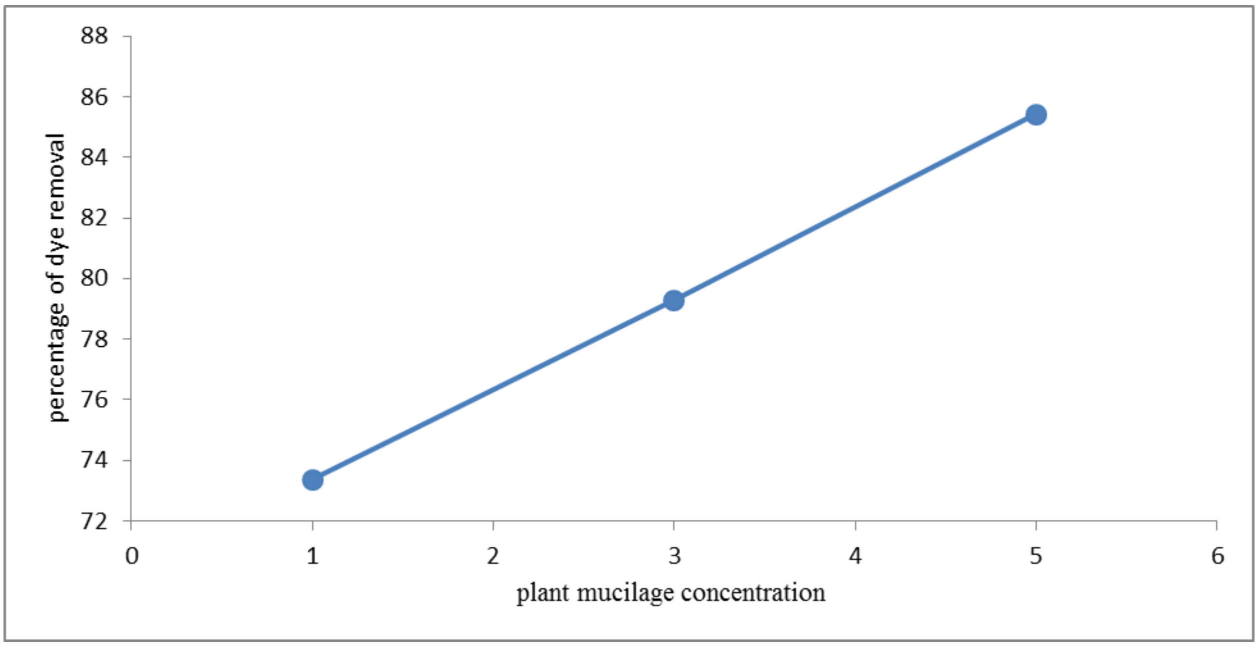

Figure 2. The effect of plant mucilage concentration in coagulation-flocculation of anionic dye.

Investigating the effect of initial dye concentration on coagulation-flocculation process

Initial dye solution concentration can be an effective parameter on dye coagulation-flocculation process. It was observed that with an increase in dye concentration in the same temperature and at the same pH with 5 mg of plant mucilage, the efficiency of dye removal by aptenia cordifolia mucilage increased. It can be interpreted that polysaccharide plays as a bridge between dye molecules. In other words a chain of dye-polysaccharide-dye was formed.

The existence of functional groups on plymers to interact with colloidal particles can increase the efficiency of removal. The functional groups on the polysaccharides adsorbs a colloidal particle on the surface. Second particle sticks to the remaining vacant sites of functional groups and so on. So, polymer acts as a bridge [34,39]. In a study by Mishra and others (2005) the application of a food grade polysaccharide assessed for textile wastewater treatment. Dye removal efficiency enhanced by increasing the dye concentration due to the possibility of forming particle-polymer-particle structure in higher dye concentration [39].

Table 3. The effect of initial dye concentration on the removal of anionic dye.

dye concentration (mg/L)	Plant mucilage (mg)	pH	Contact time (min)	Temperature (centigrade)	Percent removal
50	5	4	30	25	85.41
25	5	4	30	25	59.36
12.5	5	4	30	25	40.46
6.25	5	4	30	25	22.86

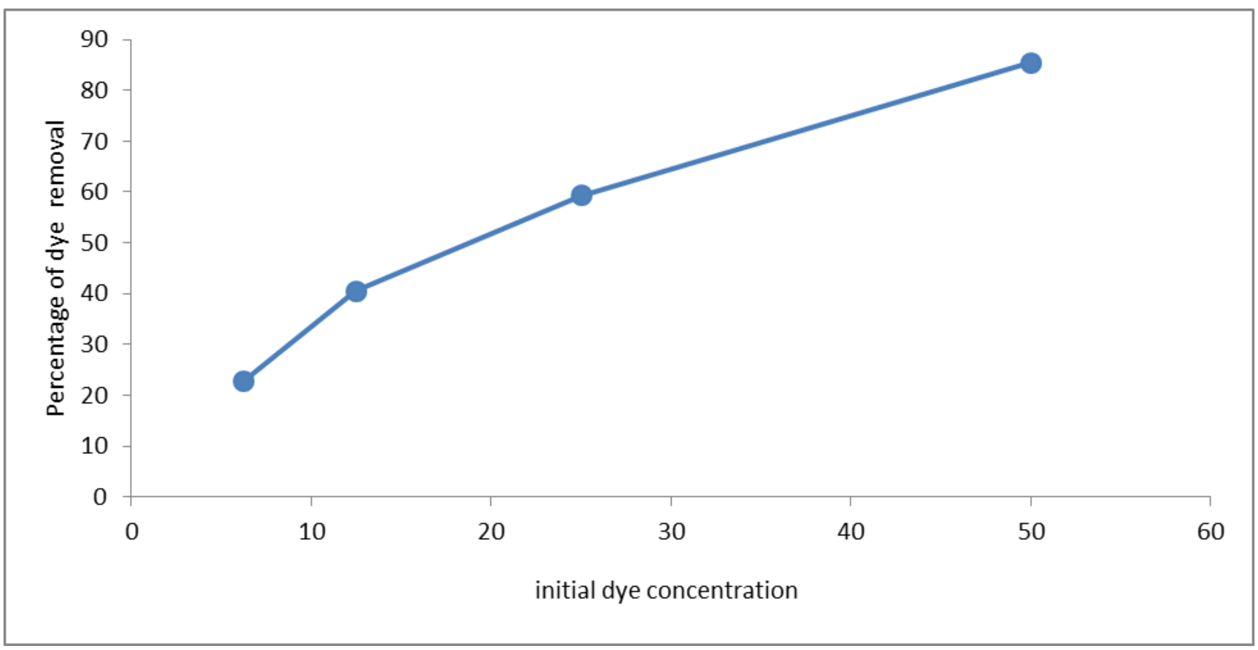

Figure 3. the effect of initial dye concentration on the removal of anionic dye.

The effect of temperature on coagulation-flocculation process

Temperature is one of the most important parameters in chemical-physical processes and can be operative in coagulation-flocculation process for dye removal. It was perceived that increasing the temperature decreased the dye removal from 85.41 to 27.79. According to the obtained results, aptenia cordifolia mucilage has better performance in removing the anionic dye AR252 from aqueous solution in lower temperatures. Higher temperatures break the molecular chains and stimulate the structural deformation in configuration and robustness of polysaccharides and prevent the formation of effective grafting between dye particles in dye solution[28,42] and hence decrease the possibility of dye flocculation by polysaccharides present in aptenia cord folia. In a research done by Wang and others (2015), cationic copolymer, Xylan METAC was used to treat the textile wastewater. Increasing the temperature higher than 80 centigrade did not improve the charge density, grafting ratio and removal performance. This might be due to termination of the reaction in this temperature[43].

Table 4. the effect of temperature on anionic dye removal.

Optimization of temperature	Plant mucilaget (mg)	pH	Contact time (min)	Dye concentration (mg/L)	Percent removal
25	5	4	30	50	85.41
40	5	4	30	50	41.86
60	5	4	30	50	27.79

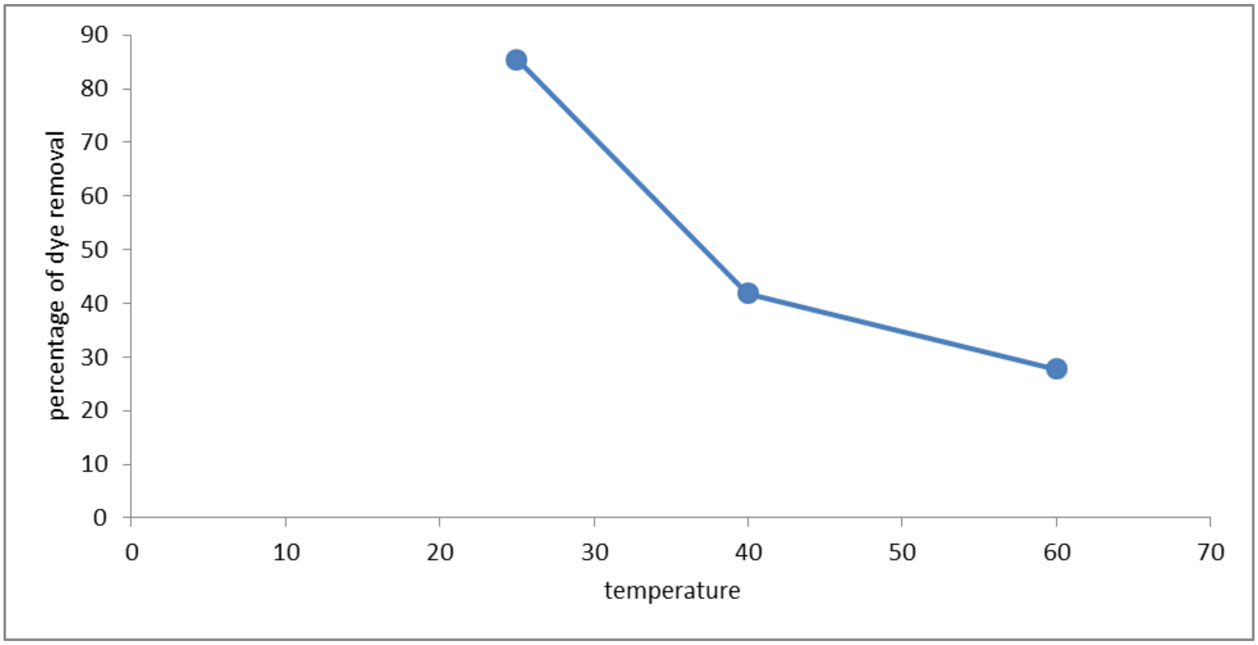

Figure 4. the effect of temperature on anionic dye removal.

Investigating the effect of dendrimer polypropylene imine 3rd generation in coagulation-flocculation process

To improve the performance of aptenia Cordifolia in anionic dye removal and to study the efficiency of poly(propylene imine) dendrimer on this process, dendrimer amount increased in optimum conditions. The result revealed that with increasing the amount of dendrimer, the efficiency of dye removal increased as well. Dendrimers consist of a focal core and many treelike branches. On one hand these particles are able to embed and enclose dye molecules in their branches. On the other hand, they can connect and carry different molecules on their surface due to the presence of various functional groups on the surface. Consequently, these particles can be used as an efficient adsorbent and facilitator for improving the plant extract performance in dye removal.

Table 5. investigating the efficiency of dendrimer poly propylene imine 3rd generation on anionic dye removal.

Optimization of dendrimer amount (mg)	Plant extract (mg)	pH	Initial dye solution concentration (mg/L)	Temperature (centigrade)	Percent removal
0	5	4	50	25	85.41
0.2	5	4	50	25	88.79
1	5	4	50	25	92.18

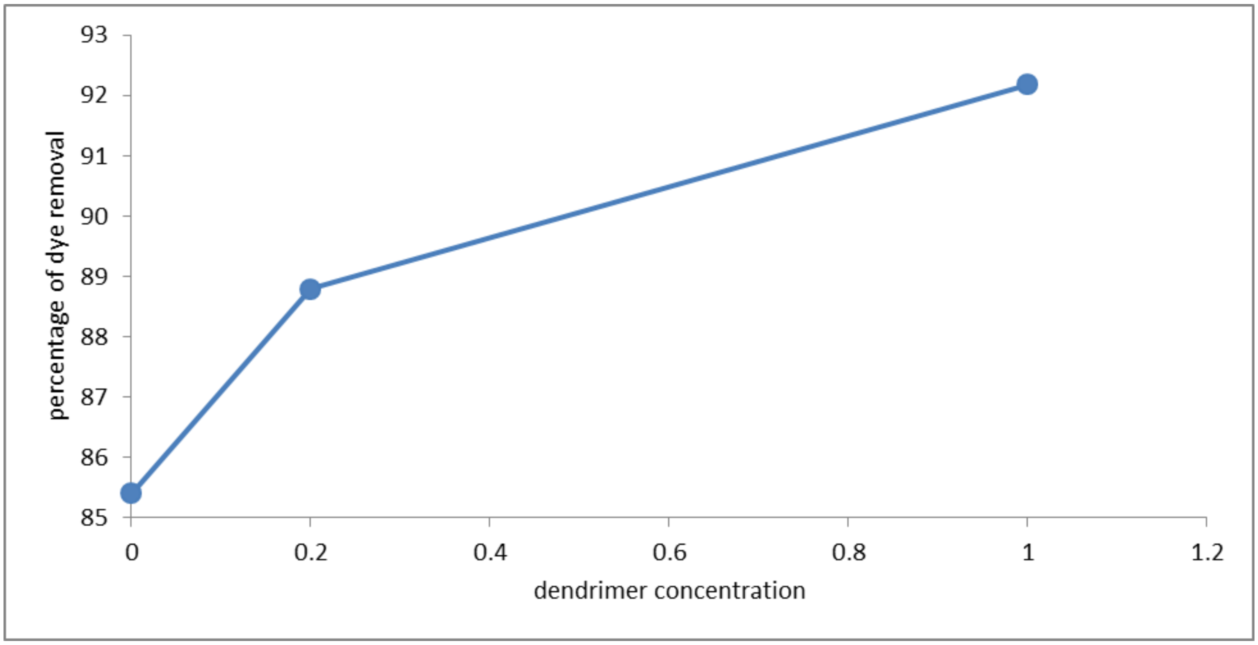

Figure 5. efficiency of dendrimer poly propylene imine 3rd generation on anionic dye removal.

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