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DYE WASTEWATER TREATMENT: REMOVAL OF REACTIVE DYES USING INORGANIC AND ORGANIC COAGULANTS

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ABSTRACT

Wastewater treatment in textile and dye industry mainly involves treatment of highly colored wastewater containing variety of dyes in different concentrations. The wastewater needs to be treated prior to discharge by effectively removing dye color in order to protect environment and as per the statutory guidelines. Coagulation is the most commonly used method in the treatment of textile wastewater. In the present work, we report experimental work on treatment of synthetic waste containing pure reactive dyes, removal of reactive blue dye, using various commercial coagulants such as alum, polyaluminium chloride (PAC), polydiallyldimethylammonium chloride (poly-DADMAC) and polyamines. The optimum coagulant dose and pH values were determined for these inorganic and organic coagulants by comparing color removal efficiency. It was found that for inorganic coagulant PAC, the color removal was near total at concentration of 200 ppm and above, whereas for alum much higher dosage was needed (> 500 ppm). In both the cases, the color removal efficiency was strongly dependent on pH and best colour removal efficiency was observed only at pH of 5. For polyamine and poly-DADMAC, the optimum coagulant dose was much less as compared to inorganic coagulants. Further, it was found that the color removal efficiency is practically insensitive to pH and the results were good in the pH range of 3 to 10. The results clearly indicate that organic coagulants are more effective in treating wastewaters containing reactive dyes. The study also confirms the strong dependence of color removal efficiency on coagulant type, coagulant dosage and pH apart from its dependence on nature of dye. In an attempt to combine effects of both inorganic and organic coagulants, studies have also been carried out on development of formulations by various combinations of inorganic coagulants such as alum, PAC and organic coagulant-PolyDADMAC. The results of such formulations have been compared with those obtained using individual inorganic and organic coagulants. The results clearly indicate that development of formulations can offer superior solution to coagulation problem as compared to individual coagulants.

INTRODUCTION

Textile and dyeing mills use many kinds of artificial composite dye and discharge large amounts of highly colored wastewater. These wastes must be

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treated prior to discharge in order to comply with the environmental protection laws for the receiving waters. Biological treatment processes are frequently used to treat textile effluents. These processes are generally efficient for biochemical oxygen demand (BOD) and suspended solids removal (TSS) but they are largely ineffective for removing color from the waste (Aziz *et al.*, 2007; Basava Rao and Maohan Rao, 2006). Consequently treated waste effluents may contain appreciable amounts of color when discharged. Now, the treatments are physico-chemical treatment operations, including adsorption, oxidation, chemical precipitation etc. Each has its merits and limitations in application.

In recent years, considerable research has been done on the removal of color from textile effluents. In general these studies involved use of inorganic coagulants such as alum, lime, ferric or ferrous sulfate and polyaluminum chloride (PAC) etc. In most cases coagulation has been effective in removing color especially for wastewaters, containing dissolvable solids. However high chemical dosages are usually required and large amounts of sludge volume must be disposed off. The cost of sludge disposal results in relatively high process costs.

Organic coagulants such as polydiallyldimethylammonium chloride (poly-DADMAC) and polyamine have been shown to be an effective alternative to inorganic coagulants and can enhance removal of impurities or pollutants in the treatment of wastewater. The zero production of sludge almost eliminate sludge disposal problems and significantly reduce treatment costs.

The objective of this study is to compare the performance of organic coagulants like poly-DADMAC and polyamine with inorganic coagulants such as ferrous sulfate, alum and PAC for removing color from the dye waste water and to attempt further development of a suitable formulation combining inorganic and organic coagulants for enhancing coagulation effects with reduced coagulant quantities. Dye used is Reactive blue. The study concentrates on the effects of pH and coagulant dosage on color removal efficiency. The reason for selecting reactive blue dye for testing effect of combined coagulant system or formulation is that it is one of the most difficult dye to treat using conventional coagulation. The reactive dyes pose the greatest color removal problems and even concentration as low as 0.005 ppm is visibly detectable. Further, reactive dyes are not easily biodegradable and even after extensive treatment, color is likely to remain in the effluent. Thus, in the absence of any effective conventional treatment methodology for reactive dyes, it is, but, required to develop an effective formulation in the form of combined coagulants for effective color removal of reactive dyes. Further, at present investigations into reactive dye wastewater treatment are rarely reported in literature (Kim *et al*., 2004; Klumuik *et al*., 1999).

MATERIALS AND METHODS

Coagulants poly-DADMAC, polyamine and PAC used in this study are of commercial grade. Ferrous sulfate, Alum and reactive dye are of A.R. grade. The Reactive Dye used is of commercial grade.

Laboratory tests were conducted on synthetic reactive dye wastes which were prepared by adding 100 mg of the dyestuff in 500 ml of water. Synthetic waste were studied in order to determine the effectiveness of various inorganic and organic coagulants for removal of color of the dye-stuff from the water. It was hoped that use of pure dye solutions would make it possible to obtain information about the color removal efficiency of coagulants for individual dye. The optimum pH value and optimum dosages of reactants required for color removal were determined by a jar test procedure. One liter beakers containing 500 mL of dye waste were placed on Jar Test Apparatus having six stirrers (Make-Kumar Sales Corporation, Bombay). The coagulants, dye and distilled water were initially mixed at highest rpm for 2 min. Then followed a 15 min period of slow stirring at 40 rpm and 1 hr of sedimentation. After settling, an aliquot of suparnatant was pipetted out from each jar and then analyzed on colorimeter (Elico make, Model CL 157) to determine % color removal. The characteristic wavelength for the dye was determined on a UV-Visible spectrophotometer (Jasko make, Model V-530). The maximum absorbance wavelength (λmax) was used for absorbance reading on the colorimeter. Percentage of color removal was calculated by comparing the absorbance values for the wastes after treatment to the absorbance value for the original waste. Distilled water served as reference. The % color removal was calculated by the formula (Javaid Mughala, 2013;atel *et al*., 2012).

$$\% \text{ Color removal} = (Abs_o - Abs_t) \times 100/Abs_o$$

Where C_o and C_e are initial and equilibrium con-

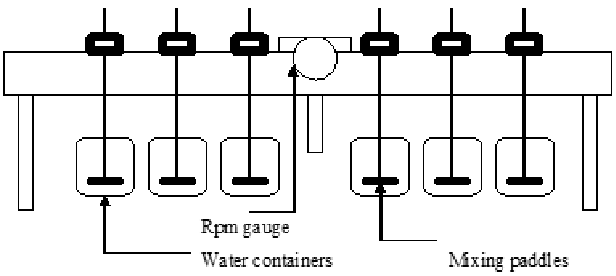

Fig. 1 Schematic Experimental set up: Jar test apparatus

centrations of the dye, respectively

The jar test apparatus (Figure 1) contains six paddles, which stir the contents of six 1liter containers. One container acts as a control while the operating conditions can be varied among the remaining five containers. A rpm gauge at the top-center of the device allows for the uniform control of the mixing speed in all of the containers.

The jar test is a common laboratory procedure used to determine the optimum operating conditions in coagulation/ flocculation studies. This method allows adjustments in pH, variations in coagulant or polymer dose, variation of mixing speeds, testing of different coagulant or polymer types, on a small scale in order to predict the functioning of a large scale treatment operation. A jar test simulates the coagulation and flocculation processes that encourage the removal of suspended colloids and organic matter, which can lead to color, turbidity, odor and taste problems (Ismail *et al.*, 2012).

RESULTS AND DISCUSSION

Dye water treatment using inorganic coagulants

Typical inorganic coagulants are aluminum sulfate, aluminum chloride, ferric chloride/sulfate, calcium/ magnesium oxide etc. As discussed earlier, the cation of the coagulant is mainly responsible for the neutralization of negative charge and the trivalent cations have much stronger ability as coagulants than divalent. The mono valent cations of coagulants have practically very weak coagulating action and are less commonly used. Alum (aluminum sulfate) is one of the most widely used inorganic coagulant and it produces insoluble precipitate of aluminum hydroxide and works better in the narrow pH range close to 5. Alum is comparatively cheaper than most other coagulants. However, its effectiveness is limited in many wastewater treatment applications. Ferric compounds such as ferric chloride and sulfate are also some of the popular inorganic coagulants which work over a same range of pH as Alum (Merzouk *et al*., 2011; Aziz *et al*., 2007).

In general, inorganic coagulants produce smaller and lighter flocs that require larger time to settle. It also reflects into the fact that the sludge volume is always greater with inorganic coagulants. Another disadvantage with most inorganic coagulants is that they are pH sensitive and therefore work only in a narrow pH range. Some of the disadvantages of common inorganic coagulants can be avoided with the use of organic coagulants or using formulations of both inorganic and organic coagulants. Recently, use of inorganic coagulants such as polyaluminum chloride (PAC) or polyaluminum sulfate (PAS) has gained wide importance in many applications. The PAC products are comparatively less sensitive to pH. Further, there is some manipulation possible with respect to molecular weight of PAC and its basicity. For the PAC with general formula of

$Al_n (OH)_n Cl_{(3n-m)}$

The basicity is defined as:

Basicity = m/3n = number of (OH) groups per Al

It is to be noted that, so far, no criteria or guidelines are available for preparation of PAC with specific molecular weight and basicity for any particular application. This therefore requires forming recommendations for any particular product on the basis of experimentally obtained data for a specific application.

Effect of coagulant concentration

When PAC was used as a coagulant for treating water containing Reactive blue dye, following were the observation made :

1. Below 100 ppm dose of coagulant less color removal
2. Above 100 ppm dose of PAC nearly complete color removal was observed.
3. Nearly 100 % colour removal can be obtained using PAC above 100 ppm dosage.

It was also observed that sensitivity of color removal efficiency with change in coagulant dose above 150 ppm was negligible.

In case of alum it can be seen that the color removal efficiency increases very slowly with increase

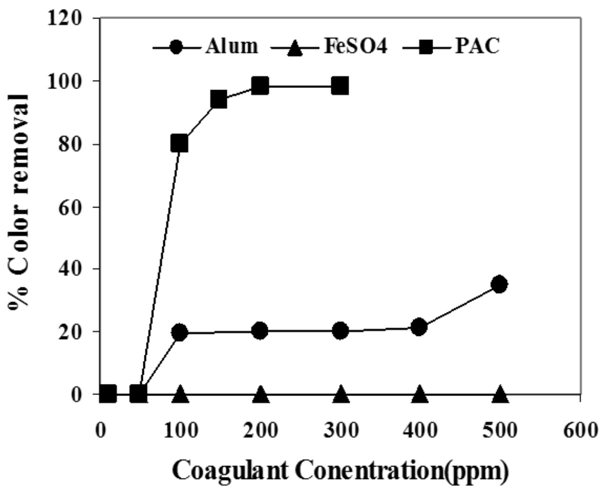

Fig 2. Effect of coagulant concentration on % colour removal using inorganic coagulants

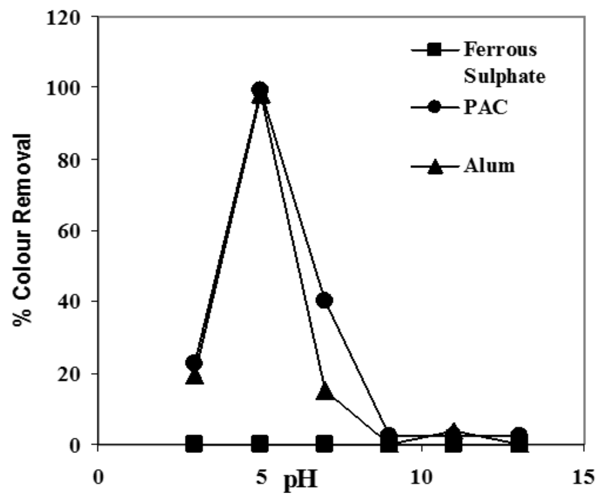

Fig 3. Effect of pH on % colour removal using inorganic coagulants

in coagulant concentration. The optimum coagulant concentration lie above 500 ppm. The quantity of alum required as compared to PAC is very high.

Ferrous sulfate was found to be largely ineffective in removal of color of reactive dye from solution. The color removal is almost zero at all concentrations of ferrous sulfate.

Effect of change in pH

It was found that pH value for reactive blue solution has sharp dependence on color removal efficiency. Maximum color removal was observed at pH 5. Color removal was largely incomplete above and below pH 5. Drastic reduction in color removal was observed above and below pH 5. The performance of PAC as well as alum in case of change in pH was observed to be same. Change of pH did not improve performance of ferrous sulfate and at values of pH (i.e. from 3 to 12) the color removal was almost incomplete.

The PAC is an inorganic coagulant and its trivalent aluminum ions react with hydroxyl and other alkaline ionic species. The hydroxides of aluminum are almost insoluble at neutral pH, which precipitate and settle down. Although it is believed that, precipitate of aluminum hydroxide ($Al(OH)_3$) is the final product, there are many intermediate species formed in the process of coagulation which are of the type: MOH_2+, $M(OH)_2+$ and $M(OH)_4-$. It is also possible that the intermediate hydrates can polymerize into chain of metal hydrate units, which carry particle charges along the chain. The role of intermediate metal hydrate is important in the development of coagulation process. The polymeric chains, especially are capable of physically capturing the colloidal solids and thereby its physical removal. The extent of coagulation as well as rate of coagulation greatly depends on concentration of coagulant and pH. The pH has its strong effect through generation and dictating lifetime of intermediate reaction products and hence optimum pH is the key to effective coagulation.

Klimiuk *et al.* (1999) have reported study on use of PAC in the case of reactive dyes and also found strong dependence on dye type and coagulant dose. Joo *et al.* (2007) found that very high dosage of inorganic coagulant and polymeric coagulant are required for reactive dye wastewater treatment. The mechanism of the dye removal is largely abstract. The main reason for this is the fact that different dyes have different chemical structure. Dyes are composed of groups responsible for dye colour containing azo (N=N), carbonyl (C=O), methine (-CH=), nitro ($-NO_2$) and quinoid groups. There are also electron withdrawing or donating groups responsible for colour modification such as amine ($-NH_3$), carboxyl (-COOH), sulfonate ($-SO_3H$) and hydroxyl (-OH). Thus, depending upon the dye type, interaction of coagulant can substantially change because of the nature of the functional groups, number of functional groups. Hence there is substantial variation in dye removal performance even with same coagulant system for different dye types. Gao *et al.* (2007) have attempted to study mechanism of

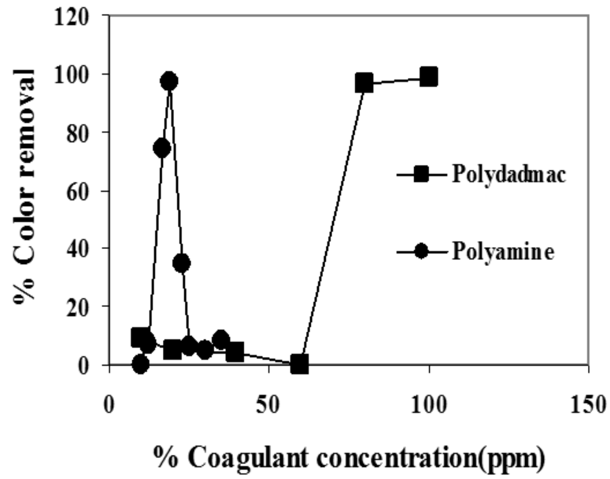

Fig. 4 Effect of change in concentration on color removal efficiency of organic coagulants

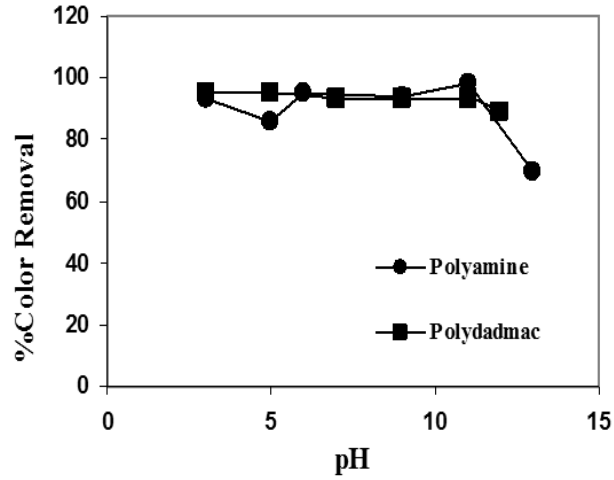

Fig 5. Effect of pH on % colour removal using organic coagulants

dye removal using $MgCl_2$ as a coagulant. They also indicated strong dependence of color removal efficiency on the type of dye, pH and dosage of coagulant. Their results were, however, indicated good color removal only at pH values greater than 12 for both reactive and disperse dyes, indicating color removal through precipitation of magnesium hydroxide at pH 12 and above. This is quite diffe- rent proposition and action than that reported in this work.

The pH dependence of dye removal is also found in adsorptive separations. Santhy and Selvapathy (2006) have reported significant pH dependence in the case of adsorption of reactive dyes on activated carbons and found satisfactory colour removal only in the pH range of 1-3. Flavio *et al.*, (2007); Basava Rao and Mohan Rao (2006); Joo *et al.*, (2007) and Isa *et al.* (2007) also supported such pH dependence in adsorptive removal of dyes. Further, in the case of adsorption using activated carbons, the carbon affinity for reactive blue was found lowest indicating severe difficulty in its removal (Santhy and Selvapathy, 2006).

Dye water treatment using organic coagulant

Effect of Coagulant concentration

It was found that no color removal was there up to 60 ppm of poly-DADMAC and maximum at 80 ppm and above. The trend is almost similar to inorganic coagulant PAC. However lower concentration of organic coagulant (poly-DADMAC) was required for same separation as compared to inorganic coagulant PAC.

In case of polyamine the color removal efficiency of reactive dye increases with increase in the concentration of coagulant. The color removal is found to be almost complete at concentration of 22 ppm. There is drastic change in color removal efficiency below and above this coagulant concentration.

When performance of poly-DADMAC and polyamine is compared it can be seen that the trend of color removal efficiency v/s change in coagulant concentration is entirely different in both the cases. In case of polyamine color removal efficiency first increases and then decreases whereas in case of poly-DADMAC color removal increases with increases in coagulant concentration.

Effect of change in pH

The experimental results have shown that sensitivity to pH was less in case of organic coagulant as compared to inorganic coagulant for reactive blue. However maximum color removal was achieved at pH 3 and 5.

As discussed earlier in the case of inorganic coagulants, polyelectrolytes or organic coagulants are largely pH insensitive. Thus organic coagulants can be used in fairly wide range of pH for dye wastewater treatment application. Our results on the use of organic coagulant - poly-DADMAC and polya- mine also supports these observations. For reactive dyes behavior with organic coagulants, poly-DADMAC agrees well with that reported in the literature. Further, as compared to PAC, the sensitivity to coagulant concentration was much more in case organic coagulants. Similar to dosage of polymer in our studies, Gao *et al.*, (2007) have indicated much higher coagulant dose for reactive blue. Also, Kang (2007) studies on reactive dyes showed requirement of high dose of ploy-DADMAC for satisfactory removal. The large molecular weight of poly-DADMAC is believed to have significant role in determining the coagulant concentration. More the molecular weight, better performance of the coagulant can be expected through larger interaction with positively charged nitrogen groups and efficient physical removal of suspended colloidal particles. The effect of molecular weight is also important in the case of PAC. However, since data on the polymerization conditions, molecular weight of the polymer and characterization in both organic and inorganic polymer is not reported in the literature, it is difficult to comment on these issues and quantify results on the basis of these parameters at present. Yu *et al*. (2002) have attempted characterization of interactions of sulfonic acid groups and carboxyl groups in the case of PAN-DCD polymer but without any quantitative analysis. The data on these parameters will also be useful in development of mathematical models by incorporating charge interactions and surface interactions corresponding to different dye species.

The approach to developing solution to coagulation problems is, by and large empirical and a priori selection of any coagulant system for any specific application is difficult. Both inorganic and organic coagulants have their own advantages and disadvantages. In many cases, use of one coagulant alone (inorganic or organic) may not give effective coagulation. Further there are variations in size, type and concentration of suspended solids apart from dye quantity etc in the wastewater, which may necessitate use of a better coagulant system combining advantages of both inorganic and organic coagulants. It is known that polyelectrolytes are often successfully used with inorganic coagulants and in many cases result of using alum in combination with organic coagulant is more beneficial than using either single inorganic or single organic coagulant. In principle, any two or more coagulants can be combined to enhance the effect of coagulation. The combination can be among class of inorganic coagulants, organic coagulants or it can be in both inorganic and organic coagulants. The inorganic coagulants usually produce smaller, lighter and fluffier flocs and thereby have problems of settling as compared to that in case of organic coagulants and it creates disposal problem. As found earlier in these studies and also reported in literature, inorganic coagulants are much more pH sensitive as compared to organic coagulants and therefore application of inorganic coagulants alone is limited. Thus it makes sense to combine inorganic and organic coagulants to exploit advantages of both types in effective coagulation. The objective in developing such formulation would be to have efficient coagulation process operating over a wide pH range with better settling properties and minimum sludge volume. Although, the results with polyamine were quite good, especially with respect to the quantity of coagulant, it was observed that the performance with polyamine was not completely reproducible. Therefore, further experiments were mostly focused on poly-DADMAC organic coagulant. In this work we attempt to study such an approach of developing formulations with combinations of Alum, PAC and poly-DADMAC for treating reactive blue dye wastewater. Reactive dyes, in general, are quite difficult to treat using conventional coagulation methodology apart from biodegradability problems. In the absence of any effective conventional treatment methodology for reactive dyes, it is instructive to investigate development of a suitable formulation in the form of combined coagulants for effective color removal of reactive dyes. Further, at present investigations into reactive dye wastewater treatment are rarely reported in literature (Kim *et al*., 2007). In view of this, we have also attempted to study development of such formulations by combination of inorganic coagulants such as alum and polyaluminum chloride and organic coagulant- poly-DADMAC.

Formulation of PAC with poly-DADMAC

In the first combination, for developing formulation of PAC with poly-DADMAC, a PAC concentration of 200 ppm was used in all the cases, while poly-DADMAC concentration in the formulation was varied from 20 ppm to 100 ppm. It is to be noted that poly-DADMAC alone effects poor colour removal up to a concentration of 60 ppm. This is indicated by flat dotted lines in Figure 6 for poly-DADMAC concentrations of 20 ppm and 60 ppm. For poly-DADMAC concentration of 100 ppm, when used alone, the colour removal was very good of the order of 98 % over a wide range of pH. However, when the same concentration is used in combination with 200 ppm PAC, the performance was good only at pH of 3 and color removal was drastically lower at higher pH. This could be due to more prominent effect of PAC in the combined formu- lation. This is also evident from the fact that PAC alone gives no

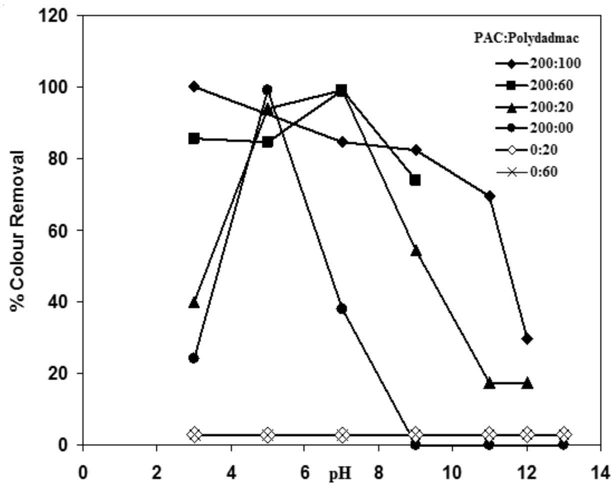

Fig 6. Effect of pH on % colour removal using PAC and poly- DADMAC formulation

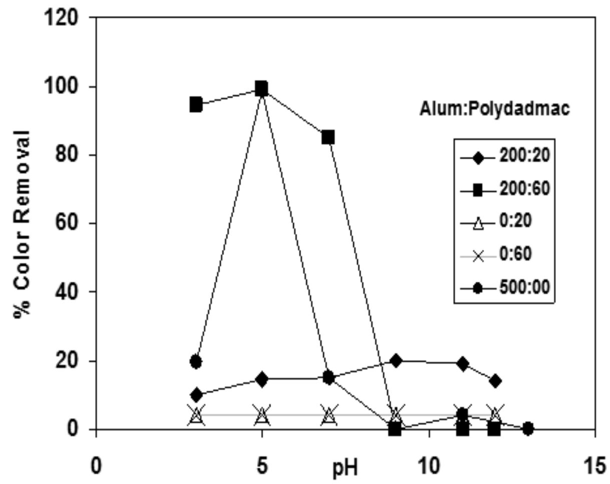

Fig 7. Effect of pH on % colour removal using alum and poly- DADMAC formulation

color removal at pH greater than 7.

However, as evident from Fig. 6, when PAC or poly-DADMAC alone are not effective, the formulation of PAC and poly-DADMAC works much better. Thus, color removal percentage is much higher with the formulation of PAC 200 ppm and poly-DADMAC 20 and 60 ppm as compared to individual coagulants. It is also to be noted that there is also modification in the operating pH range and the formulation works better in a wider pH range than that for individual coagulants. Another interesting observation, which can be made for the above formulation studies is that the effect modification is prominent in the later pH range which can be attributed to the contribution of poly-DADMAC since in this particular pH range PAC is largely ineffective.

Formulation of Alum with poly-DADMAC

It was observed earlier during preliminary experiments that inorganic coagulant, Alum is required in much larger quantity as compared to most coagulants, both inorganic and organic, although it is widely used in many industrial applications due to its lower cost. In developing formulation with alum, therefore, some trial experiments were required to obtain reasonably effective quantity of alum for coagulation. Using this information, a quantity of 200 ppm of alum was used in the formulation and quantity of organic coagulant, poly-DADMAC was varied from 20 ppm to 60 ppm.

The results with the two formulations developed using Alum and poly-DADMAC are clear indicative of how color removal efficiency can be enhanced by combining inorganic and organic coagulants. It should be noted that use of alum alone as a coagulant results in significant pH dependence for both coagulant dosage and percentage color removal. Alum dosage of 500 ppm would be required for effective color removal at pH of 5. Below and above pH 5, the color removal with Alum alone even with this high dosage is highly inefficient. However, it can be seen from the above Fig. 7, the developed formulation with lowered quantity of alum and with poly-DADMAC concentration of only 60 ppm results in a substantial enhancement in terms of colour removal efficiency and also in terms of increased pH range (pH 3-8) for its application. The developed formulation is thus, far better than the Alum alone or poly-DADMAC alone in removing reactive blue dye from wastewaters.

The comparison of the two poly-DADMAC formulations, one with PAC and other with Alum can not be avoided. However, although their performance compares well, the concentrations are different with inorganic coagulants and thus, the selection of such formulation would be mostly on the basis of economics of the process for similar performance.

Formulation of Alum +PAC + poly-DADMAC

The results of development of binary coagulant system clearly indicate trends in the modification of range of pH for operation and also possibility of lowering dosage of individual coagulant in combined coagulant systems.

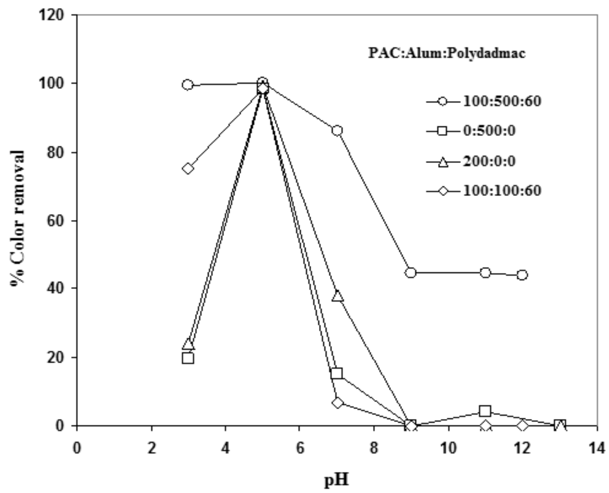

Fig 8. Effect of pH on % colour removal using alum, PAC and poly- DADMAC formulation

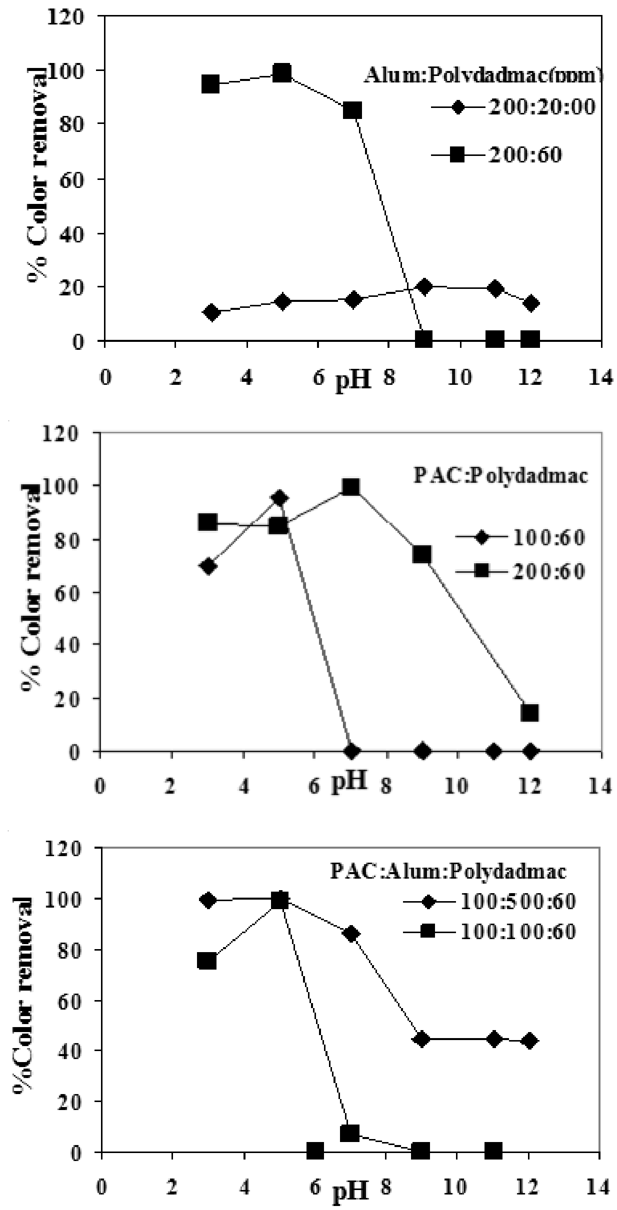

Fig. 9 pH range modification in 2-component and 3-component formulations

Performance behavior is, however, not straightforward and the approach is empirical. It would be however instructive to study effect of adding third component in the formulation. The results of Fig. 8 show such three-component formulation behavior for the removal of reactive blue dye. The formulation of PAC and poly-DADMAC (100 ppm and 60 ppm) works only in the range of pH 3-5 with maximum color removal at pH 5 similar to that obtained with only PAC (but at higher concentration), while the formulation of PAC, Alum and poly-DADMAC (100 ppm of inorganic and 60 ppm of poly-DADMAC) only slightly modifies the color removal behavior. However, with little increased concentration of Alum in the three component formulation (alum 500 ppm) changes the color removal behavior significantly and the formulation now works in the wider pH range of 3-12 with high color removal efficiency in the pH range 3-7, which is not the case with individual inorganic coagulants.

Discussion

Reactive dyes usually have more number of functional groups such as azo (N=N), sulfonic acid (-SO3-), hydroxyl (-OH) etc. Thus, these dyes are more stable apparently due to charge distribution, which could be one reason why reactive dyes are more difficult to remove using conventional coagulation. Thus, we need here a system or formulation for effective charge neutralization. In this respect, our results have clearly indicated improvements in the color removal efficiency by combining two or more coagulants for better utilization of coagulant quantity and also for widening pH range for the application of coagulant system. Gao *et al*. (2007) have also reported some studies on formulation of poly ferric chloride and poly-DADMAC for removal of reactive blue dyes and disperse blue dyes. Their results indicated dosage of coagulant formulation similar to that shown in our studies. In this respect, it is possible to conclude from our studies that PAC or Alum based formulations are comparable to that ferric chloride based formulations.

The results of this work clearly indicate that use of coagulant formulation is far superior to using individual coagulant in removal of dyes from dye wastewater. In this case, the advantages of both inorganic and organic coagulants can be incorporated in formulation to a great extent. In fact, the recent trend in the industry has been to use blends of inorganic and organic coagulants to optimize the treatment program. Although such formulations are available in the market for specific applications, the research on these formulations is lagging far behind and many aspects involved in making such formulations are less understood. Our research in this work is expected to provide impetus in this regard.

CONCLUSION

The conclusions from the present study are,

1. The research on the use of inorganic coa'ulants (polyaluminum chloride and Alum) confirms the finding of the previous pesearchers on pH sensitivity. The inorganic coagulants were found to be highly sensitive to pH as far as removal of dyes is concerned and optimum pH for most c`ses was found to be pH 5. Thus, these coagulants mostly work in limits pH range.
2. The color removal efficiency was also found to be strongly dependent on the dosage of the coagulant. The optimum dose of a particular coagulant was, however, dictated by the dye type. In general, dose of inorganic coagulant was more as compared to organic coagulant-poly DADMAC. Further, in the same class of inorganic coagulant, optimum dose of Alum was far higher than that of polyaluminium chloride.
3. PAC concentration of the nrder of 100- 200 ppm is required for the removal of reactive dyes for good color removal.
4. As compared to inorganic coagulants, the organic coagulant, poly-DADMAC and polyamine have shown negligible sensitivity to pH for almost all dye types. These finding are in agreement with those reported in the literature.
5. The optimum concentration obtained with organic coagulants is generally less as compared inorganic coagulants and is 100 and 25 ppm for poly-DADMAC and polyamine respectively. Further, as compared to PAC, the sensitivity to coagulant concentration was much high in case organic coagulant.
6. When PAC or poly-DADMAC alone are not effective, the formulation of PAC and poly-DADMAC works much better. The color removal percentage with respect to reactive blue dye is much higher with the formulation of PAC 200 ppm and poly-DADMAC 20 and 60 ppm as compared to individual coagulants. Further, there is modification in the operating pH range and the formulation works better in a wider pH range than that for individual coagulants. Another interesting observation, which can be made for this formulation is that the effect modification is prominent in the later pH range, which can be attributed to the contribution of poly-DADMAC since in this particular pH range PAC is largely ineffective.
7. The results with the two formulations developed using Alum and poly-DADMAC are clear indicative of how color removal efficiency can be enhanced by combining inorganic and organic coagulants. The developed formulation with lowered quantity of alum and with poly-DADMAC concentration of 60 ppm results in a substantial enhancement in terms of color removal efficiency and also in terms of increased pH range (pH 3-8) for its application to the removal of reactive dyes. The developed formulation is thus, far better than the Alum alone or poly-DADMAC alone in removing reactive blue dye from wastewaters.
8. The three component formulation attempted in this study for the removal of reactive blue dyes was found to change the color removal behavior significantly and the formulation was found to work in wider pH range of 3-12 with high color removal efficiency in the pH range 3-7, which is not the case with individual inorganic coagulants.
9. The results of this work clearly indicate that use of coagulant formulation is far superior to using individual coagulant in removal of dyes from dye wastewater. In this case, the advantages of both inorganic and organic coagulants can be incorporated in formulation to a great extent.

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