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**S Jindal, R C Singh**
**Coagulation and flocculation by polyelectrolytes**

INTRODUCTION

Coagulation is an important unit process in water treatment. The primary purpose of coagulation is to reduce the turbidity of water to the point where sand filters can remove the balance without excessive clogging. Another purpose of the coagulation is reduction in bacteria, viruses and other microorganisms as well as various specified and unspecified dissolved solids. Coagulation can reduce the total bacterial count by 65 percent (1-2). Efforts must be made to improve it further because it is desirable that terminal chlorination has to deal with as small number of microorganisms as possible. In fact it should be regarded as a final safe guard, rather than a method of removing organisms from water. The unsatisfactory state of affair in water treatment in developing countries like India is occurring on account of the fact that process preceding the chlorination, like coagulation and filtration, are neglected in the found hope that chlorine will take care of all the organisms.

The conventional metal salt coagulation is being substituted by polyelectrolytes for the reasons that these are more efficient, give a very tough floc resulting in an increased settling and filtration rate with reduced fragmentation of the floc, sludge produced is thick, compact and less in volume which is easier to handle. The cost of the chemicals are expected to be half (3-5). Alum sludge alone is posing a great problem for disposal all over the world. Till now the use of synthetic polyelectrolytes in the treatment of water has not been able to gain grounds in the developing countries due to their non-availability and fear that these may be toxic and water treated with them may be harmful for the human consumption. But now-a-days standards have been laid down by International authorities such as WHO, EPA, of the USA etc. so that they may be used without harmful effects to

human beings (6,7). A recent survey shows that all the three types of polyelectrolytes i.e. cationic, Anionic and Monionic are in use in water treatment in technically advanced countries like U.S.A. (8) as a primary co-agulants or coagulant aids (9-12). To obtain the maximum removal of colloidal and suspended particles through the coagulation, use of polyelectrolytes is a very successful solution. In the words of Adih, Bahuman and Cleasby (13)-

"The conclusion which may seem heretical to some, is that alum is unlikely to be used as a sole flocculant in modern treatment systems, which obviously tend to use higher rates than in past and want to reduce their sludge disposal problems. The use of polyelectrolytes either alone or in combination with alum has become inevitable".

The use of polyelectrolytes enables production of water of very low turbidities (0.1 JTU) which has a great public health importance in the inactivation of virus by chlorination (14). Besides this the treatment capacity can be increased tremendously by 100-200 percent (15).

In view of the above circumstances it has become of vital importance to evaluate the efficiency of polyelectrolytes vis-a-vis metallic coagulants in removing turbidity and bacteria, so that the effect of substitution of polyelectrolytes in place of metallic coagulants in water treatment may be judged in this respect. Till now very information is available for the effect of these synthetic polyelectrolytes on bacteria.

The purpose of the present work is to study the relative efficiencies of the conventional coagulant i.e. alum and polyelectrolytes for the removal of bacteria and turbidity by flocculation in chemically defined water and the effects of coagulant dosages, pH, initial concentration of bacteria and external turbidity on removal efficiencies of these two and to

extend the existing knowledge of the mechanism(s) involved in removal of bacteria from water by coagulation and flocculation.

#### MATERIALS AND METHODS

E.Coli was selected as the model bacteria for this study because it is the indicator of bacteriological pollution of water.

Pure E.Coli strain was obtained from the Deptt. of Microbiology, AIIMS, Delhi. It was cultured on MacConkey's Agar slants at  $37^{\circ}\text{C} \pm 1^{\circ}$  for 24 hr.  $\pm 1$  hr. The cells were taken into 0.85 percent sterile saline solution and centrifuged for 10 minutes at 5,000 rpm. Cells were washed 2-3 times to wash off the medium adhering to the cells membranes. The centrifuged cells were resuspended in sterile deionized water and were diluted in different concentrations as required at the time of experiments according to their optical density. A standard curve was drawn for the relationship of bacterial enumeration was done by standard plate count method on nutrient agar plate (16). The plates were done in triplicate.

Reagents : All the three polyelectrolytes is cationic, MAGLOC 851, anionic MAFLOC 900 & nonionic MAFLOC 720 were obtained from MAZER chemical s Inc, Illi, U.S.A.. These all were EPA of USA approved for the use in potable water. Alum of BDH was used. Stock 1% solution of the polyelectrolytes were prepared and diluted at the time of experiments. Alum solution of 10 mg/ml was prepared every week. Kaolinite earth clay was used as the source of external turbidity. The clay was suspended in the deionized water and stirred for 6-8 hrs. after which it was left for 24 hrs. & the supernatant was collected. It was again diluted with deionized water as required at the time of experiments. pH was adjusted by 0.1 N  $\text{H}_2\text{SO}_4$  and 0.1 N NaOH. All reagents used were of analytical grade of BDH. All the solutions were sterilized before conducting the experiment. All the glassware were cleaned with chromic acid and Teepol solution. Finally these were rinsed with deionized water and sterilized in an hot air oven at  $200^{\circ}\text{C}$  for 2 hrs.

Coagulation studies were performed using a Jar test apparatus on the following types of the samples - (a) Controlled E.Coli suspension (early log growth phase) (b) Synthetic turbidity with Kaolin (c) Mixed suspension of E.Coli & Kaolin. The experimental parameters varied to study their effects on the removal efficiencies were (i) Dosage of the coagulants (ii) pH (iii) External turbidity (iv) Initial input concentration of bacteria.

Jar test were conducted for all the experiments by flash mixing for 2 minutes at 100 rpm, flocculation for 20 minutes at 10 rpm & quiescent settling for 30 minutes. Samples for the bacterial enumeration, turbidity & pH measurements were withdrawn by a pipette at the middle of the supernatant after the quiescent settling. The first beaker in all the experiments was kept as the control & no coagulant was added. Readings of the control were used to compute the percent removal of bacteria & turbidity.

The optimum dosages of the alum & polyelectrolytes were obtained by the coagulation studies using a pH range of 5 to 9.

The effect of input bacterial conc. on the removal efficiencies of these coagulants was studied at the optimum dosages & optimum pH observed. Effect of external turbidity was observed using the constant bacterial input concentration in between 270-280 colonies/ml to avoid the enumeration problem as well as to avoid experimental mistakes due to low counts.

To find out whether the removed bacteria could be regenerated, settled flocs were resuspended in sterile deionized water & stirred for 2 hrs. Samples were pipetted out for the bacterial enumeration.

In a definite bacterial concentration (280/ml.) different dosages of polyelectrolytes were added, the samples were mixed very gently & incubated at  $30^{\circ}\text{C}$  for 24 hrs. to observe if toxicity is associated with polymers or the monomer. After incubation plate count was done to enumerate the bacterial conc.

#### RESULTS AND DISCUSSION

Fig.1 & 2 show the percent removal

of coliform bacteria & turbidity by alum, Fig. 3 & 4 show the percentage removal of bacteria & turbidity using cationic polyelectrolyte at different pH & different dosages.

In case of alum it was found that 45-50 mg/l is the optimum dose for the maximum removal of bacteria & turbidity both & for the best floc formation. Maximum removal of bacterial was 88% at lowest of the pH range studied i.e. 5-2. It is observed that pH has got an pronounced effect on removal efficiencies in case of alum. As pH increases, percentage removal decreases & beyond pH 7.0 there is a sharp reduction in percentage removal of bacteria & turbidity both (as in fig. 1,2). A reduction from 88% at pH 5.2 to 80% at pH 8.2 & further to 60% at pH 9.2 is observed. The turbidity removed by alum was 96% at pH 5.2. It reduces to 82.5% at pH 8.2 & further to 71% at pH 9.2.

In the case of polyelectrolytes it is observed that bacterial removal exceeded the turbidity removal. Effect of pH is rather small. Neutral pH is the optimum pH value where we get the maximum percentage removal. As the pH increases or decreases the percentage removal decreases but the decrease is more pronounced in acidic range. In the case of polyelectrolytes exact monitoring of the dosage is very essential, beyond the optimum dose the percentage removal of bacteria & turbidity starts decreasing (as in fig. 3,4).

It is observed that only the cationic PE is of promising use out of all the three tried. In the case of cationic PE the optimum dose is 0.45-0.5 rpm at pH 7.2 as shown in fig.3. It removed greater percentage of bacteria 94.5-95.0% while only 87.5% of turbidity is removed. Floc obtained by this polyelectrolyte (PE) was very big, tough having a 50% less settled volume as compared to alum, which is one of the great advantages of PEs use.

Monionic PE also gives good removal but the dose required is very high as compared to cationic PE. while the amonic is completely non-effective in the dosages range tried. Higher dosages were not tried because it is not economical as well as it affects the viscosity of water. Fig. 5 & 6 show the percentage removal of bacteria & turbidity with monionic &

amonic PEs at pH 7.2 at different dosages. It is observed that monic PE can remove 76% of the bacteria & 73.5% of turbidity & amonic PE could not go beyond 7.5% removal of bacteria and was totally noneffective in removing turbidity. From fig. 7 & 8 it is inferred that the presence of external turbidity has got a remarkable influence in all the cases. More presence of 50 NTU of external turbidity increased the percentage removal of bacteria from 88 percent to 97 percent at pH 5.2 in the case of Alums & from 85 to 98% in case of cationic PE. The same was applicable to nonionic PE too, reaching up to 80 percent from 76 percent. In our studies higher external turbidities were also tried and it was found that the efficiency of the removal was further increased but the increase is not so significant.

Further the experiments were conducted to observe the effect of initial bacterial concentration on removal efficiencies because different waters may have different bacterial concentration. It is observed that the percentage removal increased as the initial concentration was increased but the optimum dose was also increased a little bit as shown in the table No 1. The effect is more pronounced in the case of cationic polyelectrolyte.

The resuspension of the floc gave a recovery 70.75% in the case of alum while very less could be recovered in case of polyelectrolytes.

It is also observed that the PEs are neither toxic nor nutritive to the bacteria in the range used.

#### MECHANISM INVOLVED IN REMOVAL OF TURBIDITY :

The removal pattern of bacteria using alum agrees with the previous studies (17). It is observed that in the case of alum, bacterial removal paralleled the turbidity removal. Alum undergoes rapid hydrolytic reactions and form hydroxy products in the pH range 5-6, which form Aluminium-Colloids precipitate which aggregate to form flocs thus removing the turbidity and bacteria. It is obtained in our experiments that the dosages of alum depended more on the turbidity than on bacterial counts. Higher percentage removal of bacteria at higher dosage can be explained on the basis of

trapping between the flocs of turbidity.

The interaction of bacteria-Al(III) is superficial, it may be adsorption or a very low energy chemical bond (if at all it is formed) so that it can be broken by stirring only as 70-75% bacteria could be recovered as in Table (2).

Polyelectrolytes remove the turbidity by either the (i) charge neutralization or (ii) by bridging in between the turbidity particles and the segments of the polymer chain (18). Both the mechanisms may be operative together depending on the conditions. In bridging the polymer chain is adsorbed on the bare surface of the particle and bridges between them with the aid of free segments of the adsorbed polymer to form a three dimensional agglomerates of sufficient size to be settleable.

SPECIAL MECHANISM PROPOSED FOR THE BACTERIAL REMOVAL BY POLYELECTROLYTES :

It is observed that percentage removal of bacteria is higher than the percentage removal of turbidity which shows that the bacteria are not acting just as a colloidal particle and their biological structure has got some influence on flocculation by PEs. The surface of the bacteria is composed of a variety of proteins, lipids, polysaccharides and nucleic acids in the form of cell wall, capsules, flagella. The composition of this surface changes as their physiological conditions alter, thus it is very difficult to give a confirmatory mechanism(s) for the cell flocculation by PE.

It can be postulated that the polyelectrolytes added as flocculant is interacting with the cell surface with any of the groups present on the cell wall components. The interaction may be either a chemical bond formulation or a co-ordinate complex formation. As the recovery of bacteria is very less from the performed floc leads to believe that there is a strong interaction.

Once the PE interact with the surface of bacteria the other branches of the polymer chain can interact with the bare surfaces of the available bacteria resulting in a big floc.

The cationic PE were able to flocculate bacteria more efficiently, it appears that in the interaction of PEs with cells, electrostatic forces help in flocculation by bonding the positively charged segments of polyelectrolyte to the bacteria surface. The hypothesis confirms the uneffectiveness of anionic PE in removing the bacteria. There may be some charge repulsion which inhibits this polyelectrolyte bacteria surface interaction or the polymeric materials excreted or exposed at the surface of bacteria may be interfering with the anionic PE and preventing the complex formation resulting in non flocculation. The mechanism applies to monionic PE also explaining why we get the removal by this PE.

Requirements of higher dosages of PE for the optimum removal of bacteria in higher co input concentration further supports the mechanism. It can be assumed that a certain fraction of the surface of the cells has to become covered with extended polymer segment and as more of the surface material is there, more dosage would be needed for the complex formation.

Phenomenon of Overdosing : Can also be explained on the basis of the above proposed mechanism. The excessive amount of the polymer will cover the bacterial cell completely by complex formation, making a single cell stabilized and leaving no bare surface for the floc formation.

Work is in progress to confirm this mechanism by isolating the bacterial component which interact with PE.

CONCLUSIONS

The following conclusions can be drawn from the paper:

- (i) Cationic polyelectrolytes are the most efficient flocculants in removing bacteria.
- (ii) Removal of bacteria is more as compared to turbidity by cationic polyelectrolyte while in case of Alum more turbidity can be removed.
- (iii) The optimum pH for the polyelectrolytes is 7.2 which is an added advantage, no pH adjustment has to be done.

- (iv) Optimum dose of cationic PE is very less as compared to Alum thus saving the cost of chemicals.
- (v) Nonionic and anionic PEs are of little use for the removal of bacteria.
- (vi) The only disadvantage of PE being one should be very careful while dosing and must follow the International standards.
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TABLE 1 : EFFECT OF 'INPUT' BACTERIAL CONCENTRATION ON REMOVAL EFFICIENCY

S. No.	Name of the coagulant	Conc. of bacterial input in Col./	Optimum dose	% removal obtained
1.	Alum	50	40	86.5
		100	40-42	87.0
		250	42-45	88.0
		500	50	88.5
2.	Cationic Polyelectrolyte	50	0.35	90
		100	0.40	92.5
		250	0.45	95.0
		500	0.55	98.0

TABLE 2 : RECOVERY OF BACTERIA

Name of the Coagulant	Recovery after $\frac{1}{2}$ hr. stirring
Alum	72-75%
Cationic PE	15-17%
Nonionic PE	10-15%
Anionic PE	Could not be performed

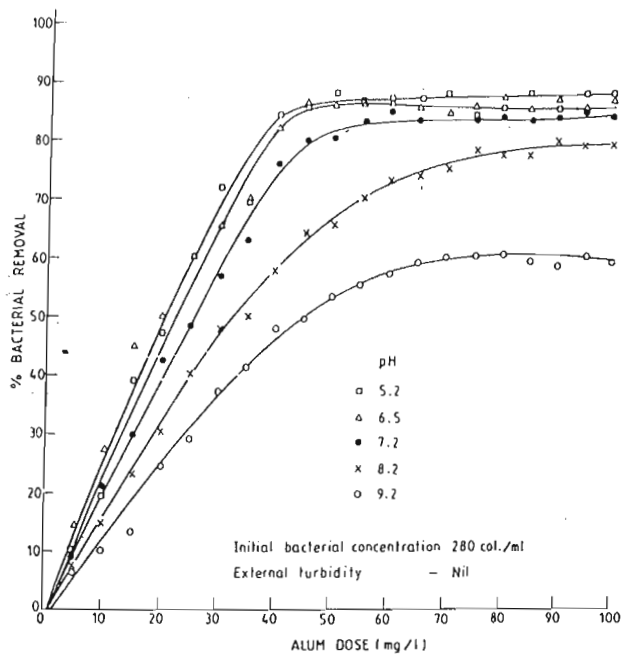


FIG.1. REMOVAL OF BACTERIA AT DIFFERENT pH AND WITHOUT TURBIDITY USING ALUM AS A COAGULANT

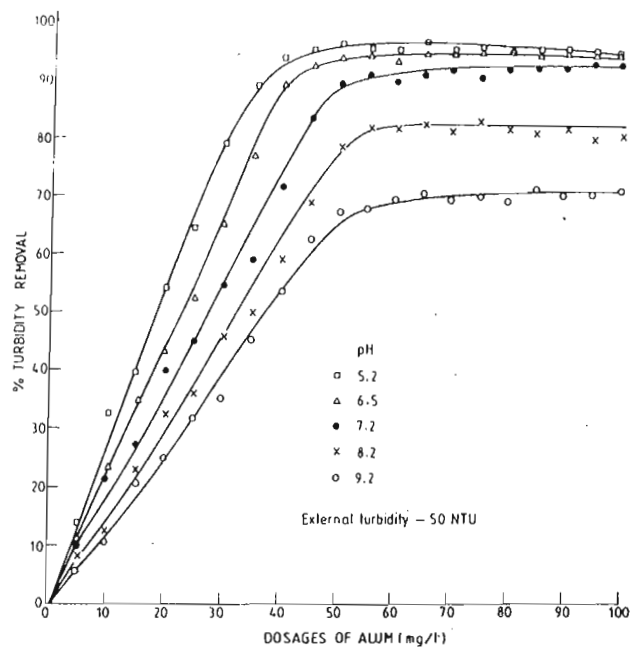


FIG.2. REMOVAL OF TURBIDITY AT DIFFERENT pH AND DIFFERENT DOSAGES OF ALUM

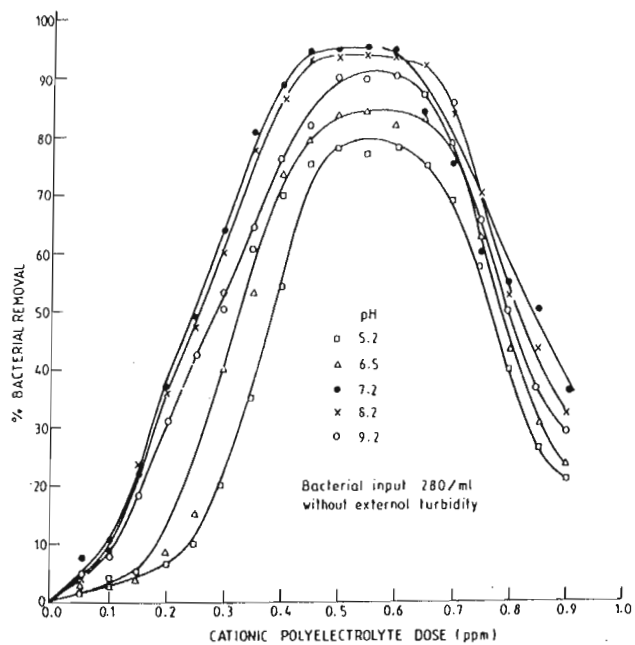


FIG.3. REMOVAL OF BACTERIA AT DIFFERENT pH USING DIFFERENT DOSAGES OF CATIONIC PE

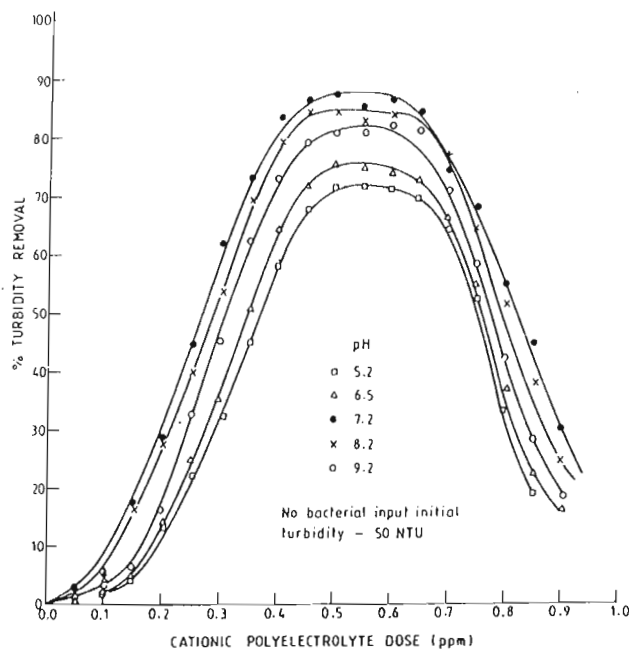


FIG.4. REMOVAL OF TURBIDITY AT DIFFERENT pH AND DIFFERENT DOSAGES OF CATIONIC POLYELECTROLYTE

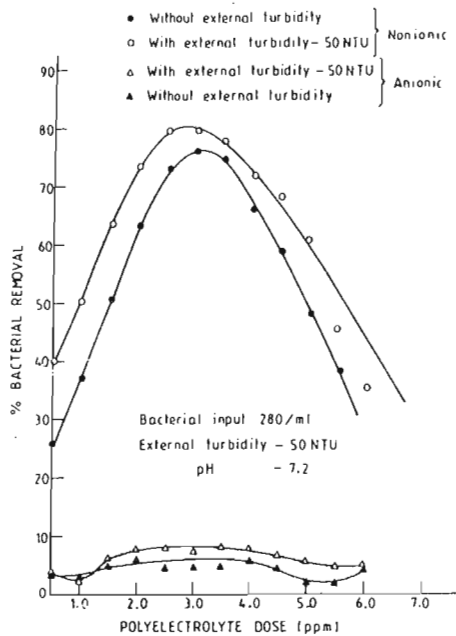


FIG. 5. REMOVAL OF BACTERIA USING NONIONIC AND ANIONIC POLYELECTROLYTES AT DIFFERENT DOSAGES

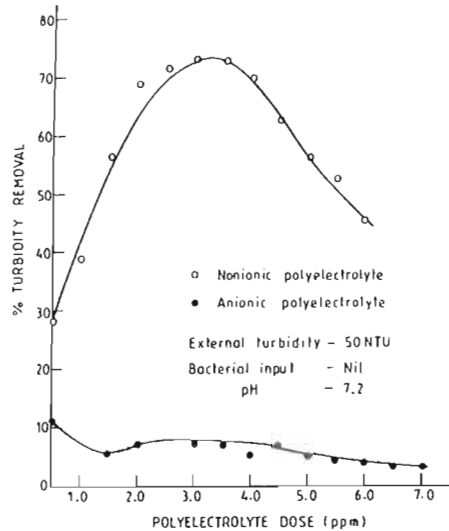


FIG. 6. REMOVAL OF TURBIDITY AT DIFFERENT DOSAGES OF NONIONIC AND ANIONIC POLYELECTROLYTES

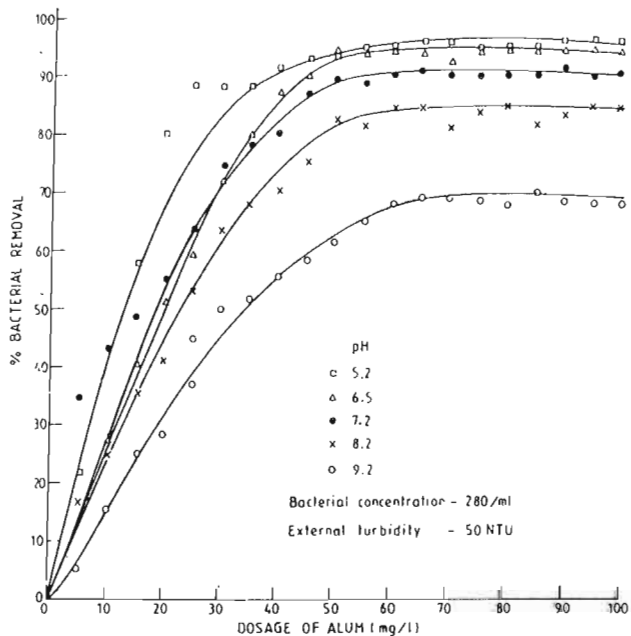


FIG. 7. REMOVAL OF BACTERIA AT DIFFERENT pH AND WITH EXTERNAL TURBIDITY OF SONTU USING ALUM AS A COAGULANT

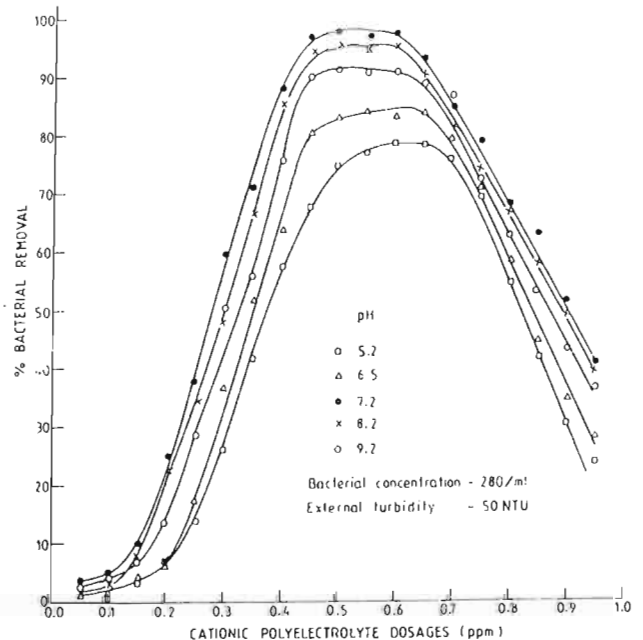


FIG. 8. REMOVAL OF BACTERIA AT DIFFERENT pH AND DIFFERENT DOSAGES USING SONTU OF EXTERNAL TURBIDITY