Water purification and demineralization technology

Muxammadali Abdullayev[1], *Sherali* Juraev[2*], *Yoqutxon* Yoqubjanova[2], and *Axror* Akramov[3]

[1]Samarkand State University of Architecture and Construction, 140147 Samarkand, Uzbekistan
[2]Namangan engineering constriction institute, 100000 Namangan, Uzbekistan.
[3] Ferghana Polytechnic Institute, 150107 Ferghana, Uzbekistan

Abstract. The paper has investigated flocculation capacity of polyhexamethylene guanidine hydrochloride during joint treatment with aluminum sulfate of the Narin and Sirdarya River water in the spring period. It has been shown that additives 1–5 mg/dm^3 of the matter under study during coagulation treat- ment of the river waters improves the process of their clarification, increases the filtrate quality in terms of main indicators and makes it possible to reduce by 10–20 mg/dm^3 the dose of the coagulant. A pronounced flocculation effect manifests itself when treating the waters in question by the doses of a polymer preparation respectively 3–5 and 2–3 mg/dm^3. Polyhexamethylene guanidine (without a coagulant) ensures a standardized quality of the filtrate in terms of color and turbidity only when treating weakly stained water of the Sirdarya River. It has been found that ≥ 99% of polyhexamethylene guanidine is retained by the medium of a sand filter.

1 Introduction

The joint employment of coagulants and flocculants intensifies the operation of treatment facilities and increases the quality of the clarified water. Small additives of flocculants substantially reduce stability of dispersions, speed up the forma- tion and settlement of the flocs of aluminum or iron hydroxides, increase the density of sediments and the clarification degree of the water. At present synthetic organic flocculants (nonionogenic, anionic and cat-ionic) have been widely used. Lately one can observe an increased interest in synthetic bactericidal polyelectrolytes based on guanidine compounds simultaneously manifesting properties of the flocculants of the cationic type [3, 4]. However, the impact of such a type on the drinking water quality has not been sufficiently studied [3, 5].

The objective of the given investigation is an assessment of flocculating properties of Polyhexamethylene guanidine hydrochloride (PHMG- $(C_7H_{15}N_3)_n$) during coagulation treatment of the water of the Narin and Sirdarya Rivers.

* Corresponding author: jorayev1981@mail.ru

2 Materials and methods

We have studied the impact of additions of both (polyhexamethylene guanidine hydrochloride) PHMG alone and together with $Al_2(SO_4)_3$ treatment in combination with the subsequent settlement and filtration on the treatment degree of river waters by a complex of indices. We monitored the following: turbidity (T), residual concentrations of aluminum and PHMG in the settled water; color (C), cloudiness, residual concentrations of aluminum and PHMG, permanganate oxidiz ability (PO), COD, the content of total organic carbon (TOC) in the filtrate.

The study of the flocculation properties was conducted on several samples of the Narin and Sirdarya waters sampled in April–May 2019-2022 (Table 1). Cloudiness of the initial Narin water during the period of investigations was much lower than that of the Sirdarya River water, however, an extremely fine disperse suspension was weakly retained both by paper and sand filters.

The concentration of the reagent was varied within the interval of doses necessary for initial disinfection of the waters being studied (1–5 mg/dm^3) [10]. The PHMG doses in question were of the same order that of other low-molecular flocculants of the cationic type (e.g., VPK-402) [9]. Sulfate aluminum was used as coagulant. The concentration of AS working solutions (in terms of $Al_2(SO_4)_3$) and PHMG are respectively 10 and 1 g/dm^3. The procedure of treating waters with a coagulant and flocculant and the time interval between their introduction corresponded to the mode of water treatment accepted at national water stations. The water settlement time is 3 h, which is in line with the data of [5].

Table 1. Characteristics of river waters being investigated.

Water	Sam- pling date	T, °C	pH	Alkalin- ity, mg- eq/dm3	Turbid- ity, mg/dm3	Color, deg	PO	COD	TOC, mg/dm3	Coli- index, CFU/dm3	Total microbial number, CFU/cm3	PHMG dose for initial dis- infection, mg/dm3
							mg O/dm3					
Sirdarya River	04.04.23	9	8.4	4.3	7.8	27	8.5	24	13.0	93	21	1.5–2.0
Narin	11.04.23	9	7.8	2.2	4.5	60	15.0	32	15.7	636	35	> 3.0
River	25.04.23	12	7.8	2.3	3.8	62	15.4	42	15.9	280	12	4.0
	10.05.23	15	8.0	2.3	4.5	79	19.7	55	20.0	180	20	–

An assessment of the impact of the PHMG additives on the degree of treating river waters by different doses of $Al_2(SO_4)_3$ was carried out by two methods. According to the first method a series of water samples (V–0.5 dm^3) in 10 cylinders with ground stops was treated by different doses of $Al_2(SO_4)_3$, stirred by 5 – 10 fold tilting of closed cylinders 3 min later design volumes of the PHMG working solution were added, again stirred, settled for 3 h and filtered through a paper filter (white ribbon). According to the second method large volumes of river water (5 dm^3) were treated in the same way; in this case the settled water (3 h) was separated by means of a siphon through a sand filter. The sand filter is a glass funnel (diameter 3.8 cm, height 1 m) with a glass plate soldered in the lower part with holes and filled with granite chippings (grain diameter 3- 5 mm, medium height 60 mm). The water was filtered from top to bottom at the rate 5–6 m/h, the filter was washed with tap water from bottom to top.

3 Results and discussion

The process of formation and sedimentation of flocs of aluminum hydroxides in the waters under investi- gation at low temperature (11–15°C) proceeds slowly both with the treatment

with only AS and in the presence of PHMG. Though the formation of the flocs in the presence of PHMG was accelerated complete pre- cipitation of the flocs within 3 h was not achieved and rather high turbidity of the settled water held on even with $Al_2(SO_4)_3$ doses (D) 60–80 mg/dm^3 (Figs. 1a, 2a). The turbidity of the settled Narin water diminished com- pared with the initial one only with the treatment of doses of the coagulant and PHMG respectively 60 and 3–5 mg/dm^3. The analysis of residual concentration of aluminum in the settled waters (see Figs. 1b, 2b) demonstrated that an addition of 2–5 mg/dm^3 of PHMG with AS doses 40–60 mg/dm^3 increased the degree of aluminum hydroxide flocs' sedimentation in the Sirdarya and Narin River waters respectively by 13–39 and 20–45%. The degree of the PHMG influence on the process of the settling of the waters in question depended on the doses of the coagulant and flocculant. With the PHMG dose 1 mg/dm^3 the flocculating effect was unstable (see Fig. 1). A more pronounced flocculating effect revealed itself during the treatment of the Narin and Sirdarya River waters with the PHMG doses respectively 3–5 and 2–3 mg/dm^3. Macromolecular flocculants of the cationic type are capable to cause aggregation and sedimentation of negatively charged particles of colloids and suspensions without coagulants. Binary additives of cationic type polymers are also used (both low- and macromolecular) [9]. In this case an additive of a macromolecular floculant (~5×10^5 amu) destabilizes the particles of humus matter, while a macromolecular component (up to 15×10^6 amu) is conducive to the formation of large aggregates easily separated from the liquid phase by sedimentation and filtration.

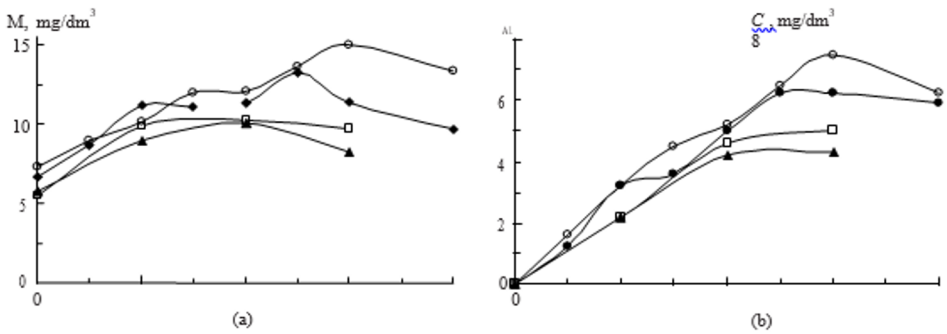

Fig. 1. Variation of the turbidity of the Sirdarya River water (a) and residual concentrations of aluminum (b) in it after treatment of different doses of aluminum sulfate and PHMG and settlement within 3 h. The PHMG doses, mg/dm^3: 0 (1), 1 (2), 2 (3), 3 (4).

The treatment of the waters under investigation by the PHMG alone (without $Al_2(SO_4)_3$) increased the turbidity of the settled water (Figs. 1 and 2) and noticeably decreased the values of all controlled parameters in the filtrate (Tables 2 and 3). For example, at the PHMG dose 3 mg/dm^3 the turbidity and color of the filtrate of the Sirdarya River water decreased respectively by 42 and 35%, the Narin onerespectively by 55 and 45% compared with the filtrate of untreated river waters. The PHMG consumption constituted 1 mg per 6–9 deg of color, which corresponded to the published data for other cationic flocculants [2]. However, in the absence of a coagulant color of the filtrate of the Narin River water exceeded the normalized one for drinking water even at the PHMG dose 5 mg/dm^3 (see Table 3). The filtrate quality corresponded to the standards o'zdst_950_2011of Uzbekistan [11] for drinking water by the indicated parameters when treating weakly-dyed water of the Sirdarya River by a dose of PHMG 3 mg/dm^3 (Table 2).

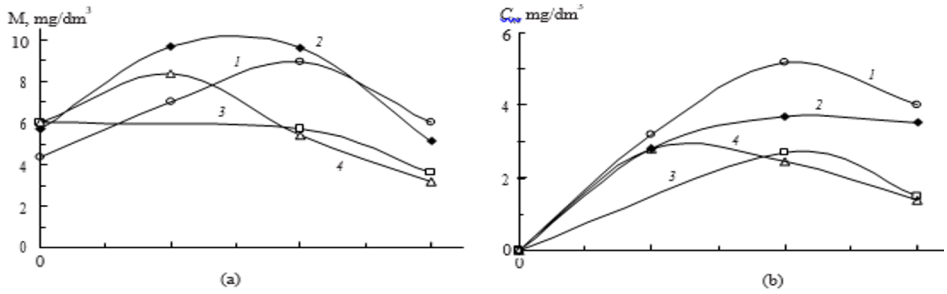

Fig. 2. Variation of turbidity of the Narin River water (a) and residual concentrations of aluminum (b) in it after treatment of different doses of aluminum sulfate ($Al_2(SO_4)_3$) and PHMG and settlement within 3 h. The PHMG doses, mg/dm^3: 0 (1), 2 (2), 3 (3), 5 (4).

The PHMG concentration in the residual Sirdarya River water constituted 50–70% of the introduced dose (1–3 mg/dm^3) without a coagulant, while with the joint treatment including a coagulant it varied 38 to 100%. The PHMG residual concentration in the filtrate (without treatment with a coagulant) equaled 0.27–0.50 mg/dm^3 (10–30% of the introduced dose); with the joint treatment of the water including the coagu- lant—it varied 0,12–0,18 mg/dm^3 (7–15%) to < 0,015 mg/dm^3 (< 0,5%) with AS doses respectively 10–30 and 40–80 mg/dm^3 (Table 2). Thus, similar to the flocculants of other types, PHMG by 99% was retained by the medium of the sand filter. In the settled Narin River water the PHMG concentration constituted 86–100% of the introduced dose (2–5 mg/dm^3 of PHMG) without a coagulant and 45–100% in combination with the treatment by the coagulant.

The PHMG residual concentration in the filtrate was varied < 3 to 20% of the introduced dose without a coagulant (20–60 mg/dm^3) and with the treatment including the coagulant it decreased up to < 0,1 mg/dm^3. A low residual concentration of PHMG in the filtrate, with the introduced dose 5 mg/dm^3 without a coagulant was determined, most likely, by sorption ability of aluminum hydroxide as part of deposits in the intergrain space of the filter medium with respect to (polyhexamethylene guanidine hydrochloride) PHMG since before several water samples were passed through the filter, the samples were treated together by both reagents.

Table 2. Degree of purifying the Sirdarya River water by different doses of aluminum sulfate and PHMG in combination with filtration through the sand filter (~ 6 m/h).

Dose of, mg/dm3		pH	Turbidity	C_{Al}	C_{PHMG}	Color, deg	COD	PO	TOC, mg/dm^3
coagulant	PHMG		mg/dm^3				mg O/dm3		
0	0	8.0	1.6	< 0.02	–	24	21	7.3	10.9
	2	7.9	1.5	< 0.02	0.50	23	19	6.9	8.9
	3	7.9	0.9	–	0.30	16	17	5.7	8.6
20	0	7.8	0.9	0.29	–	19	18	5.7	7.1
	2	7.7	0.5	0.05	< 0.10	13	16	5.3	6.9
40	0	7.5	0.9	0.19	–	14	16	5.3	5.6
	2	7.4	0.8	0.08	< 0.015*	11	15	4.0	5.5
60	0	7.2	0.8	0.05	–	10	14	4.9	5.1
	2	7.3	0.3	0.03	< 0.015*	8	12	4.1	5.0

Note: *By the method of solid-phase photometry [6].

With such a treatment of the waters being investigated by the coagulant and PHMG the filtrate quality by main indicators (turbidity, color, residual concentrations of aluminum) conspicuously improved compared with the treatment by one coagulant even with least

studied doses of PHMG-1 and 2 mg/dm^3 respectively for the Sirdarya and Narin River water (Figs. 3 and 4). The degree of filtrate purification by the said parameters increased respectively by 17–40; 4–18; 25–65% and 50–61; 9–52 and 79–94%.

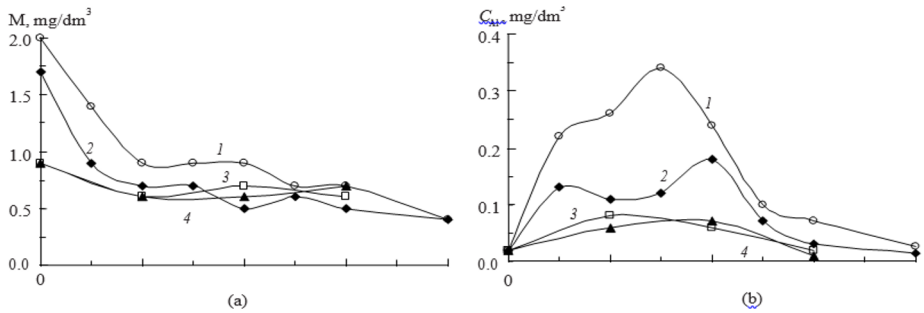

Fig. 3. Variation of turbidity (a) and residual concentration of aluminum (a) in the filtrate of the Sirdarya River water treated by different doses of aluminum sulfate and PHMG. The PHMG doses, mg/dm^3: 0 (1), 1 (2), 2 (3), 3 (4).

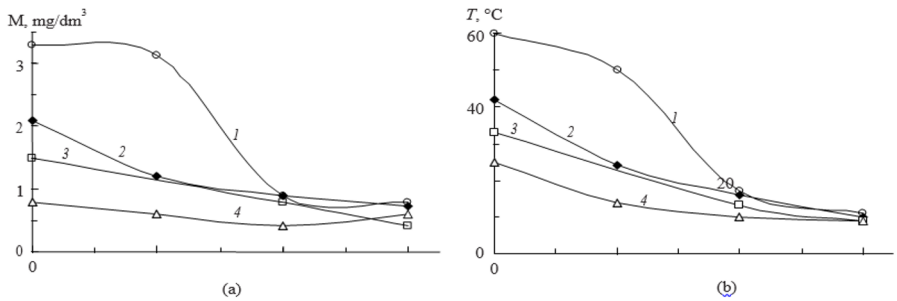

Fig. 4. Variation of turbidity (a) and color (b) of the filtrate of the Narin River water treated by different doses of aluminum sulfate and PHMG. PHMG doses, mg/dm^3: 0 (1), 2 (2), 3 (3), 5 (4).

An increase of the PHMG dose to 2 mg/dm^3 when treating the Sirdarya River water produced the greatest effect on the residual concentration of aluminum in the filtrate (see Fig. 3b) with a noticeable increase of its quality and by other indicators (see Table 2). Further increase of the PHMG dose up to 3 mg/dm^3 with the said doses of the coagulant did not improve the quality of the filtrate of the Sirdarya River water by organoleptic parameters and residual concentration of aluminum, however it was conducive to a more complete removal of organic impurities. At the same time,when treating the Narin River water an increase of the PHMG dose up to 5 mg/dm^3 with the doses of the coagulant 20–40 mg/dm^3 substantially increased the degree of filtrate purification in terms of organoleptic indicators (Fig. 4) and in terms of the parameters characterizing the content of organic impurities (Fig.5), but had a minor effect on residual concentration of aluminum. The treatment of the Narin River water by the coagulant dose 60 mg/dm^3 without PHMG additives decreased turbidity and color of the filtrate respectively by 76–83 and 79–82% compared with the filtrate untreated with water reagents and ensured compliance of its quality to the standards of SanPiNa N. 950/2011 for drinking water by the main indicators (color, turbidity, and residual concentration of aluminum) (Table 3). The PHMG additives with the coagulant dose in question decreased turbidity and residual concentration of aluminum in the settled water (Fig. 2) and slightly improved the quality of the filtrate in terms of the content of organic impurities (COD, PO, and TOC) (Fig. 5).

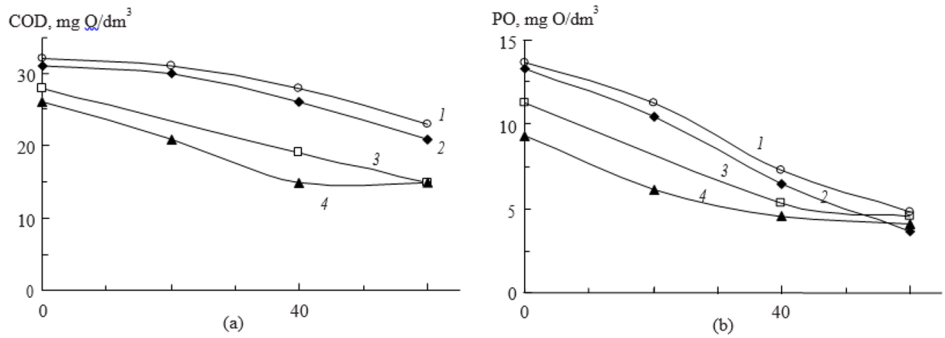

Fig. 5. Variation of COD (a) and permanganate oxidizability (b) of the filtrate of the Narin River water treated by different doses of aluminum sulfate and PHMG, mg/dm^3: 0 (1), 2 (2), 3 (3), 5 (4).

The values of COD, PO, and TOC of the PHMG solution in distilled water at the concentration 20 mg/dm^3 equaled 33 and 4 mgO/dm^3 and 8.4 mg/dm^3 at the design values of COD and TOC—respectively 28.8 mgO/dm^3 and 9.46 mg/dm3. Thus, at the dose of PHMG 1–5 mg/dm3 additional water pollution with organic impurities by COD, PO, and TOC increased respectively by ~2–8; 0.2–1.0 mgO/dm3 and 0.4–2.5 mg/dm^3. However, since residual concentrations of PHMG were retained by the filter medium virtually completely, its additions made it possible to slightly raise the degree of filtrate purification of organic impuri- ties compared with the filtrate of the waters under investigation treated by one coagulant (Fig. 5, Tables 2).

A decrease of the PO of the filtrate of the Sirdarya River water up to ≤ 4 mgO/dm^3 (O`zt № 950/2011 of Uzb [11]) was achieved under the following combinations of the doses of the coagulant and PHMG (mg/dm^3): 40 + 2; 60 + 2; while up to ≤ 5 mgO/dm^3 and guidelines 98/83/EC) [11, 12] at the following combinations of the doses of the coagulant and PHMG (mg/dm^3): 20 + 3; 40 + (2–3): 60 ± (0–3); 80 + 1. The PO of the filtrate of the Narin River water corresponded to the O`zt № 950/2011 of Uzb for drinking water only when treating the first sample of the water by doses of the coagulant and PHMG respectively of 6 and 5 mg/dm^3 (Table 3) and did not exceed 5 mgO/dm^3 at the following combinations of the doses of the coagulant and PHMG (mg/dm3): 40 + 5; 60 + (0–5). In the presence of the additions 2–3 mg/dm^3 of PHMG equal or better quality of the Sirdarya River water filtrate by most indicators was achieved at lower doses (by 10–20 mg/dm3) of AS, while those of the Narin River water- at the lower (by 20–40 mg/dm^3) dose of AS after addition of 5 mg/dm^3 of PHMG. Lower doses of PHMG did not make it possible to retain the filtrate quality at the lower doses of the coagulant[11-24].

When washing the sand filter with tap water we assessed the degree of removing PHMG from the filtrating medium, strength of PHMG binding by the dumping of the settled washing water to the river. The PHMG concentration in the settled washing water (V ~ 10% of the volume of the effluent) equaled 1.5–1.9 mg/dm^3.

Consequently, there is a probability of the ingress of the existing amounts of PHMG (in the given case 16–32% from 9.5–24.4 mg contained in 7–12 dm^3 of the settled Narin River water passed through the filter) to the environment.

4 Conclusion

Thus, PHMG in the water medium manifests the qualities of the cationic flocculant. The PHMG doses 1–5 mg/dm^3 necessary for initial disinfection of the Narin and Sirdarya River waters also improve the clari- fication process with the joint treatment with SA of river waters and conducive to an increase of the filtrate quality by the main indicators. However, the said

doses by 1–2 orders of magnitude exceed the recommended doses of effective macromolecular flocculants of the cationic type [9]. In the process of filtration 99% of PHMG are retained by the medium of the sand filter, which causes the necessity for the final disinfection of the filtrate. In the case of repeated treatment of the PHMG filtrate [3–5] for achieving normalized quality of the effluent by microbiological indicators the PHMG dose at least 1 mg/dm^3 is necessary [10], which may lead to secondary pollution of the effluent with organic impurities compared with an effect achieved due to its flocculating effect at the first stage of the purification. For preventing the ingress of PHMG to the environment it is necessary to control its content in the washing waters being discharged.

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