Application of Modified Wastes from Phenol-Formaldehyde Resin and Expanded Polystyrene in Sewage Treatment Processes

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Summary: A solution to environmental pollution by polymer plastic wastes can be their chemical modification in to useful products. Such a new solution is the obtaining of effective flocculants for sewage treatment from chemically modified phenol-formaldehyde resin production wastes (SE and NS novolak) and expanded polystyrene wastes. Comparative analysis of flocculation properties was performed for amino derivatives of novolak wastes, synthesized sulphonated derivatives of novolak and expanded polystyrene wastes, of standard polyacryloamide and for Praestol commercial polyelectrolyte. Amino derivatives and sulphonated derivatives of polymer plastic wastes, having properties of anionic type polyelectrolytes, exhibit good flocculation properties in purification processes of sewages with a chemical composition close to that of found in the water circulating system power plant, coal-mine, and steel plant. Application of synthesised flocculants caused a decrease of turbidity, concentration of solved impurities and improved quality parameter of purified water. It was found that synthesised polyelectrolytes could be used in industrial water treatment processes.

Keywords: functionalization of polymers; flocculation; phenol-formaldehyde resin wastes; polyelectrolytes; polystyrene wastes; sewage treatment; water soluble-polymers

Introduction

It is a well known fact, that storage of polymer plastic wastes is a serious global hazard to the environment. Perfect properties of plastics obtained from polymers as substrates are responsible for the fact that we are in permanent contact with polymers. Dumps and dumping grounds are full of polymer plastics, and their indestructibility previously regarded as an advantage has become their essential fault. Quantitative composition of polymer plastic wastes on dumping grounds approximately corresponds to the amount of produced polymer plastics considering their time of use. Dumping grounds consist mainly of wastes from

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packaging. In particular they are products made from expanded polystyrene of low density, and therefore of high capacity. Recently, the increase of the amount of polymer plastic waste processed into energy in combustion processes or processed into other useful wares is observed.[1-4] This is caused by intensive progress of studies on polymer plastic waste management and recycling of used polymer plastic ware, and particulary by law regulations in respective countries. Burdensome to environment are also faulty manufactured units, although the plants try to use them. Another problem is connected with useful products as, e.g., phenol-formaldehyde resins, which are produced to bind waste phenol and diminish its hazard to environment. This product, potentially useful, is stored on dumping grounds until it gets utilized. It may be said, that elaboration of polymer and polymer plastic wastes management technology is of great economic significance. Chemical modification of polymer wastes to polyelectrolytes can be a way of polymer and polymer plastic waste management. Nowadays the increase of environment pollution and enormous growth of power consumption in high-developed countries is observed. This led to the introduction of low-waste or no-waste technologies. Besides the elaboration of new technologies, such as used ware recycling in the mid eighties, it should be mentioned that from the beginning of 1970 intensive studies on waste usage as source of chemical and energetic raw material have been conducted in highly-developed countries. Utilization of polymer wastes instead of new synthesized polymers for ware production will limit material and energy consumption and pollutant emission to the environment. It is known, that polymer sorbents replace active carbons and unlike them can be regenerated with the use of an electrolyte solution or organic solvent.[5] The best known polymer sorbent on market is Amberlite produced by Rohm and Haas Co. However, this is a hydrophobic sorbent made from non-waste polymers and copolymers. Nowadays, the studies are conducted of the synthesis of sorbents (used mainly in sewage and water treatment processes) with the use of phenol-formaldehyde resin production wastes[6-8] and polystyrene wastes.[1-4] Modification of phenol-formaldehyde resins is essential due to the various kinds of pollution they form during the process of production waste storage. Influence of humidity, solar energy, acids, bases and other external agents, will cause inevitable transfer of phenol and formaldehyde to the environment. The formation of these wastes is independent of the production technology. Phenol-formaldehyde resin wastes are sometimes large manufactured units of novolak synthesis. They are the result of deflection from a process production parameter and as a result the obtained product properties are sub-standard. Management of these products is a serious problem for a production plant and hazard to the environment.. An advantageous solution is the modification of phenol-formaldehyde resins by chemical methods to products which can be used as flocculants. This solution saves raw material and energy and reduces environmental pollution. Highly diluted polyelectrolytes (mostly 0,1% to 0,01% solutions) used to aid flocculation in the sewage treatment process are transferred to the sludge and then are deposited in sedimentation lagoons or are used with sludge in the agriculture and power industry. To obtain effective polyelectrolytes, the synthesis of amino derivatives of phenol-formaldehyde resins, sulphonated derivatives of SE and NS novolak wastes, and expanded polystyrene wastes was performed. The synthesized compounds were utilized in the industrial sewage treatment processes. Comparative analysis of flocculation properties of synthesized polyelectrolytes from novolak and expanded polystyrene wastes, of synthesized standard polyacryloamide and of commercial Praestol-polyelectrolyte was performed.

Materials and Methodology

The novolak waste from "ERG" Plastic Factory in Pustków, nitric acid, sulphuric acid, hydrochloric acid, calcium carbonate, sodium carbonate, ammonia, stannic chloride and silver sulphate from POCh, Gliwice were used for chemical modification. Aluminium sulphate (POCh, Gliwice), as a coagulant (1% solution), commercial Praestol 2515 polyelectrolyte (anionic commercial polyelectrolyte syntesized from polyacryloamide[9]), Jaroszów clay for preparation of high turbidity water according to PN-71/C-04583 standard, coal mine water and blast-furnace circulation water from metallurgical plant were used in these studies.

Nitro derivatives of novolak were obtained by nitration of linear structure novolak with nitrating acid HNO_3/H_2SO_4. The amino derivatives were obtained by reduction of novolak nitro derivatives with mixture of $SnCl_2/H_2O$ and concentrated HCl. The obtained products were separated and purified.[6,7]

Sulphonated derivatives of novolaks were obtained by sulphonation of novolak wastes with concentrated sulphuric acid, according to the well-known sulphonation method of aromatic compound. The excess of sulphuric acid was next removed in a process of liming by means of calcium carbonate. Water-soluble product was obtained. Products were precipitated in form of sodium salts in reaction with Na_2CO_3.[8]

Sulphonated derivatives of expanded polystyrene waste were obtained by sulphonation of polystyrene waste with concentrated sulphuric acid, according to the well-known sulphonation method of aromatic compound.[3,4] The excess of sulphuric acid was removed in a process of liming with $CaCO_3$. Products were precipitated as sodium salts in reaction with Na_2CO_3, or a product of sulphonation reaction was passed through the column filled with Dowex-1 anion exchanger.

Intrinsic viscosity was determined by means of Ubbelohde viscometer with Pollena K capillary no. I ((K=0,02510 and A_p.103) at 298± 0,01 K. Measurements were conducted in a Julabo Labortechnik GMBH thermostat. Bulk density was measured according to PN-64/C-98054 standard. Identification of polymer characteristic groups and nitro- and amino- groups was done on a Spectrum One IR spectrometer. Samples were prepared in form of tablets using 1 mg of investigated compound and 100 mg of KBr. Spectra were recorded in 4000-400 cm^{-1} range. Studies of the flocculation process were conducted according to PN-71/C-04583 standard. Study of the flocculation process started after selection of an optimum dose of coagulant and then of flocculent. Before this, at the beginning of each measurement the turbidity of the investigated water was determined. Then an optimal dose of coagulant was chosen. For a known dose of coagulant an optimum dose of flocculent was then found by measuring the turbidity of the tested water. The turbidity was measured by means of a Turb 550 IR according to ISO 797/DIN 27027 standard and US EPA recommendation.

Results and Discussion

Chemical modification of the environmentally hazardous novolak and expanded polystyrene wastes gave flocculation materials useful for purification of sewages from power plant, coal mines and steel plant water circulating systems. Properties of synthesized polyelectrolytes are much better than those of polyacryloamide commercial flocculants. Comparative analysis of flocculation properties was conducted for synthesized polyelectrolytes containing maximum functional groups in a polymer. It was found, that the amount of functional groups per one constitutional unit has a great effect on the flocculation and coagulation processes.[3,4,6-9] In the process of chemical modification the amino and sulphonated derivatives of SE and NS novolak and of expanded polystyrene wastes were obtained. Nitration of SE novolak wastes (with increased by 2% content of phenol in relation to commercial product) or SE (with increased by 2% content of phenol) gave nitro derivatives containing 5.21% and 5.30% of nitrogen, respectively. IR confirmed presence of nitrogen groups. Spectra of nitro derivatives exhibit a characteristic asymmetrical stretch vibration bands of –NO at 1541 cm^{-1} and 1537 cm^{-1} and symmetrical stretch vibration band of –NO at 1344 cm^{-1} and 1340 cm^{-1}. Nitro derivatives of SE and NS novolaks were reduced with a reducing mixture (tin salt and hydrochloric acid). The obtained novolak amino derivatives contain 3.02% of nitrogen (SE novolak amino derivative) and 2.95% of nitrogen (NS novolak amino derivative), which corresponds to the presence one NH_2 amino group in two constitutional units. Reduction of SE and NS novolaks nitro derivatives is carried out an identical manner, which is confirmed by the presence of amino groups.

Amino derivatives of NS and SE novolaks are well soluble in 3% water solutions of KOH and NaOH.[6,7]

Sulphonated derivatives of SE and NS novolak wastes were obtained by sulphonation with sulphuric acid. Obtained products contain 10.62% of sulphur (SE novolak) and 9.15% of sulphur (NS novolak). It coresponds to one sulfo group per 3 constitutional units. IR spectra of sulphonated derivatives of SE and NS novolaks exhibit a characteristic asymmetrical stretch vibration bands of S=O sulfo group at 1300 cm^{-1}. It was found, that sodium salts of sulphonated novolaks derivatives are very well soluble in water and can be used as flocculents.[8]

Sulphonation of expanded polystyrene wastes in excess of concentrated sulphuric acid, gave products with a maximum sulphur concentration equal to 13.80% . This corresponds to one sulfo group per one constitutional unit.

IR analysis confirmed the presence of sulpho- groups. IR spectra of these products exhibit a characteristic asymmetrical and symmetrical stretch vibration bands of S=O sulpho group at 1370-1070 cm^{-1} range. It was found, that sodium salts of sulphonated expanded polystyrene wastes are very well soluble in water and can be used as flocculants.[3,4]

Polyacryloamide (M_η= $3{,}5 \bullet 10^5$) obtained in radical homogenise polymerisation or precipitation polymerization of acrylamide was used as a standard polyelectrolyte.[9] Commercial Praestol polyelectrolyte was also used as comparative material.

The obtained modified phenol-formaldehyde resin production wastes and expanded polystyrene wastes, and synthesized standard polyacryloamide were used for study of the flocculation process. Coal mine sewages, water from the steel plant circulating system, and high turbidity model water prepared from tap water and Jaroszów clay were used in studies of the flocculation process. Chemical composition of Jaroszów clay close to the chemical composition of sewages from the power plant was the following: SiO_2 (54-68%); Al_2O_3 (29-40%); Fe_2O_3 (1,5-2,9%); K_2O (1,2-2,4%). Water of high turbidity was prepared according to the Polish Standard – 71/C-04538.

Such choice of water for analysis resulted from the fact that both amino derivatives of phenol-formaldehyde resins and sulphonated derivatives of these resins and polystyrene wastes are polyelectrolytes of the anionic type. Such kind of polyelectrolytes are best for aiding the coagulation process of coal mine water and industrial sewages. Commercial Praestol polyelectrolyte and synthesized polyacrylamide were used as comparative material. Studies were conducted according to standard PN-79/C-04619/03. At first the optimal dose of the coagulant was selected and then the flocculent one.

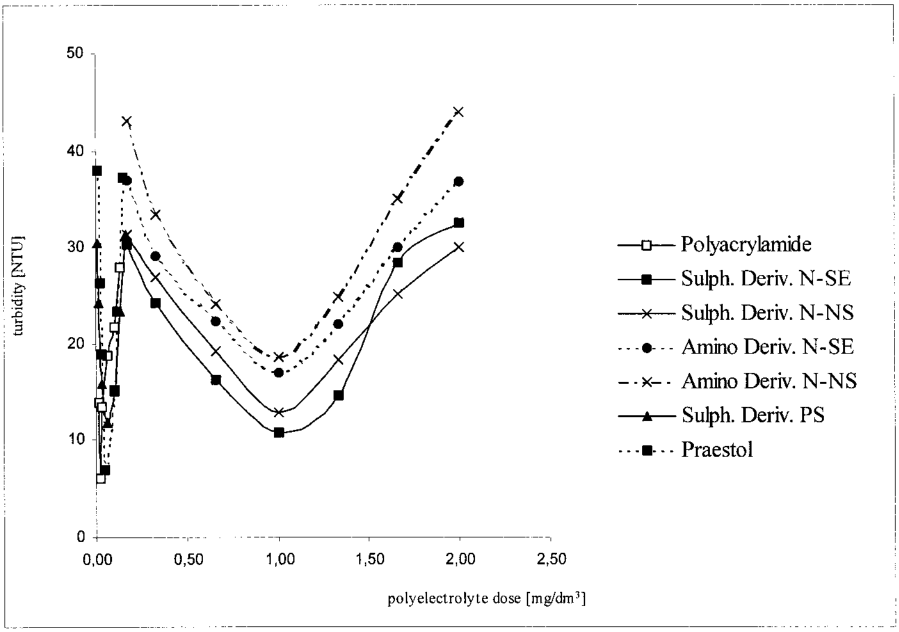

Fig. 1. Dependence of turbidity on optimal doses of synthesized standard polyacrylamide, sulphonated derivatives of SE novolak (sulphonated derivatives N-SE) and NS novolak (sulphonated derivatives N-NS) - phenol-formaldehyde resins wastes, amino derivatives of SE novolak (amino derivatives N-SE) and NS novolak (amino derivatives N-NS) - phenol-formaldehyde resins wastes, sulphonated derivatives of expanded polystyrene wastes (sulphonated derivatives PS), and commercial Praestol polyelectrolyte during purification of model water with initial turbidity of 160 NTU, at coagulant dose of 50.0 mg/dm^3 and pH=6,95.

It was found from flocculation studies with the use of sulphonated derivatives of SE and NS novolak wastes, and amino derivatives of SE and NS novolak wastes that the aiding effects of the coagulation process are comparative with those of commercial Praestol polyelectrolyte and synthesized polyacrylamide. These effects were obtained for Praestol polyelectrolyte and synthesized polyacrylamide having similar values of intrinsic viscosity but with ten times greater concentration of polyelectrolytes, due to the lower molecular weight of the obtained polymers (Fig. 1,2,3). It can be said, that amino and sulphonated derivatives of SE and NS novolak wastes appeared to be the most efficient polyelectrolytes for model water, chemically corresponding to sewages from the power plant (turbidity decrease was 72.2% for sulphonated derivative of SE novolak, 67.1% for sulphonated derivative of NS novolak, 56.3% for amino derivative of SE novolak, 52.2% for amino derivative of NS novolak) (Table 1). Similarly amino and sulphonated derivatives of novolak production wastes are efficient in a broad range of concentrations in comparison to standard polyacryloamide and commercial Praestol polyelectrolyte, and also to sulphonated derivatives of expanded polystyrene wastes.

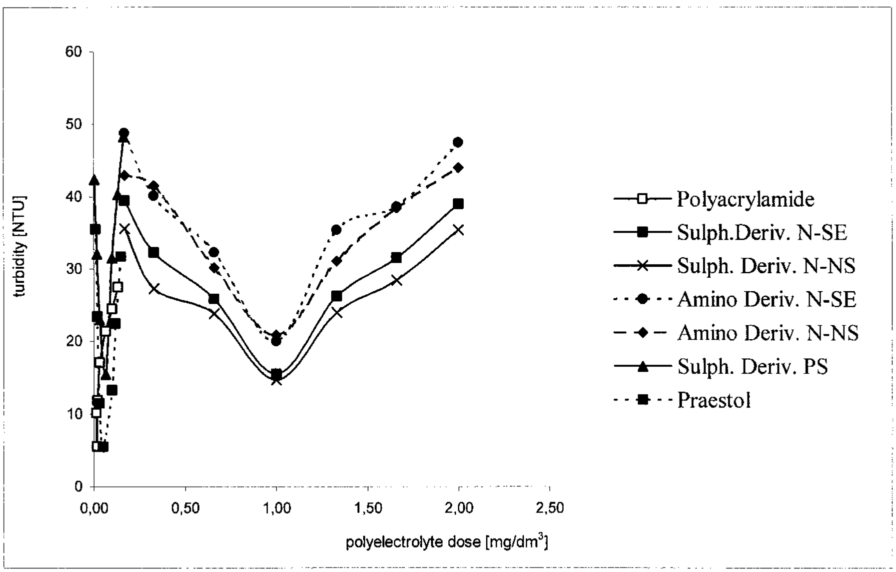

Fig. 2. Dependence of turbidity on optimal doses of synthesized standard polyacrylamide, sulphonated derivatives of SE novolak (sulphonated derivatives N-SE) and NS novolak (sulphonated derivatives N-NS) - phenol-formaldehyde resins wastes, amino derivatives of SE novolak (amino derivatives N-SE) and NS novolak (amino derivatives N-NS) - phenol-formaldehyde resins wastes, sulphonated derivatives of expanded polystyrene wastes (sulphonated derivatives PS), and commercial Praestol polyelectrolyte during purification of coal mine water with initial turbidity 177 NTU, at coagulant dose of 66.7 mg/dm^3 and pH=6.96.

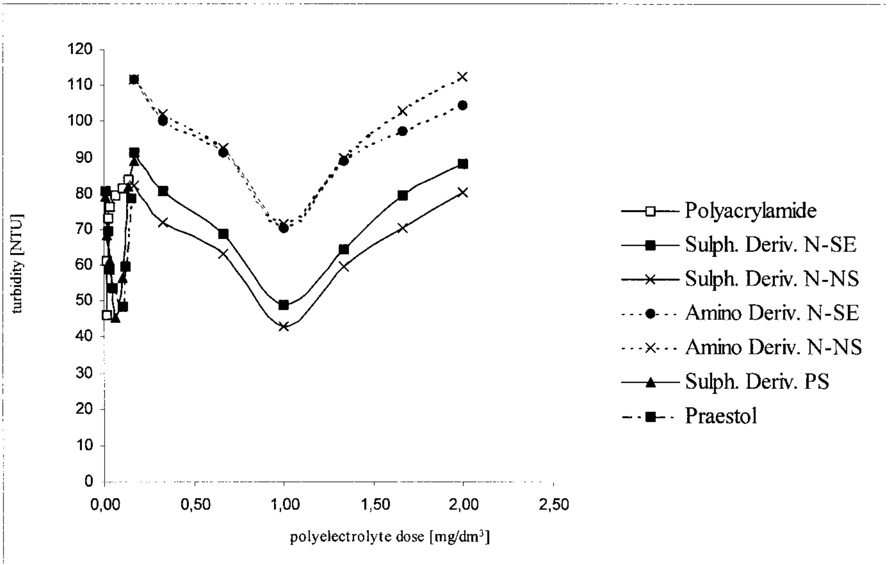

Fig. 3. Dependence of turbidity on optimal doses of synthesized standard polyacrylamide, sulphonated derivatives of SE novolak (sulphonated derivatives N-SE) and NS novolak (sulphonated derivatives N-NS) - phenol-formaldehyde resins wastes, amino derivatives of SE novolak (amino derivatives N-SE) and NS novolak (amino derivatives N-NS) - phenol-formaldehyde resins wastes, sulphonated derivatives of expanded polystyrene wastes (sulphonated derivatives PS), and commercial Praestol polyelectrolyte during purification of water from steel plant circulation system with initial turbidity 247 NTU, at coagulant dose of 100.0 mg/dm^3 and pH=6.94.

It was found from flocculation studies with the use of sulphonated derivatives of expanded polystyrene wastes, that aiding effects of the coagulation process are comparative with that of commercial Praestol polyelectrolyte and synthesized polyacrylamide obtained for slightly lower concentrations. This is particularly visible in the case of water from the steel plant circulating system (Fig. 3, Table 1). Decrease of coal mine water turbidity was 20% higher when Praestol was used. Sulphonated derivatives of expanded polystyrene wastes like commercial Praestol polyelectrolyte and standard polyacrylamide are efficient in a narrow range of polyelectrolyte concentrations.

Table 1. The highest decrease of turbidity for analyzed waters in flocculation processes with synthesized polymers and copolymers obtained from modified plastic wastes and commercial polyelectrolyte; in % of initial turbidity value.

Number	Polyelectrolyte type	The highest decrease of turbidity of analyzed waters [%]* for polyelelectrolytes concentration giving the highest decrease of turbidity [mg/dm^3]		
		model water	water from metallurgical plant circulation system	water from coal mine
1	polyacrylamide	80,2 0,020	61,0 0,017	85,9 0,066
2	commercial polyelectrolyte Praestol 2515	82,0 0,050	59,1 0,100	86,11 0,050
3	sulphonated derivatives of SE novolak	72,2 1,000	58,9 1,000	60,7 1,000
4	sulphonated derivatives of NS novolak	67,1 1,000	63,7 1,000	62,9 1,000
5	amino derivatives of SE novolak	56,3 1,000	40,6 1,000	49,5 1,000
6	amino derivatives of SE novolak	52,2 1,000	39,8 1,000	47,5 1,000
7	sulphonated derivatives of expanded polystyrene wastes	69,7 0,066	61,9 0,066	60,9 0,066

*numerator- % turbidity decrease, denominator – polyelectrolyte concentration [mg/dm^3]

Coal-mine water and water from the steel plant circulating system was analysed in the laboratories of each plant. The following analyses were conducted: reaction, solvated oxygen, biochemical oxygen demand (BZT), chemical oxygen demand (ChZT), oxygen consumption, ether extract, detergents, ammonia nitrogen, sulphates, chlorides, total iron, manganese, phenols, cyanides, solvated parts and suspension, total hardness of water for studies of synthesised flocculants efficiencies, and total hardness of water after flocculation studies (these studies were conducted 24 hours after the flocculation process). Studies were performed according to standards.[10] Results show (Table 2, 3), that the use of chemically modified polymer wastes, similarly to sulphonated derivatives of expanded polystyrene wastes and phenol-formaldehyde resin production wastes and amino derivatives of these resins give a significance decrease of all the analysed factors. However, the observed decreases are not equal for all flocculants. Particularly important and visible are the content decreases of sulphates, chlorides, and solvated parts for the coal mine water and of cyanides, ammonia nitrogen, chlorides, solvated parts, and ChZT for water from steel plant circulating system. This is due to the fact that all contamination factors higher than those permitted for water drained to water race before the flocculation process are significantly below the permitted values or below the detection threshold after the flocculation process. The obtained values of contamination factors of this water after the coagulation process show that purified sludges can be disposed to water races. It was found from the changes of the values of contamination factors of analysed water before and after the flocculation process with the use of chemically modified polymer plastic wastes that the best results were obtained for sodium salts of sulphonated derivatives of expanded polystyrene waste.

Table 2. Permissible values of pollution indicators in sewage, values for water from metallurgical plant circulation system, values for the water coagulationand flocculation processes with modified plastic wastes and synthesized model polymers.

Indicator	Permissible value of indicator for sewage	Value before treatment	Value after treatment with sulfone derivative of expanded polystyrene	Value after treatment with sulfone derivative of novolak NS	Value after treatment with amine derivative of novolak NS	Value after treatment with polyacryl-amide
Reaction (pH)	6,5-9,0	6,9	7,38	7,62	7,45	6,60
Solvated oxygen (mgO_2/dm^3)	-	49,0	8,2	7,5	7,9	7,0
Phenols (mg/dm^3)	0,5	5,1	< 0,005	< 0,005	< 0,005	< 0,005
Cyanides (mg/dm^3)	0,1	1,8	< 0,005	< 0,005	< 0,005	< 0,005
Oxygen consumption- dichromate method (mgO_2/dm^3)	150,0	185,4	< 10,0	25,5	157,8	59,9
Oxygen consumption- permanganate method (mgO_2/dm^3)	-	49,0	4,6	3,1	59,6	25,0
Ether extract (mg/dm^3)	50,0	12,9	11,5	10,5	8,0	15,5
Ammonia nitrogen (mg/dm^3)	6,0	284,0	0,18	0,30	0,41	31,3
Sulphates ($mgSO_4/dm^3$)	500,0	141,8	60,9	71,2	95,5	553,1
Chlorides ($mgCl/dm^3$)	1000,0	1386,0	33,6	12,5	42,3	1423,0
Total hardness ($mval/dm^3$)	70,0	13,67	2,3	2,2	2,3	15,4
Suspension- total amount (mg/dm^3)	50,0	13,2	20,8	32,0	10,8	19,6

Table 3. Permissible values of pollution indicators in sewage, values for water from coal mine, values for the water coagulation and flocculation processes with modified plastic wastes and synthesised model polymers.

Indicator	Permissible value of indicator for sewage	Value before treatment	Value after treatment with sulfone derivative of expanded polystyrene	Value after treatment with sulfone derivative of novolak NS	Value after treatment with amine derivative of novolak NS	Value after treatment with polyacryl-amide
Reaction (pH)	6,5-9,0	7,0	7,16	7,34	7,21	6,85
Solvated oxygen (mgO_2/dm^3)	-	7,7	8,3	7,7	8,1	7,5
Five-day biochemical oxygen demand (mgO_2/dm^3)	30,0	4,9	1,1	0,9	1,9	1,3
Oxygen consumption- dichromate method (mgO_2/dm^3)	150,0	15,8	< 10,0	<10,0	170,8	<10,0
Oxygen consumption- permanganate method (mgO_2/dm^3)	-	6,5	3,8	3,0	63,6	2,3
Ether extract (mg/dm^3)	50,0	5,0	2,5	8,0	2,0	1,0
Detergents (mg/dm^3)	5,0	<0,1	< 0,1	<0,1	< 0,1	0,57
Ammonia nitrogen (mg/dm^3)	6,0	0,18	0,23	0,13	0,50	0,10
Sulphates ($mgSO_4/dm^3$)	500,0	1510,0	28,0	59,7	51,4	92,2
Chlorides ($mgCl/dm^3$)	1000,0	1710,0	12,5	19,2	13,5	47,1
Total iron ($mgFe/dm^3$)	10,0	0,45	< 0,50	<0,50	< 0,50	< 0,50
Manganese ($mgMn/dm^3$)	-	0,85	< 0,15	< 0,15	< 0,15	< 0,15
Total hardness ($mval/dm^3$)	70,0	50,7	2,1	2,3	2,4	2,9
Solvated parts- total amount (mg/dm^3)	2000,0	5120,0	168,0	124,0	280,0	336,0
Suspension- total amount (mg/dm^3)	50,0	78,0	11,4	20,8	12,8	22,8

It is also observed that a small increase of investigated water basicity, is probably due to the use of sodium salts of sulphonated derivatives or with the use of diluted KOH solutions of amine derivatives. However, the pH values do not exceed the values permitted for sewages disposed to water race.

Polyacrylamide, used as a polyelectrolyte turned out to be very effective for the turbidity decrease of the analysed water (Fig. 1,2,3 Table 1) at a very low concentration of this polyelectrolyte. Decrease of the values of the investigated contamination factors of water from the coagulation process with the use of this polyelectrolyte is not so marked as in the case of the used chemically modified polymer wastes. It is particularly visible for total amounts of solvated parts and suspension.

It can be said, that sulphonated and amino derivatives of SE and NS novolak wastes, and sulphonated derivatives of expanded polystyrene, (but not always as efficient as the Praestol and standard polyacryloamide) aid the flocculation process. In the case of phenol-formaldehyde resins derivatives the concentrations should be significantly higher. In despite of this they often exhibit better efficiency than the standard polyelectrolytes.

Chemical modification was conducted for different expanded polystyrene wastes and for different NS-type phenol-formaldehyde resins from unsuccessful manufactured unit. The obtained results were reproducible.

Conclusions

It was found that the phenol-formaldehyde resins wastes and expanded polystyrene wastes can be used as substrates to obtain effective polyelectrolytes. This offers a new solution for their management. It was ascertained, that amino and sulphonated derivatives of phenol-formaldehyde resins production wastes and sulphonated derivatives of expanded polystyrene exhibit good flocculation properties comparable to those of standard polyacryloamide and commercial Praestol polyelectrolyte. They can be used as polyelectrolytes aiding flocculation and improving sedimentation conditions of water having properties analogous to those of sewage from the power plant, the coal mine, and the water from the steel plant circulating system.

This result is an additional argument for further studies concerning processes of chemical modification of polymer plastic wastes and for search of new application areas for these modification products.

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