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SUGGESTION AND FEASIBILITY OF APPROPRIATE TREATMENT METHODS FOR THE REMOVAL OF SULFATE AND ORGANIC MATTER FROM PETROCHEMICAL WASTEWATER EFFLUENT- A CASE STUDY

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Abstract

There are many petrochemical industry in Iran which paying attention to the output wastewater effluent of them have a great importance, because the chemical industry's wastewater have a potential for contaminating the environment. This study first evaluated the output wastewater effluent quality of a petrochemical in south of Iran and then assessed the effluent quality in terms of chemical and physical parameters with the discharge standards of surface water (as an acceptor resource) and eventually for parameters with unacceptable limit (more than the standards). Appropriate treatment method would presented in order to reduce the unacceptable parameters to an acceptable limit. For conducting this study, the sampling have done during six month and the taken samples were evaluated for 13 physical and chemical parameters based on the standard method. The results showed that among the evaluated parameters, the BOD_5 , COD and sulfate level in output wastewater effluent of petrochemical faced with troubles for discharging into the sea water, because their level were higher than the determined standard level. For reducing the parameters level which were more than the standard limit, various methods have been presented. The methods presented based on the efficiency, functionality and economic cost. Based on the obtained results, the three-step reconciliation process (electrocoagulation$\rightarrow$ anaerobic process ABR$\rightarrow$ anaerobic activated sludge process with extended aeration) have presented as a final suggestion. The aim of the mentioned final method was based on the fact that the electrocoagulation was consider for SO_4 reduction and

anaerobic ABR process used for more reduction of COD+ SO_4 and eventually aerobic process (activated sludge with extended aeration) was used for more reduction of SO_4 , COD and BOD.

Keywords: Industrial wastewater, petrochemical industry, wastewater treatment, sulfate, organic compound

Introduction: The urbanization and the population's growth and expansion of industries, emphasized on the importance of environmental pollution control. Wastewaters are one of the environmental pollution factors which should be collected and treated in a hygienic form and if possible, return to the water cycle in nature again (1, 2). The industrial wastewater have various and more pollutants in comparison with municipal wastewaters. Therefore, the treatment and discharging them into the environment needs more care and attention than the municipal wastewaters (3-5). Because the industrial wastewater have lower produced amount than the domestic wastewater, and according to having more dangerous pollutants such as heavy metals, various types of colors, various hydrocarbon compound, toxins and… it could affected the environment particularly the aquatic ecosystems (6-13). In order to prevent the adverse effect of effluent entrance to the environment, some strict legal for the pollutant level of treated effluent discharge into the environment have been established (14).

To meet the environmental standards (related to the effluent discharge) the raw wastewater effluent should be treated before discharging into the environment (1, 14, and 15). There have been various methods for industrial wastewater treatment and pollutants removal and chemical, physical and biological methods in various form have been used for wastewater pollutant removal (1, 2). In physical methods the power and physical properties of materials have used for their removal. The screener, grain retention, chemical precipitation, filtration, sedimentation and other similar methods are the types of physical wastewater treatment methods. In chemical methods for removing pollutants, the chemical reaction and materials have been used. Aeration, coagulation, flocculation, adsorption, ion exchange, pH adjustment and other similar methods considered as chemical methods. In biological methods, the biological process have used for pollutants removal.

The biological methods divided into two aerobic and anaerobic classes (16-23). Some biological process carried out in the presence of dissolved oxygen which called aerobic process and the methods which used aerobic process called aerobic biological process. The activated sludge method, activated sludge with extended aeration, RBC, SBR and MBR are the examples of aerobic biological process (20, 22). The process which carried out in the absence of oxygen called anaerobic process. In biological wastewater treatment methods these process have used. UASB, FBR, ASBR methods are the examples of anaerobic wastewater treatment methods (4). Petrochemical products, according to their number and property's variety have an important role in elimination of daily requirements such as manufacturing pharmaceuticals, foods, clothing, color and paintings, electric, agriculture, hygiene…. 10% of today's automobile's structure, 12% of packaging materials and 35% of textiles are among the petrochemical materials. In petrochemical industries more than 500 various process have been used such as polymerization, hydrogenation, halogenation, crocking, isomerization and crystallization. Therefore, the wastewater of petrochemical have a potential for contaminating the environment (24).

Based on the mentioned content, the aim of this study is to first, evaluating the output wastewater effluent quality of petrochemical in south of Iran and then assessing the effluent quality in terms of physical and chemical parameters with discharging into surface water standards (as an acceptor resource) and eventually for the unacceptable parameters (higher than the standards), appropriate treatment method have presented in order to reduce these unacceptable parameters level to the acceptable limits.

Material and Methods

For conducting this study during the second six month of the 2014 year, 6 time sampling have done from the studied petrochemical wastewater effluent which will discharge to the sea (in each month one sample have taken). For each sample the parameters which have mentioned in table1 have tested based on the water and wastewater standard method book (25).

After evaluation of parameters level, the mean and the standard deviation results of each parameters have calculated by using SPSS ver.16 software.

After that, the mean of each parameters were compared with the wastewater effluent discharge into surface water environmental standard (1, 14). The mean of each unacceptable parameters were compared with the standard level by the statistical one-sample T-test in order to ensure that the average differences of acceptable parameters with the standard level was not accidentally and this differences was noticeable and significant.

Eventually, the parameters which were more than the acceptable limit (standard) were determined with a high assurance and by evaluating the experiences of the previous studies, some suggestion with different choices and priorities have presented.

Results

The results of evaluating the physical, chemical and microbial quality of output petrochemical effluent in table 1 showed that among the evaluated parameters just the BOD_5, COD and sulfate parameters were not consistent with the discharging to the surface water standards and other parameters were consistent with the mentioned standards. It should be noted that the average differences of BOD_5, COD and sulfate parameters with the related standards was in a way that the differences was statistically significant (by one-sample T-test) in a (α=0.05 level) ($P<0.05$). In table1, the mean, standard deviation, minimum, maximum of the evaluated parameters and these parameters level in sea water (acceptor resource) have been presented.

The acceptable limit of some discharged effluent parameters (sulfate and the total dissolved solid) are depend on these two parameters in acceptor resource.

In a way that the petrochemical output effluent discharge should not increase the sulfate and total dissolved solids of acceptor resource more than 10%. According to the fact that the total dissolved solid level of acceptor resource was 48412.5 mg/l and if the mentioned 10% level (48412.5mg/l) was added to that number (48412.5 mg/l) the results would be 53253.75mg/l which considered as the acceptable limit of total dissolved solids of output effluent for discharging into acceptor resource.

Based on this fact, the total dissolved solids level of evaluated petrochemical output effluent (49637mg/l) have evaluated less than the acceptable limit. (53253.75mg/l) and as a results, the discharge of the mentioned effluent have not faced with a serious troubles in terms of total dissolved solids.

But according to the sulfate level of acceptor resource (3200mg/l) and the average level of sulfate in evaluated petrochemical output effluent (8081mg/l) and based on the similar calculation which have done for the total dissolved solids, the acceptable limit for sulfate in discharged effluent into the sea water was determined 3520mg/l, therefore, the evaluated petrochemical output effluent discharge was faced with trouble for the sulfate level (Figure1).

Figure2 showed that the BOD_5 and COD level of the output effluent was more than the acceptable limit of discharging into the acceptor resource (sea).

In summary, although most of the evaluated parameters were lower than the acceptable limits (the environmental protection agency of Iran) but for safe discharge of evaluated petrochemical output effluent into the sea water, the BOD_5, COD and sulfate level should first reduced by an appropriate method at least to an acceptable standard limit and then discharged into the sea.

Table-1: Mean, minimum and maximum of physical and chemical parameters of petrochemical wastewater effluent and seawater (Acceptor resources).

Parameters		Unit	Quality of wastewater effluent for discharge to see				Quality of water see (Acceptor resources)	Discharge standard to surface water
			Mean	SD	Min	Max		
pH	-	-	7.5	0.14	7.38	7.73	8.5	6.5-8.5
Hydrogen sulfide	H_2S	mg/l	0.011	0.003	0.008	0.015	0.018	3
Detergent	ABS	mg/l	0.04	0.010	0.02	0.052	0.018	1.5
Oil &Grease	O & G	mg/l	2	1.2	1	4.2	1.3	10
Biochemical oxygen demand	BOD_5	mg/l	225.17	27	189	260	17.1	30 (momentary: 50)
chemical oxygen demand	COD	mg/l	437.36	47.74	368.5	494	60.5	60 (momentary: 100)
Total suspended solids	TSS	mg/l	10.83	4	6	17	9	40 (momentary: 60)
Total dissolved solids	TDS	mg/l	49637	3210	44560	53121	48412.5	The discharge of effluent into the sea, seawater TDS should not exceed 10% change (1)
Phosphate	PO_4	mg/l	0.42	0.10	0.3	0.55	0.33	6
Ammonia	NH_3	mg/l	0.88	0.50	0.21	1.4	0.95	2.5
Color	Dye	TCU	28.83	9.35	19	45	5	75
Electrical conductivity	EC	µs/cm	51033	3696	45000	55500	60000	-
Sulfate	SO_4	mg/l	8081	151	7850	8230	3200	The discharge of effluent into the sea, seawater sulfate should not exceed 10% change (1)

(1) Discharge at concentrations higher than the amount specified in the table if will allow the effluent, concentrations of chloride, sulphate and soluble receptor source in a radius of 200 meters does not increase more than 10%.

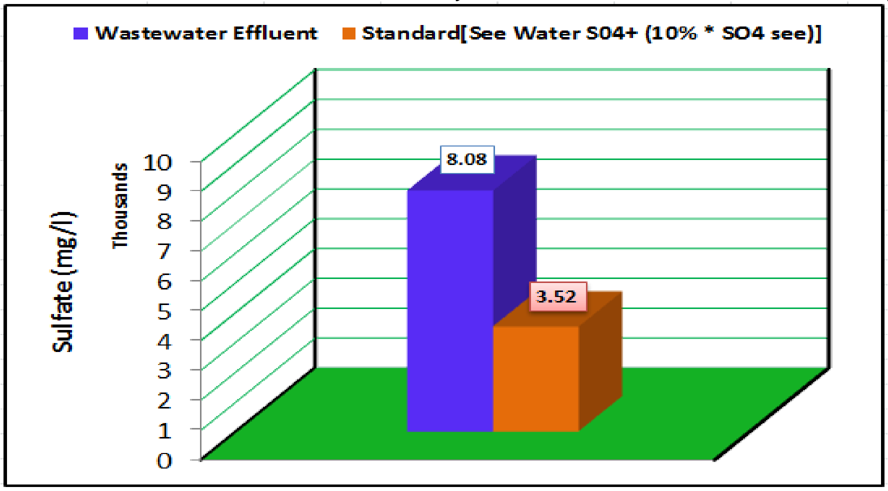

Figure-1. Comparasion of sulfate rate of petrochemical wastewater effluent with standard rate for discharge.

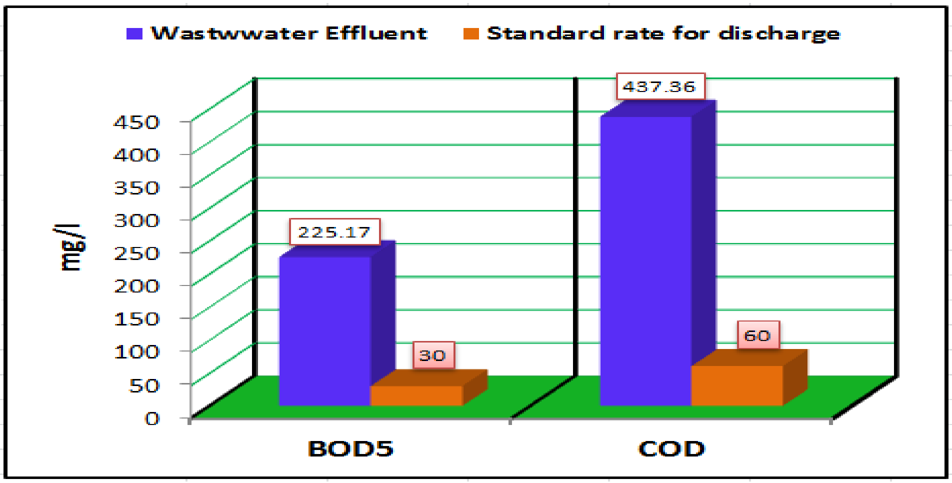

Figure-2. Comparasion of BOD_5 and COD rate of petrochemical wastewater effluent with standard rate for discharge.

Discussion

For reducing the three out of standards parameters (sulfate, BOD_5 and COD) from the evaluated output effluents, first the operational and executive procedures of sulfate reduction have evaluated and then the methods and scenarios of BOD_5 and COD reduction will be introduced. According to the fact that usually the nature of process and sulfate removal mechanism with organic compound (COD and BOD_5) from the wastewater are different, for sulfate removal the physical and chemical method and for organic compound removal the biological method was used (1) and due to the high level of sulfate in the output effluents, first in a particular process, the sulfate level was reduced and then in a separate process the BOD_5 and COD were reduced too. In relation with the above mentioned fact, reminding two points is necessary. First, for removing sulfate, the biological methods (such as anaerobic baffled reactor (ABR) maybe used but as it mentioned, the physic-chemical methods are most common. The second points is that, in the mentioned primary process which aimed to reduced sulfate level, the BOD_5 and COD level may reduce to an particular limit too (26, 27, 1). But perhaps it is not possible to ensure that this level of organic compound reduction was in a standard and acceptable limit. Various techniques such as advanced methods which includes ion exchange, reserve osmosis, nano-filtration (28) electro dialysis (29) chemical precipitation (30) adsorption (31) crystallization (32) electrocoagulation (33) and biological methods (26, 27) were used for the sulfate removal from the aquatic solutions. But each of these methods have advantages and disadvantages which according to our requirements in this research the appropriate and required method will suggested by the prioritization respectively. Maybe among the above methods, the best choice and first priority be the electrocoagulation because the other mentioned methods have problems such as the increased of minerals which include Na, the high operation cost and per capita, requirement for a high initial investment (28) a requirement for final effluent treatment and residual sludge disposal (30, 31). Also there are number of promising and hopeful techniques which are based on the electro chemical technology and don't have the above mentioned problems. Similar studies in relation with sulfate removal from wastewater of aquatics environment have been done which some of them are Mahvi et al.(2010) study which have done by using electro coagulation process with the help of aluminum electrode in a pilot scale and evaluate the sulfate removal level . this study in addition to evaluating the efficiency of process in sulfate removal, evaluated the variables such as pH, applied voltage, the initial sulfate concentration and the reaction time when the maximum removal voltage was achieved 30 volt, and the reaction time was 60min and pH=11 which in this situation for the concentration of 350, 700mg/l the efficiency was reported 58.74 and 44.2% respectively (12). The second priority which have proposed in relation with sulfate removal after electro coagulation process, is a biologic process which named ABR (anaerobic baffled reactor). This type of reactors is a kind of anaerobic system with high loading which developed in the Stanford University of U.S for the first time (4, 35). In Fox P, Venkatasubbiah V (1996) study about the ABR reactor for the pharmaceutical wastewater treatment which contain high sulfate (COD=20000mg/l and sulfate= 2500mg/l and COD/SO_4 =8) in a stable condition, removing 50% of COD and reclamation of 95% sulfate in wastewater have done during one day (36). The third priority which have proposed in relation with sulfate removal is the use of one kind of membrane process which named nano-filtration. This process in addition to sulfate removal, reduced BOD, COD and even TDS in high level completely. For this reason one of the common way of providing fresh water from sea water is the membrane process particularly nano-filtration. But from the other points of view the initial cost investment and its high operation, coagulants in wastewater treatment pay lower attention to it in comparison with other biological methods. A study which have done by Farajollahi et al. (2013) in relation with evaluating the possibility of aluminum and sulfate removal from the anodizing industry's effluent as a input flow to nano-filtration pilot have TDS and sulfate of 6970±330 and 4230±150mg/l respectively which the TDS and sulfate efficiency of 89-97% and 94% in various temperature have removed by nano-filtration method (37).after reduction of sulfate with various mentioned methods, the treated effluents (which their sulfate have reduced) BOD and COD would reduce by one of the biological process. One of the common biological process which used for treating oil wastewaters is membrane bioreactors which using them have a long history, therefore the first priority which proposed for biological wastewater treatment of petrochemicals output, is using membrane bioreactors (38, 39). Shirvani et al. (20120 have done a study about evaluating the refinery treatment of wastewater in oil refinery by using membrane bioreactors. The results of this research showed that the average percentage of COD, BOD_5 , TOC, TSS, oils and colors parameters removal were 97.32%, 92.58%, 96.36%, 99.99% and 95.58% respectively (39).

The second priority which proposed is related to reducing the COD and BOD of output effluent in activated sludge system with extended aeration. Due to the high TDS level of output effluent which enter this system, the activated sludge with extended aeration (accustomed with high TDS) should be used. A study which have done by Dolati et al. (2012) about evaluating the biological treatment of oil refinery's effluent by activated sludge method reported that the activated sludge system could reduce the BOD and COD level in a significant level (40).

The third priority which have proposed is related to COD and BOD reduction of oil wastewater and using electro coagulation method which in comparison with mentioned biological methods, required more cost and energy. Based on the Saeidi et al. (2009) study which have done in relation with COD reduction of output effluent in south pars gas refinery by the electrocoagulation method, showed that by applying an electric flow of 40 ml Amper per cm^2 in 90min, more than 90% of COD level was removed (41).

The fourth priority which have proposed based on the previous study, and according to the development and application of it, is the use of anaerobic methods for treatment of evaluated petrochemical wastewater (such as reduction of organic compound). In Mehrdadi et al. (2006) study which evaluated the efficiency of anaerobic reactor (UASB/Filter) for Hybrid wastewater treatment of Arak petrochemical complex, it was revealed that after 30 weeks, the COD removal efficiency in organic loading was 2kg COD/m^3.d and the 18 hours hydraulic retention time was related to 70.3% (42).

The fifth priority which have proposed is using the advanced filtration method such as microfiltration and ultrafiltration. It should be noted that the first priority in relation to organic compound removal from the various wastewaters is a biological method, because these methods in comparison with other methods in terms of economic aspects have lower cost (1).

Rekabdar et al. (2010) study which have done with ultra-filtration process application in oil effluent treatment of oil refinery unit in a pilot scale, showed that ultrafiltration process could remove oil and grease, TOC, COD, BOD_5 , TDS, TSS and turbidity for 97.2%, 75.4%. 63.3%, 42.3%, 23.1%, 100% and 79.9% respectively (43). In other foreign studies which have done, they emphasized this fact that the process which rely on membranes could reduce the organic compound of oil wastewater in a significant level (44, 45).

Conclusion

Based on the obtained results and studies, it could be concluded that three mentioned parameters (sulfate, BOD5 and COD) could be removed simultaneously just with one method and the second choice is removing sulfate with a separate method. However selecting just one method for removing the three parameters simultaneously is preferred due to having lower cost in comparison with using separated and sequential methods. By practical and executive evaluation of it in pilot scale, the efficiency of the only selected method for simultaneous removal of three parameters in high and acceptable limit have argued. Overall according to the studies which have done, the final results of proposed choices for evaluated petrochemical output wastewater treatment, have presented in table2. (In the mentioned table, in 2 choices priority, the aim of using first choice was removing sulfate, and the aim of using second choice was COD and BOD removal). Again it should be reminded that, the choices are proposed just based on the world's experiences, the evaluation mechanism for oil wastewater treatment According to the natures of the pollutants, the obtained efficiency in similar studies and considering economic aspects. and if the final choice or in other words one choice have used for m petrochemical output wastewater effluent treatment, the mentioned priority in bellow tables would considered and according to the poor financial and available technology, one choice would be selected and eventually the efficiency would be evaluated in pilot scale. If the efficiency in pilot scale would confirmed, then it would be run out at full scale.

Table -2: Suggested options for wastewater treatment, petrochemic.

Single option priorities	Double-priority option (consecutively)
First priority: Electrocoagulation	**First priority:** Electrocoagulation→MBR process
Second priority: MBR process	**Second priority**: Electrocoagulation→ activated sludge process with extended aeration accustomed with high TDS
Third priority: Activated sludge process with extended aeration accustomed with high TDS	**Third priority**: Electrocoagulation→ ABR process
Fourth priority: ABR process	**Fourth priority:** Electrocoagulation→ Hybrid filter process /UASB
Fifth priority: Hybrid filter process /UASB **Sixth priority:** A combination of microfiltration and ultrafiltration process	**Fifth priority:** Electrocoagulation→ SBR process

Based on the obtained results, finally three-step reconciliation process (electrocoagulation→ anaerobic process ABR→ anaerobic activated sludge process with extended aeration) would proposed. The electrocoagulation process have used for reducing sulfate, then the anaerobic process ABR for more reduction ($COD+SO_4$) and eventually the aerobic process (activated sludge with extended aeration) for more reduction (sulfate, COD and BOD_5).

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