

Modification of the Multi-Stage Treatment (Anaerobic- Anoxic- Oxidic) by Adding Almond Shells as Biological Carriers

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Abstract

In this study, the treatment process Anaerobic- Anoxic- Oxidic (A₂O) was modified by adding almond shells as biological carriers. The performance of it has been evaluated in two groups for simultaneous removal of organic matter and nutrients (nitrogen and phosphorous) from wastewater. In both groups the hydraulic retention time (HRT) was 12.5 hours and mixed liquor suspended solid (MLSS) was 2000 mg/L. In the first group: The Oxidic bioreactor was filled with packing up to 7% on an effective volume basis. In the second group: All bioreactors were filled with packing up to 7% on a volume basis for each tank. The efficiency of the two groups was compared by analyzing the removal efficiencies of COD, SS, NH₄⁺-N, PO₄³⁻. The removal efficiency in the first group was 94.71%, 90.52%, 95.73% and 92.55%, respectively, while in the second group was 96.53%, 89.82%, 98.29% and 95.61%, respectively. The treatment when adding shells in all bioreactors is the best, but the SS removal efficiency decreased slightly, the reason for this is due to the degradation of shells (organic matter). Obtained results indicated the good stability of the modified system without adding any external carbon sources whereas the almond shells have the ability to release carbon. Almond shells have the ability to adsorb pollutants and they were excellent carriers for bacteria (Biofilm).

Keywords: Almond Shells, (Anaerobic- Anoxic- Oxidic) A₂O Treatment, Biofilm, Biological Carriers, Carbon Source.

Introduction

Activated sludge (AS) was used as a significant method in wastewater treatment plants (WWTP) for decades. AS processes and configurations have been enhanced for many years. Among these enhancements is the Anaerobic/Anoxic/Oxidic (A₂O) process, which removes organic compounds, total nitrogen and phosphorous (CNP) in three tanks¹. Municipal wastewater causes eutrophication of natural water bodies, if not adequately treated before final disposal because it contains reasonable

amounts of nitrogen (N) and phosphorus (P)². Wastewater requires a more effective Biological Nutrient Removal (BNR) process to ensure effluent N and P satisfy the increasingly stringent discharge standard². The A₂O system is the most used among BNR systems due to its cost-effectiveness and good efficiency because its coupled with aerobic autotrophic nitrification of ammonium (NH₄⁺) to nitrite (NO₂⁻) and nitrate (NO₃⁻) and anoxic heterotrophic denitrification of oxidized nitrogen to

N₂ (nitrogen gas)³. Phosphorus is also removed in it, as it is within anaerobic tank poly phosphate accumulating organisms (PAOs) in the sludge deplete the organic substrates from the wastewater and store them as Poly-β-HydroxyAlkanoates (PHA), as well as release phosphate from the stored poly-P in the sludge. In the next oxic tank, PAOs use PHA as energy to grow and to take up P from the wastewater. Thus, there are many deep problems with the A₂O process such as: the denitrification process being affected by the internal recycling ratios⁴, substrate (carbon) competition for N and P removal between nitrifying bacteria and phosphorus removal bacteria. However, when treating low C/N ratio wastewater makes the performance of N and P removal is not satisfied (When C/N is lower than 5 in the influent)⁵. This process causes high sludge production and low sludge retention time (SRT) during operation. Also, WWTP overloading obligates operators to produce more sludge to get effluent quality standards¹. Waste sludge is unpleasant because of its large volume, odor and high pollutants content. Untreated waste sludge may affect the environment⁶. The treatment and disposal of waste sludge have become a big problem due to its high proportion of volatile solids (VS), bulky amounts of water, and harmful elements such as heavy metals and organic pollutants. Treatment and disposal of waste sludge require energy and chemical agents, which result in significant increases in the carbon footprint and resource utilization of the wastewater treatment process⁷.

The Waste Activated Sludge (WAS) treatments cost up to half of the operating costs in WWTP. Furthermore traditional sludge disposal as landfilling is forbidden by administrative authorities⁸. Therefore, there is a need to design an efficient nutrient removal process with modifications A₂O process by integrating carriers⁹.

Integrated Fixed Film Activated Sludge (IFAS) is a modification process consisting of carriers added to suspended growth reactors. This provides attachment surfaces for bacterial growth, which increases total microbial concentrations without physically expanding existing facilities, and carriers are helpful in stabilizing nitrifying bacteria. The

suspended growth for PAOs is IFAS¹⁰. The biofilm reactor contains two types of biomass, the suspended growth resulting from the return activated sludge (RAS) and the attached growth resulting from the biofilm. The A₂O biofilm combines the advantages of the biofilm reactor and activated sludge process³.

At present, most studies on the biofilm system focus on supplementation of carriers by using synthetic biological carriers in A₂O treatment. For example, Tabraiza, S, et al used polyethylene sheets in all three bioreactor chambers in A₂O¹¹. Jaafari J, et al studied the efficiency of A₂O to achieve simultaneous removal of organic matter and nutrients from municipal wastewater, when Kaldnes K3 carriers with 60% filling were placed in all Tanks¹². Wang c, used polyester carriers to modify nitrogen removal efficiency in A₂O treatment¹³.

Nowadays, carriers made of polypropylene, polyvinyl chloride, polyester, fiber and polystyrene are widely used in the biofilm reactor. However, these raw materials are derived from nonrenewable oil resources, and would not be easily degraded once wasted¹⁴. However, the cost of plastic bio-carriers makes them less attractive for applications in developing countries¹⁵. Therefore, a considerable amount of research has focused on the preparation of biofilm carrier which is easily available, and degradable materials. Searching for the natural carriers as fixed media for supporting microbial growth and replacement with the synthetic carriers to decrease the cost of treatment is considered an important process in low-income communities¹⁶. Agricultural waste is high water absorption, which facilitates cell proliferation and biofilm formation, and thus reduces the time required to operate the treatment system¹⁴. The application of natural materials has attracted significant attention¹⁵. For example, Researchers Zainab, A., et al studied the performance of several anaerobic reactors using natural organic materials such as fiber, coconut husk fiber, and wood chips as carriers¹⁷. Kanwar, R M A, et al investigated the efficacy of using carriers from corncob arborvitae and date palm fibers to support biofilm growth¹⁸. Huang L, et al found that TN removal efficiencies were improved by 24% and 8.98% after adding biodegradable corn cob

carriers and inert commercial fibers respectively at low C/N ratio¹⁹. Shehab D, found that the best filling ratio of pumic stone in aerobic tank activated sludge was 25%²⁰. Agricultural wastes are characterized by their ability to adsorb. Al-Saed Ka, et al used *Phragmites australis* (P.a) Iraqi Plant to adsorb Pb from an aqueous solution²¹.

However, the effect of adding carriers from agricultural wastes into the A₂O treatment is still not clear. The knowledge gained from this study will help to expand the application of the biofilm system with the A₂O process for the complete treatment of municipal wastewater. The aim of the research is to evaluate the efficiency of processing using almond shells as biological carriers.

Materials and Methods

Experiments were conducted in the Al-Duwair treatment plant in the city of Homs in Syria, which is a plant using activated sludge treatment as shown in Fig. 1. In this study, almond shells were dried

and filled with 7% of the effective volume of the tank. The shells were distributed on sieves in three layers to distribute them over the height of the tank.



Figure 1. Part of Al-Duwair treatment plant

Experimental plant components:

An experimental multi-stage biological treatment plant (Anaerobic - Anoxic - Oxidic) was designed

based on the Metcalf reference⁴, where the highest hydraulic retention time (HRT) was relied upon within the fields shown in table 1.

Table 1. Typical design indicators for the most commonly used BNR methods⁴

Design parameter/ process	SRT D	MLSS mg/L	H			RAS, % of influent	Internal recycle, % of influent
			Anaerobic zone	Anoxic zone	Aerobic zone		
A/O	2-5	3000-4000	0.5-1.5	-	1-3	25-100	
A ² /O	5-25	3000-4000	0.5-1.5	1-3	4-8	25-100	100-400
Modified Bardenpho	10-20	3000-4000	0.5-1.5	1-3 (1 st stage) 2-4 (2 nd stage)	4-12 (1 st stage) 0.5-1 (2 nd stage)	50-100	200-400
UCT	10-25	3000-4000	1-2	2-4	4-12	80-100	200-400 (anoxic) 100-300 (aerobic)
VIP	5-10	2000-4000	1-2	1-2	4-6	80-100	100-200 (anoxic)

SBR	20-40	3000-4000	1.5-3	1-3	2-4	50-100	100-300 (aerobic)
Phostrip	5-20	1000-3000	10-12		4-10		10-20

Depending on the HRT in Table 1 and at the value of the flow entering the experimental plant (100L / day =4.2 L/h), the volumes shown in Table 2 were obtained (Volume = Flow× Hydraulic Retention Time).

Table 2. Designed tank size

Indicators	Tank	Hydraulic Retention Time (hour)	Volume (L)
Anaerobic		1.5	6.3
Anoxic		3	12.6
Oxic		8	33.6
Secondary Sedimentation		2	8.4

Fig. 2 shows schematic diagram for the designed experimental treatment plant with scale in cm.

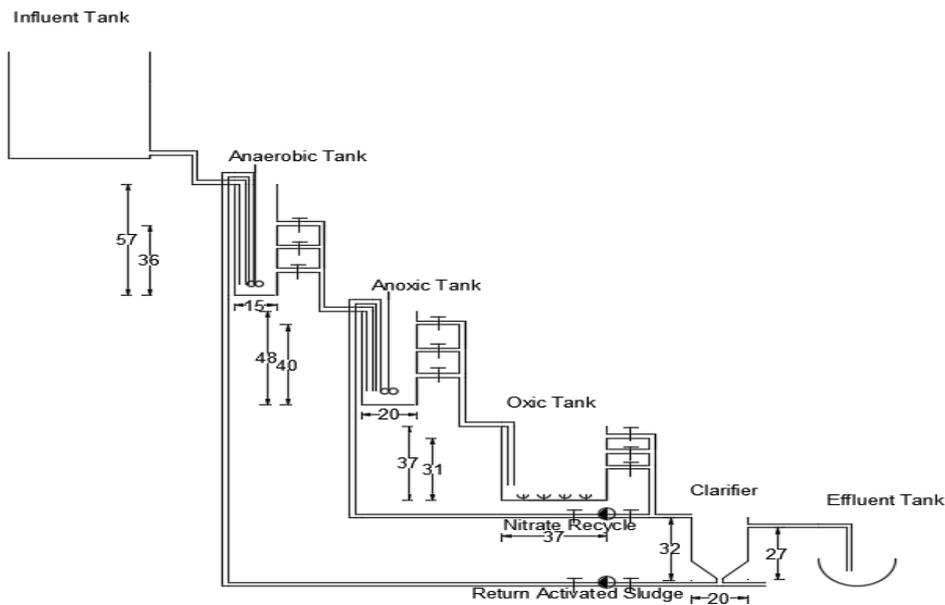


Figure 2. Schematic diagram for the designed experimental treatment plant

Fig. 3 shows the inlet of the designed experimental treatment plant with a capacity of 100 L and is equipped with a valve to control the amount of water flowing, and Fig. 4 shows the sections of the Designed experimental treatment plant sections.



Figure 3. The beginning of the experimental plant



Figure 4. The designed experimental treatment plant

Where:

A- Anaerobic Tank: (Diameter 150 mm, Height 570 mm, Volume 10L), equipped with a mixer to keep the sludge suspended in the tank and made of PVC.

B- Anoxic Tank: (Diameter 200 mm, Height 480mm, Volume 15L), also equipped with a mixer to keep the sludge suspended in the tank and made of PVC.

C- Oxic Tank (Diameter 370mm, Height 370mm, Volume 40L), equipped with an aeration system that provides the tank with a dissolved oxygen concentration of 2 mg/L, made of PVC.

D- Secondary sedimentation tank (Diameter 200mm, Height 320mm, Volume 10L).

E- Tank to collect effluent water.

There were two recirculation flows, An Internal recycle (IR) from the aerobic tank to the anoxic tank by a return pump and the return activated sludge (RAS) from the secondary sedimentation tank to the anaerobic tank by a return pump. The waste sludge is removed manually.

Results and Discussion

Practical experiments were started after about a month of placing the shells within the A₂O treatment in order to give enough time for the formation of the biofilm on the shells. Fig. 6 shows

Experimental plant components:

-The experimental plant was supplied with water from the outlet of the primary sedimentation tank of the Al-Duwair treatment plant at a rate of 100 L/day = 4.2 L/h. It was also supplied with sludge from the aeration tank of the Al-Duwair treatment plant to start the work of the experimental plant directly.

- The Internal recycle from the oxic tank to the anoxic tank was set at 100% of the flow entering the treatment based on the range given in Table 1 and equal to 4.2 L/h using the return pump.

- The returned activated sludge was set at 25% of the flow entering the treatment based on the range given in Table 1 and equal to 1.05 L/h using the return pump, the excess sludge was removed manually to achieve a concentration of MLSS = 2000 mg/L within the oxic tank.

- Almond shells have been used as a biological carrier which was dried as shown in Fig. 5.

- The indicators shown in the tables were measured at the Homs Wastewater Treatment Plant in Al-Duwair, and at the Faculty of Civil Engineering at Al-Baath University.

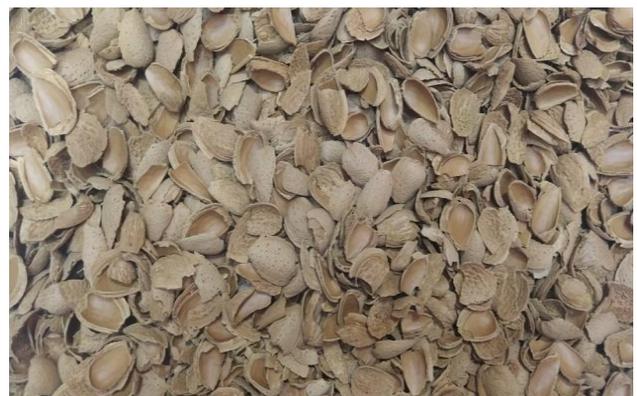


Figure 5. Almond shells after dried

almond shells after a week and Fig. 7 shows almond shells after a month.

Practical experiences are in two groups: The first group: with biological carriers only in the Oxic

tank. The second group: with biological carriers within all Tanks (Anaerobic - Anoxic - Oxidic).



Figure 6. Almond shells after a week



Figure 7. Almond shells after a month

The first group:

Almond shells were placed at a rate of 7% of the effective oxidic tank volume, and they were distributed within three layers, as shown in Fig. 8.



Figure 8. Almond shells distributed over three layers

The results were as shown in Tables 3, 4, 5.

Table 3. Experimental results for five samples at MLSS = 2000 mg/L, HRT = 12.5 hours (COD, SS, MLSS analysis)

Sample Number	MLSS (mg/L)	COD in (mg/L)	COD out (mg/L)	SS in (mg/L)	SS out (mg/L)
1	2100	191	10	74	6
2	2200	204	11	79	7
3	1999	210	12	100	11
4	1800	232	11	112	12
5	2000	240	13	99	8
Average	2109.8	215.4	11.4	92.8	8.8
Removal Efficiency %		94.71		90.52	

Table 4. Experimental results for the five samples at MLSS = 2000 mg/L, HRT = 12.5 hours (Nitrogen ions)

Sample Number	NH ₄ ⁺ -N in (mg/L)	NH ₄ ⁺ -N out (mg/L)	NO ₃ ⁻ -N in (mg/L)	NO ₃ ⁻ -N out (mg/L)
1	48	2	4.2	4.6
2	40	1	5.1	4.8
3	34	1	5	4.7
4	50	3	4.3	4
5	39	2	4.5	4.2
Average	42.2	1.8	4.62	4.46
Removal Efficiency %	95.73		-	

Table 5. Experimental results for the five samples at MLSS = 2000 mg/L, HRT = 12.5 hours (Phosphorous ions)

Sample Number	PO ₄ ⁻³ in (mg/L)	PO ₄ ⁻³ out (mg/L)	P ₂ O ₅ in (mg/L)	P ₂ O ₅ out (mg/L)	P in (mg/L)	P out (mg/L)
1	23	2	17.19	1.49	7.51	0.65
2	22	1.5	16.44	1.12	7.18	0.49
3	18	1.3	13.45	0.97	5.87	0.42
4	26	2.2	19.43	1.64	8.48	0.72
5	21	1.2	15.69	0.89	6.85	0.39
Average	22	1.64	16.44	1.22	7.18	0.53
Removal Efficiency %	92.55		92.58		92.62	

COD removal:

From the previous tables, we find that the COD removal efficiency is 94.71%. Longer HRTs usually increase the organic matter removal because microorganisms have an important chance to get in touch with the substrate and consume it. The A2O reactor with added carriers also features high Sludge Retention Time (SRT) values for this system which provided sufficient time for microorganisms, especially in the attached form¹².

Removal of phosphorus and nitrogen:

COD is the main source of VFAs for PAO bacteria, the conversion of COD to VFAs occurs through fermentation within the anaerobic tank so the more organic matter is removed the more cell growth and thus more phosphorus will be removed. This supports the resulting high COD, phosphorus and nitrogen removal efficiency of this system. This is consistent with what researchers. Leyva-Díaz, J C et al when treated with the A2O method in the presence of a membrane reactor with and without supports, they concluded that the reactors with supports had the best performance because the attached growth can form anaerobic, anoxic and oxic zone so the biofilm can enhance TN removal in

the oxic zone and avoid the transfer of nitrate into the anaerobic zone, which consume COD and prevent TP removal²².

The second group

Almond shells were placed at a rate of 7% of the effective Anaerobic- Anoxic- oxic tank volume, and they were distributed within three layers, as shown in Figs. 9, 10, 11.



Figure 9. Almond shells distributed on three layers within the Anaerobic tank



Figure 10. Almond shells distributed over three layers in the Anoxic tank



Figure 11. Almond shells are distributed over three layers within the Oxidic tank

The results were as shown in Tables 6, 7, 8.

Table 6. The results of experiments for five samples when MLSS= 2000 mg/L, HRT= 12.5 hours (COD, SS, MLSS analysis).

Sample Number	MLSS (mg/L)	COD in (mg/L)	COD out (mg/L)	SS in (mg/L)	SS out (mg/L)
1	2100	226	5	99	11
2	2200	244	7	93	9
3	1979	231	11	72	7
4	2108	196	6	78	8
5	2006	199	9	100	10
Average	2078.6	219.2	7.6	88.4	9
Removal Efficiency %			96.53		89.82

Table 7. The results of the experiments for the five samples at MLSS= 2000 mg/L, HRT= 12.5 hours (Nitrogen ions analysis)

Sample Number	NH ₄ ⁺ -N in (mg/L)	NH ₄ ⁺ -N out (mg/L)	NO ₃ ⁻ -N in (mg/L)	NO ₃ ⁻ -N out (mg/L)
1	30	0.4	5	5.5
2	35	0.6	5.2	5.4
3	36	0.7	4	5.3
4	38	0.8	5.6	5
5	37	0.5	5.3	5.1
Average	35.2	0.6	5.02	5.26
Removal Efficiency %		98.29		-

Table 8. The results of the experiments for the five samples at MLSS= 2000 mg/L, HRT= 12.5 hours (Phosphorus ion analysis)

Sample Number	PO ₄ ⁻³ in (mg/L)	PO ₄ ⁻³ out (mg/L)	P ₂ O ₅ in (mg/L)	P ₂ O ₅ out (mg/L)	P in (mg/L)	P out (mg/L)
1	16	0.6	11.96	0.45	5.22	0.19
2	18	0.7	13.45	0.52	5.87	0.23
3	20	0.8	14.95	0.59	6.53	0.26
4	21	1	15.69	0.75	6.85	0.33
5	23	1.2	17.19	0.89	7.51	0.39
Average	19.6	0.86	14.65	0.64	6.39	0.28
Removal Efficiency %		95.61		95.63		95.62

Nitrogen Removal:

NH₄-N removal: Despite the favorable conditions for Anammox in the anoxic zones, stable Anammox has been seldom confirmed, because the SRT of a WWTP is not enough to retain low-growth Anammox bacteria. Biofilms are an effective strategy to increase low-growth biomass²³. Gao R., et al found that Anammox activity gradually increased within the biological carriers and in sludge flocculants while a higher abundance of Anammox was observed in biocarriers than in suspended masses²⁴. Analysis of microbial populations showed that Anammox bacteria were found within biofilms in the anoxic zone and their abundance was much higher than sludge flocculants²⁵.

NO₃-N removal: Although the ammonium removal efficiency increased significantly, we notice a slight increase in the resulting nitrate concentration compared to the addition of the carriers in the aerobic tank only. The researchers Li J, et al used A₂O treatment and found that the nitrate concentration in the anoxic zone was significantly reduced compared to the stage before adding the carriers, and ammonia was reduced in the anoxic zone which is rarely seen in conventional activated sludge systems. Typical ammonia-oxidizing bacteria (AOB) Nitrosomonas in sludge flocculants only, as it was not found within the biofilms in the anoxic zone, and the presence of nitrospira

oxidizing bacteria was observed in sludge flocculants and biofilms²⁵.

Phosphorus removal:

We notice an increase in the efficiency of removing phosphorus compared to adding carriers within the air basin only. Tabraiza, S, et al studied the efficacy of A₂O treatment with biological carriers in all tanks and concluded that anoxic conditions significantly increased phosphate uptake due to the presence of organic matter (such as COD)¹¹. The increase in phosphorous and nitrogen removal efficiency could be due to the ability of cellulosic almond shells to adsorb pollutants.

SS removal:

We notice a decrease in the removal efficiency of SS when compared with the placement of shells within the oxic tank only. This could be a result of the degradation of shells (organic matter). Researchers Le H T, et al studied the effect of adding corncob carriers, and the effluent of the corncob reactor contained turbidity from corncob degradation, in the same time corncob carrier can release the soluble carbon which was used for denitrification²⁶.

Fig. 12 shows a comparison between the results of the shells in all tanks at HRT = 12.5 hours and when placing the shells in the air tank only at HRT = 12.5 hours.

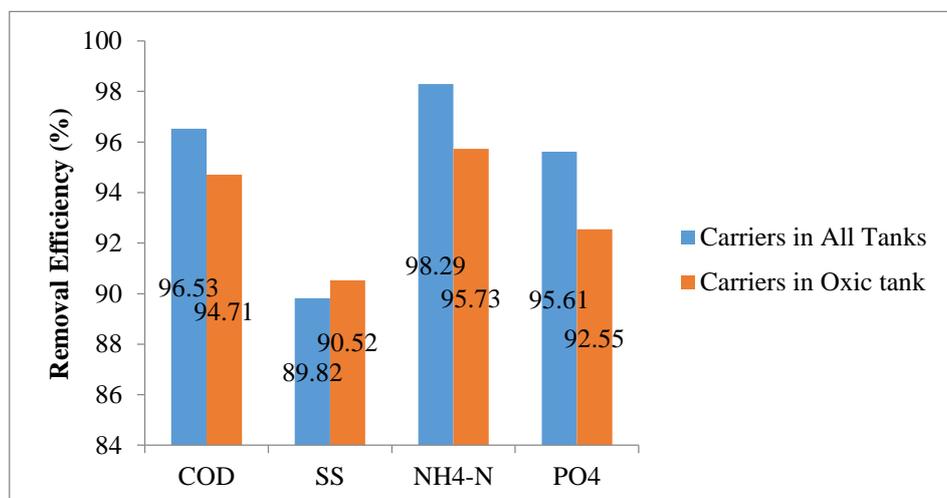


Figure 12. Comparison of the removal efficiency (%) of COD, SS, NH₄-N, PO₄ between A₂O treatment with the placement of carriers in all tanks and the results of A₂O treatment with carriers in the oxic tank

Conclusion

A₂O system with biofilm carrier was excellent biodegradation efficiency for nutrients (N and P) and carbon (COD). An optimum filling: All bioreactors were filled with almond shells with packing up to 7% on an effective volume basis. The employment of the fixed film was a promising approach showing stability and robustness in

removal performance for all ways of filling without any external carbon sourcing. Almond shells have the ability to release carbon, excellent carrier for bacteria (Biofilm) and ability to adsorb pollutants. It is recommended to modify the A₂O treatment by adding almond shells as biological carriers.

Author's Declaration

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are ours. Furthermore, any Figures and images, that are not ours, have been included with the necessary permission for

- re-publication, which is attached to the manuscript.
- Ethical Clearance: The project was approved by the local ethical committee in University of Al-Baath.

Authors' Contribution Statement

All authors contributed to the design and implementation of the research, to the analysis of the results and to the writing of the manuscript.

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تعديل المعالجة متعددة المراحل (اللاهوائية- منقوصة الأكسجين- الهوائية) باستخدام قشور اللوز كحوامل بيولوجية

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الخلاصة

في هذه الدراسة تم تعديل عملية المعالجة A2O (الأحواض اللاهوائية- منقوصة الأكسجين- الهوائية) بإضافة قشور اللوز كحوامل بيولوجية وتم تقييم أدائها في مجموعتين لتحقيق الإزالة المتزامنة للمواد العضوية والمغذيات (النتروجين والفوسفور) من مياه الصرف الصحي، في كلا المجموعتين كان تركيز الكتلة الحيوية المعلقة ضمن السائل المختلط $MLSS = 2000 \text{ mg/L}$ وزمن المكث الهيدروليكي $HRT = 12.5 \text{ hours}$. المجموعة الأولى: تم ملء المفاعل الحيوي الهوائي بما يصل إلى 7% من الحجم الفعال، المجموعة الثانية: تم تعبئة جميع المفاعلات الحيوية بما يصل إلى 7% من الحجم الفعال لكل حوض. تم مقارنة كفاءة المجموعتين من خلال تحليل كفاءات إزالة COD، SS، $NH_4 + -N$ ، $PO_4 - 3$. كفاءة الإزالة للمجموعة الأولى 94.71% و 90.52% و 95.73% و 92.55% على التوالي بينما في المجموعة الثانية 96.53% و 89.82% و 98.29% و 95.61% على التوالي. إن المعالجة عند إضافة القشور في جميع المفاعلات الحيوية هي الأفضل، لكن كفاءة إزالة الـ SS انخفضت بشكل طفيف والسبب في ذلك يعود إلى تحلل القشور (المادة العضوية). أظهرت النتائج الثبات الجيد للنظام المعدل دون إضافة أي مصادر كربونية خارجية حيث أن قشور اللوز لديها القدرة على إطلاق الكربون للبكتيريا، امتزاز الملوثات وتعتبر حامل ممتاز للبكتيريا (الغشاء البيولوجي).

الكلمات المفتاحية: قشور اللوز، المعالجة بالأحواض (اللاهوائية- منقوصة الأكسجين- الهوائية)، الغشاء البيولوجي، الحوامل البيولوجية، مصدر كربون.