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The effectiveness of using a combination of eggshell waste and natural zeolite as an adsorbent for treating laundry waste

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ABSTRACT

In Indonesia, the laundry industry is expanding extremely quickly. There are worries over the concentration of surfactants accumulated in the aquatic environment because the laundry industry has expanded into both large cities and rural regions, yet control over the disposal of laundry waste is still quite poor. In order to lessen surfactant waste in the aquatic environment, we must control laundry waste. Eggshells are not used to their full potential because of a lack of public understanding. In addition to figuring out the ideal composition of the optimum ratio of zeolite to eggshell that can lower detergent levels in laundry waste, this study attempts to ascertain how successful it is to combine chicken eggshells with active zeolite in decreasing laundry waste. In this investigation, two adsorbents were used as a treatment for synthetic laundry waste (LAS) and laundry waste. There are two iterations of each treatment. A 1: 50 ml sample of 1.0 g of adsorbent in 50 ml surfactant samples (laundry waste) was employed in this investigation. It then splits into five different treatments: Zeolite 1.0 g and chicken eggshell 0 g (A), zeolite 0.0 g and chicken eggshell 1 g (B). Chicken eggshell 0.3 g and 0.7 g of active zeolite (C), 0.5 g and 0.5 g of active zeolite (D), and 0.7 g and 0.3 g of active zeolite (E). The findings demonstrated that the detergent content decreased from 50.507 mg/L to P1 (A) 39.535 mg/L, P2 (B), 44.794 mg/L, P3 (C), 38.311 mg/L, P4 (D), 42.063 mg/L, and P5 (E) 37.396 mg/L. These results corresponded to an advisory capacity of A; 0.5484 mg/g, B; 0.2855 mg/g, C; 0.6095 mg/g, D; 0.4222 mg/g, and E; 0.6553 mg/g. The percentage efficiency of each treatment was determined to be 21.724%, B: 11.311%, C: 24.148%, D: 20.073%, and E: 25.959%. E saw the biggest drop, coming in at 37.396 mg/l. According to the study's findings, treating chicken eggshells has a 25.959% efficiency rate and an adsorption capacity of 0.6553 mg/g. The ideal mixture is 0.3 gr. of zeolite and 0.7 gr. of eggshell. Eggshell and natural zeolite are combined in the absorption process of LAS/SLS compounds, with a contact time of 30 minutes and a discharge rate of 8 ml/minute.

ABSTRAK

Di Indonesia, industri laundry berkembang sangat pesat. Ada kekhawatiran mengenai konsentrasi surfaktan yang terakumulasi di lingkungan perairan karena industri laundry telah berkembang baik di kota-kota besar maupun daerah pedesaan, namun pengendalian terhadap pembuangan limbah laundry masih sangat buruk. Untuk mengurangi limbah surfaktan di lingkungan perairan, kita harus mengendalikan limbah laundry. Cangkang telur belum dimanfaatkan secara maksimal karena kurangnya pemahaman masyarakat. Selain untuk mengetahui komposisi ideal perbandingan zeolit terhadap cangkang telur yang optimal yang dapat menurunkan kadar deterjen pada limbah laundry, penelitian ini juga berupaya untuk mengetahui seberapa sukses kombinasi cangkang telur ayam dengan zeolit aktif dalam menurunkan limbah laundry. Dalam penelitian ini digunakan dua adsorben sebagai pengolahan limbah laundry sintetik (LAS) dan limbah laundry. Terdapat dua iterasi pada setiap perlakuan. Sampel 1: 50 ml dari 1,0 g adsorben dalam 50 ml sampel surfaktan (limbah cucian) digunakan dalam penyelidikan ini. Selanjutnya dipecah menjadi lima perlakuan berbeda: Zeolit 1,0 g dan cangkang telur ayam 0 g (A), zeolit 0,0 g dan cangkang telur ayam 1 g (B). Cangkang telur ayam 0,3 g dan 0,7 g zeolit aktif (C), 0,5 g dan 0,5 g zeolit aktif (D), serta 0,7 g dan 0,3 g zeolit aktif (E). Hasil penelitian menunjukkan kandungan deterjen mengalami penurunan dari 50.507 mg/L menjadi P1 (A) 39.535 mg/L, P2 (B), 44.794 mg/L, P3 (C), 38.311 mg/L, P4 (D), 42.063 mg /L, dan P5 (E) 37,396 mg/L. Hasil ini sesuai dengan kapasitas penasehat A; 0,5484 mg/g, B; 0,2855 mg/g,C; 0,6095 mg/g, D; 0,4222 mg/g, dan E; 0,6553 mg/g. Persentase efisiensi masing-masing perlakuan ditetapkan sebesar 21,724%, B: 11,311%, C: 24,148%, D: 20,073%, dan E: 25,959%. E mengalami penurunan terbesar, yaitu 37,396 mg/l. Berdasarkan hasil penelitian, pengolahan cangkang telur ayam memiliki tingkat efisiensi 25,959% dan kapasitas adsorpsi sebesar 0,6553 mg/g. Campuran yang ideal adalah 0,3 gr. zeolit dan 0,7 gr. dari kulit telur. Cangkang telur dan zeolit alam digabungkan dalam proses penyerapan senyawa LAS/SLS, dengan waktu kontak 30 menit dan laju pelepasan 8 ml/menit.

Keywords: *adsorbent, detergent, eggshell, laundry waste, zeolite*

INTRODUCTION

The use of detergents is increasing every year along with the increasing population, meaning that the community's need for the use of detergents is increasing. Laundry services both in urban and even rural clothing are increasing. Disposal of laundry waste directly into the environment without prior treatment, can pollute the environment, especially waters.

The main content of laundry waste that can cause environmental pollution is surfactant (raw material for making detergents). Detergent ingredients are surfactants (the main ingredients of detergents) containing ± 22-30% active ingredients, active ingredients (phosphate compounds) and additives (bleaching and fragrance). Surfactants have polar groups that like water (hydrophilic) and nonpolar groups that like oil (lipophilic), so they can bind the two mixtures (Ety, 2014). The most important substances contained in detergents are ionic compounds in the form of sodium tripolyphosphate which acts as a builder and surfactant. Surfactants can be harmful to the environment because they are difficult to decompose by the environment. Excessive exposure to surfactants in humans can cause eye irritation and skin infections (Apriyani & Novrianti, 2020).

The adsorption process is widely used in industry because it has several advantages, namely it is more economical, does not cause toxic side effects and can remove organic matter. When a solution interacts with a solid adsorbent, the adsorbate molecules will transition from liquid to solid until the solution is in equilibrium with the adsorbate concentration in the solid (Himma, 2017) Freudlich and Langumuir investigated equations that could explain isothermal experimental data. A good adsorbent has a high adsorption capacity and adsorption percentage.

The adsorption capacity can be calculated using the formula:

$$qe= (Co-Ce)v/w$$

The percentage of adsorption (adsorbent efficiency) can be determined using the formula:

$$\%E=((Co-Ce))/Co \times 100\%$$

Where:
qe = number of metal ions adsorbed (mg/g)
Co = concentration of metal ions before adsorption
Ce = concentration of metal ions after adsorption
V = volume of solution of metal ions (L)
W = number of adsorbents (g)
%E = adsorption efficiency

Adsorption is one of the efficient and effective and inexpensive methods to reduce the concentration of a contaminant. Known types of adsorbents include activated carbon, silica gel, and zeolite. In this study, a combination of 2 adsorbents will be carried out, namely eggshells and zeolite. Adsorbents that can reduce pollutants in laundry waste, one of which is eggshells. Eggshells are potential absorbent materials (Mulyati, 2018). Porous structure of eggshell rich in calcium carbonate can be used as an adsorbent for good liquid waste treatment (Li et al., 2021). Zeolite is an aluminosilicate mineral widely found in Indonesia, so it has the ability to adsorb pollutants in laundry waste well. Eggshells and zeolite have high porosity so that both combinations can be used to adsorb pollutants in laundry waste.

Handling or treatment of laundry wastewater includes the Biosand Filter method, floating treatment wetland, moving bed biofilm reactor (MBBR), electrocoagulation, sand filter, and activated carbon adsorption. This method is quite good but has not been widely applied because it is limited in land, requires a long time and requires an aerator, requires the use of electric current and electrodes. Meanwhile, micro-scale laundry entrepreneurs need a practical method that is simple, and cheap, but effective and efficient.

The activation process can increase the surface area and pore size so that it can affect adsorption performance (Fasihah, et al., 2022). Activation of chicken egg shells as adsorbents by burning at 600 °C calcination reaction occurs. The calcination process causes a change in the composition of the eggshell. In addition to changes in composition, the calcination process also causes changes in the morphology and pore structure of the eggshell (Pardede et al., 2020).

The results of research (Apriyani & Novriyanti, 2020) revealed that the use of activated carbon and zeolite without prior activation has the potential to pollute the environment.

Some of the background methods that have been described related to handling laundry waste, in this study one solution that can be applied in the community is a simple filtration method with a combination of *zeolite* and eggshell. Based on research Fasihah et al., 2022), eggshells are effective in reducing MBAS levels where the optimum adsorbent size of 150 mesh can reduce BOD levels by 80% and MBAS levels by 38.85% and 200 mesh sizes which can reduce COD levels by 67.34%. The optimum time for reducing COD and BOD levels is at 90 minutes which can reduce BOD levels by 80% and COD levels by 74.5% and 120 minutes which can reduce MBAS levels by 35.67%.

According to (Purnamasari, 2015), natural zeolite can be used to reduce the concentration of Linear Alkylbenzene Sulfonate (MBAS) compounds in laundry waste water by 93.6 – 95.2%. The maximum decrease in the concentration of Linear Alkylbenzene Sulfonate (MBAS) compounds was achieved with the addition of

100 grams of zeolite with a decrease in MBAS concentration of 0.48 mg/L.

Based on the background above, this study aims to see the effect of using a combination of zeolite and chicken egg shells to reduce the level of laundry wastewater pollution in particular the levels of surfactant, phosphate and COD.

METHODS

Research Location

This research was conducted on laundry waste around Bantarjati, North Bogor. Sample testing was carried out at the AKA Bogor Polytechnic test laboratory and the Bogor stone mountain forestry test laboratory. The test parameters measured are Detergent (MBAS), COD and Total Phosphate. The research conducted between February and July 2023.

The research conducted uses descriptive experiments consisting of several methods that have been standardized by SNI and related journals. This research consists of 4 stages including adsorbent preparation, adsorbent quality testing, adsorbent optimization, and adsorbent application to laundry waste. Application of adsorbents (combination of eggshells and zeolite) to laundry waste with a simple filter device.

Material

The necessary materials are laundry waste water taken in one of the laundry around, eggshell, zeolite, aquades, NaOH, H_2SO_4, Benzene, buffer pillow, isopropyl alcohol, Na_2SO_4, ammonium molybdate, ascorbic acid, filter paper, potassium antimonyl tartrate, MBAS (linear alkylbenzene sulfonate), HCl, NH_4NO_3, $NaH_2PO_4.2H_2O$, buffer solution (pH 4, 7, and 10), phosphate buffer solution, $K_2Cr_2O_7$, $HgSO_4$, Ag_2SO_4, $CaCl_3$, $FeCl_3$, $MgSO_4.7H_2O$, KH_2PO_4, and NH_4Cl.

Activation of Chicken Eggshells and Zeolite

Eggshells from the Home industry of Bantarjati and Tegal Gundil regions, Bogor City, were used as biosorbent materials in this study. The eggshell is cleaned of dirt and membrane layers attached with running water, then ground and sifted using a mechanical sieve measuring 150 mesh and furnaced at 600 °C for 2 hours and then stored in a desiccator for 3 hours. *Natural zeolite* is pureed and then sifted using a sieve mechanical size 200 mesh and heated to 600 °C with a time of 2 hours and stored in the desiccator for 3 hours.

MBAS Methylene Blue Active Surfactant (MBAS) Laundry Wastewater Sample Analysis HACH .8027 Method

The principle of MBAS testing is that anionic surfactants react with methylene blue to form blue-colored ion pairs that dissolve in organic solvents. The intensity of the blue color formed was measured with a spectrophotometer at a wavelength of 605 nm. The measured absorption corresponds to anionic surfactant levels. The test procedure is in accordance with the HACH Method. The spectrophotometric method is an instrument based on Lambert's law BEER to measure a solution with a light source.

Test Procedures:

1. Insert the test sample then add 30 ml benzene and 10 ml buffer sulfate solution, insert 1 bh buffer pillow then extracted.
2. The extracted solution will be divided into two layers and then accommodated into the upper layer Erlenmeyer.
3. Once accommodated into the Erlenmeyer insert it in the spectrophotometric cuvette.
4. Enter the program number for the Detergent test parameter.
5. Rotate the wavelength appropriate for the detergent test parameters and then create a calibration curve.

Table 1. SNI adjustments.

Parameter	Method	SNI
COD	Reflux spectrophotometer	SNI 6989. 2-2009
Surfactant (detergent)	Spectrophotomete HAC H	Method 8028, surfactant anionic
Phosphate	Test phosphate levels spectrophotometrically with ascorbic acid	SNI 06.6989-31:2005

RESULT AND DISCUSSION

Zeolite and eggshell activation process

Eggshells that are clean from dirt and membrane are then dried below sunlight and smoothed use blender dry. The sieving process is carried out using a mechanical sieve of 150 mesh size. While the zeolite uses a size of 200 mesh. This sieving aims to make the eggshell more homogeneous in size and have a wider surface.

The 2 adsorbents (egg shell and zeolite) are physically activated in the muffle furnace at 600°C for 2 hours. The more surface area produced, the more pores are formed. The purpose of physical activation is to increase the adsorption trap power. According to (Napitapulu, 2009), with physical activation, it can enlarge pores, namely by breaking chemical bonds or oxidizing surface molecules so that the surface area increases and affects adsorption power.

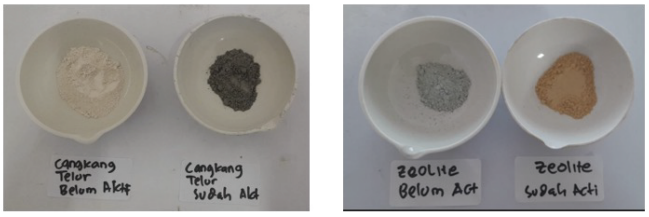

Figure 1. eggshell and zeolite before and once activated.

Based on Figure 1, you can see the difference between eggshell and zeolite before and after the activation process. Discoloration the eggshell turns black clearly because in the carbonization process the high enough hydrocarbon content in the eggshell undergoes authoring. Likewise, with activated zeolite undergoing discoloration, the purpose of physical activation is to evaporate the remaining water in the pores of the zeolite crystal, so that the surface area of the pore increases.

Basically, only the water content evaporates in the heating process, and there is no aluminum removal process or impurity separation in the zeolite structure, so there is no change in the composition of the zeolite material.

Optimization of the combination of eggshell adsorbent and zeolite

Factors that affect the adsorption process must be optimized by optimization. Determination of the optimum conditions of the adsorption process in this study is to determine the optimum weight, and the optimum concentration.

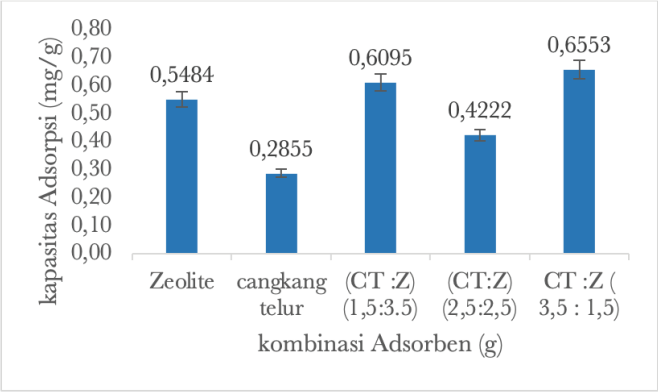

Figure 2. Optimum combination of adsorbents.

Based on Figure 2. above shows that the mass capacity of 1 g of zeolite adsorbent is greater compared to eggshell adsorbents of the same mass. Zeolite has an absorption model of silicon dioxide, ie. active groups SiO_4 and AlO_4, and in addition the largest eggshell forming component is $CaCO_3$, where this compound is included in the polar adsorbent group (Hajar, et al, 2016). Both materials also have pores that can increase their surface area.

The adsorption capacity of methylene blue increases as the mass of adsorbents on the eggshell increases, so the number of active sites on the surface of the adsorbent increases. However, under certain conditions the adsorption capacity remains or even decreases because the adsorbent is saturated due to the reduction in the number of active sites (Nurlaili et al., 2017).

The optimal ratio of eggshell and zeolite adsorption weight is 0.7:0.3 g with an adsorption capacity of 0.6553 mg/g, where the percentage of adsorption efficiency is 25.21%. This value is very different compared to zeolite and eggshell when reacted on its own. Zeolite has an adsorption capacity of 0.5484 mg/g while eggshells are 0.2855 mg/g.

Optimization of Adsorption Concentration of Laundry Waste

Determination of the optimum concentration aims to determine the adsorption capacity of the adsorbent in adsorbate and determine the optimum concentration of surfactant that can be adsorbed by the adsorbent to a saturation point thereby reducing its ability to absorb. In this study, several variations in concentration were carried out including (10, 20, 40, 80, and 100) mg / L.

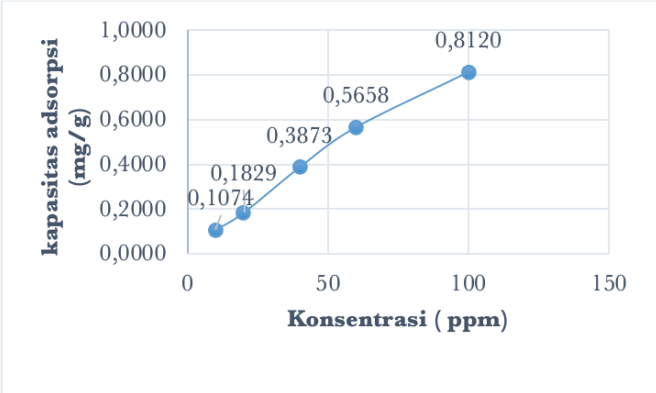

Figure 3. Optimum concentration.

Variations in the optimum concentration of adsorbents against surfactants were carried out with a weight of 3.5 grams of eggshells and 1.5 grams of zeolite and a contact time of 60 minutes and with a stirring speed of 120 rpm. Figure 3 above shows the effect of surfactant concentration on the adsorption capacity of adsorbent.

The adsorption capacity of the adsorbent increases with the increase in surfactant concentration. It refers to the active side on the surface of the adsorbent, the more active side it is. The more active sides, the higher the absorption capacity. Based on this study, the optimum concentration of eggshells and zeolite against surfactants / detergents was obtained at a concentration of 100 mg / L, with an adsorption capacity of 0.812 mg / g.

Decreased Detergent Levels After Adsorbing with Zeolite and Eggshell

The length of adsorption time affects the amount of decrease in surfactant levels in detergent wastewater samples (Figure 4). At the adsorption time that lasted for 1.5 hours, the rate of decline of SLS compounds was not too significant. The large amount of adsorbent mass in the adsorption process also affects the decrease in SLS compounds. The above is the result of anionic surfactant adsorption with fluctuations in discharge. Each discharge has different levels of surfactant.

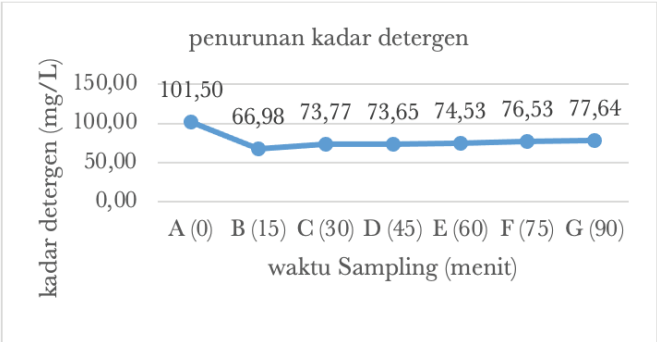

Figure 4. Decreased detergent levels flow rate 8 ml/min.

The length of adsorption time affects the amount of decrease in surfactant levels in detergent wastewater samples (Figure 4). At the adsorption time that lasted for 1.5 hours, the rate of decline of SLS compounds was not too significant. The large amount of adsorbent mass in the adsorption process also affects the decrease in SLS compounds. The above is the result of anionic surfactant adsorption with fluctuations in discharge. Each discharge has different levels of surfactant.

It can be seen from the picture that the separation is influenced by anionic surfactant (detergent) levels, this can be seen from all variations in discharge of 8 ml / minute which can provide significant surfactant reduction results. At the initial detergent level of 101.50 mg / L, and after being applied with the filtration method using a simple tool obtained at a sampling time of 15 minutes a decrease in detergent levels of 66.98 mg / L, and at the 30th minute and so on there has been an equilibrium and saturation point occurs.

The percentage of removal is around 27.32%. This is because the lower the variability of the discharge, the longer the contact time between the surfactant and the adsorbent to maximize the absorption process (Mistar et al., 2017).

Based on Figure 5. It can be seen that with an allowance rate of 18 ml / min, the content of anionic surfactant (detergent) decreases, but is less optimal than the discharge of 8 ml / minute. This is because the lower the variability of the discharge, the contact time between the surfactant and the adsorbent is longer, and vice versa the higher the discharge, the faster the contact time so that the discharge absorption process is slower and vice versa (Mistar et al., 2017).

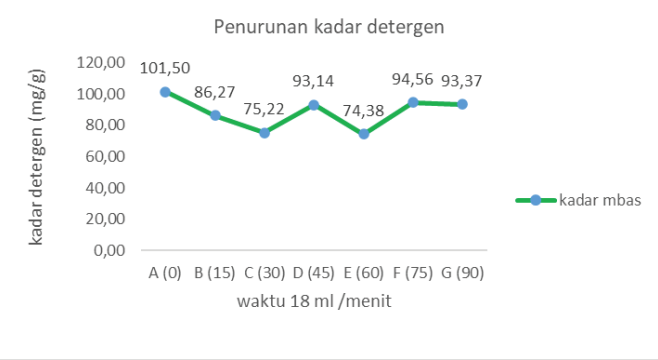

Figure 5. Decrease in detergent levels flow rate 18 ml/min.

At a discharge of 18 ml / minute shows a saturation point, which is shown at a sampling time of 60 minutes decreased after minutes 75 and 90 detergent levels rose. This shows that the higher the discharge, the less optimal the results of surfactant removal in laundry waste (Maharani, 2018). The percentage of removal of detergents is 8.24%. So it can be concluded that the optimal percentage of detergent removal is 8 ml / minute which is 27.32% at a sampling time of 30 minutes.

Effect of PO_4 Levels After Adsorbing with Zeolite and Eggshell

From Figure 6, it can be seen that the phosphate concentration dropped from the initial concentration of 51.56 ppm to 47.63 ppm which shows that phosphate is adsorbed in waste laundry with a combination of zeolite adsorbent and eggshell of 7.62% for 15 minutes at a flow rate of 8 ml / minute. But at sampling times of 30, 45, 60, 80, 75 and 90 minutes increased, this is because the adsorbent adsorption power is decreasing and the state becomes saturated. The use of eggshell adsorbents and zeolite can help lower phosphate levels in detergent waste by binding phosphate from the solution. This is in line with research by (Astuti, 2015), (Irawaty et al., 2021).

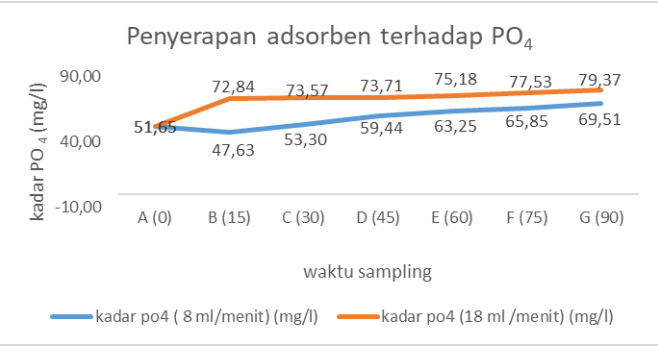

Figure 6. Adsorbent absorption against PO_4.

The sampling time of 30 – 90 minutes of phosphate level reduction is not significant because the adsorb process requires a certain time to interact with phosphate in detergent waste.

Elevated phosphate levels after detergent waste treatment using zeolite or eggshells can occur for several reasons:

1. Desorption effect, Zeolite or eggshell may not completely bind phosphate from detergent waste. When the material is saturated with phosphate, the bound phosphate can be released back into solution when conditions change or shock occurs.
2. Imperfections of Treatment, Sewage treatment process using zeolite or eggshell may not be perfect, and some phosphates may remain dissolved in solution after treatment.

Decreased COD Levels After Adsorption with Natural Zeolite and Eggshell

COD (Chemical Oxygen Demand) analysis in detergent waste analysis is very important because it provides information on the increase in the amount of chemically oxidized organic matter in the waste sample. COD values reflect the burden of organic pollution in waste, which can have a negative impact on the environment if disposed of without adequate treatment (Figure 7). A value indicating the amount of oxygen in 1 liter of a solution of oxygenated organic compounds (Setyobudiarso, 2014).

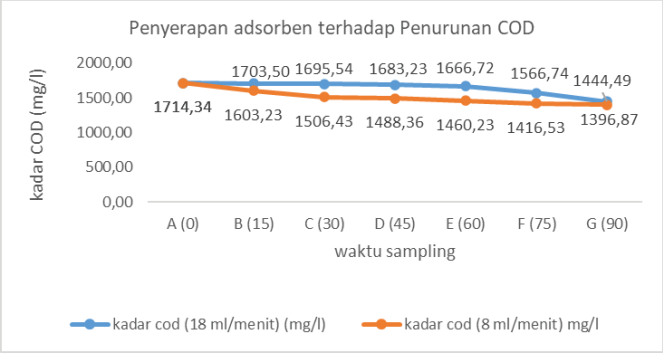

Figure 7. Adsorbent absorption against COD.

According to Mulia (2005), organic substance of COD compounds that can be processed biologically (biodegradable) and those that cannot be processed biologically (non-biodegradable). In the picture above, the initial cod content of laundry waste is 1714.34 mg / L, and the decrease in cod content after laundry waste treatment after 1.5 hours is 1397.87 mg / L or about 81.54%.

Basically, the treatment carried out causes a decrease in the COD parameters of laundry water, this can be seen from the percentage decrease in COD levels. In general, the COD number is a measure of water pollution by organic matter which can certainly be oxidized during microbiological processes, but does not necessarily cause a decrease in dissolved oxygen in waters (Setyobudiarso, 2014). Another factor that also contributes to this decrease is the flow rate adjusted to achieve a relatively long dwell time before being drained to the filter unit.

The decrease in COD levels of detergent waste after processing with eggshell adsorbents and zeolite is caused by the ability of these adsorbents to absorb organic matter resulting in high COD values. By using adsorbents it can help bind and remove potentially polluting components contained in detergent waste, thereby reducing the value of COD and its negative impact on the environment.

CONCLUSION

The results of the study found that the application of simple filtration with the optimum combination of zeolite and eggshell can reduce surfactant levels and Chemical Oxygen Demand (COD). The optimum conditions of eggshell and zeolite adsorbents in adsorbing anionic surfactants at a combination of 0.7 g eggshell and 0.3 g zeolite, flow rate 8 ml/min, and adsorption time 30 min. The maximum adsorption capacity of eggshell and zeolite adsorbent combination (0.7:0.3) against surfactants in laundry waste is 3.24 mg/g.

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