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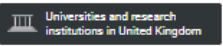
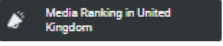
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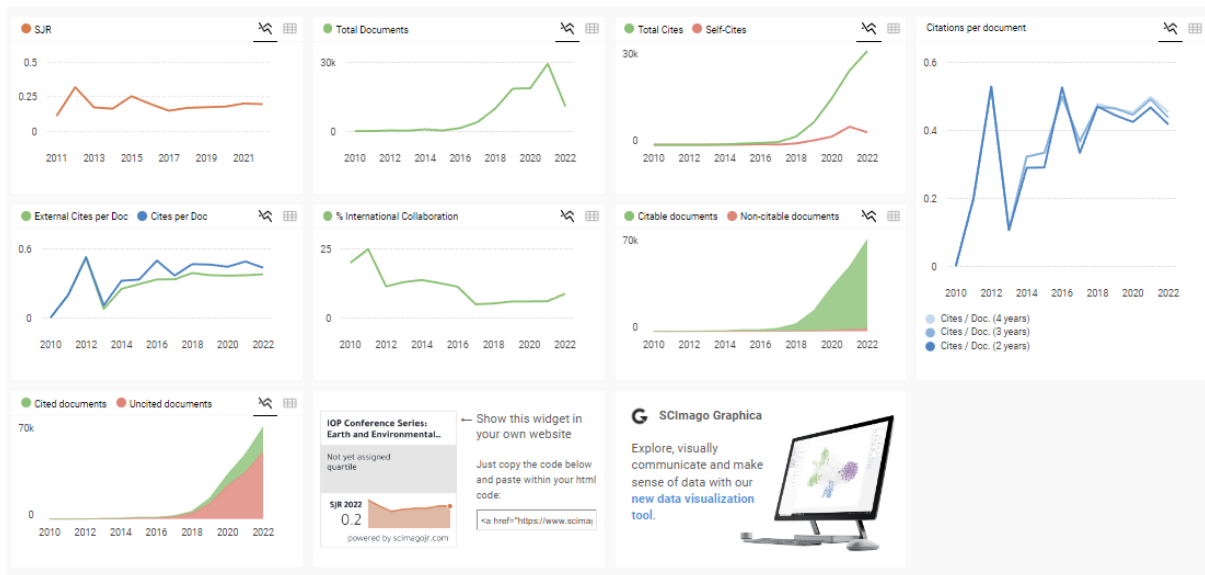
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R Pratiwi^{1,*}), D A Chusniyah¹), K Fajarwati¹), Fadliyah¹), L Satiawati¹)

¹)Department of Petroleum Engineering, Faculty of Earth Technology and Energy, Universitas Trisakti, Jakarta, Indonesia

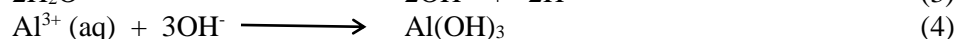
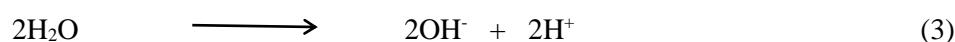
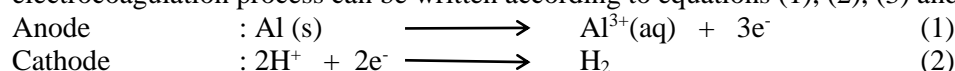
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1. Introduction

Oil exploration activities in Indonesia continue to experience growth in line with the increasing energy demand. This condition has the consequence of producing liquid waste, which needs to be controlled because of its nature which can damage the environment and threaten the lives of living things around the waste disposal area. One method that is widely used in organic wastewater treatment is electrocoagulation. This method uses the working principles of coagulation, flotation and electrochemistry [1], which in its development, are known as efficient and relatively inexpensive methods to operate. The electrochemical process will produce a coagulant, a strong adsorbent for dissolved pollutants [1, 2]. The adsorbed pollutant will then be coagulated due to its strong affinity [3]. Meanwhile, water is reduced at the cathode metal plate to produce OH⁻ anions and hydrogen gas (H₂). The electrocoagulation method is known as a method that is quite effective in removing pollutants dissolved in the liquid phase. In addition, during the electrocoagulation process, hydrogen gas is also produced as an alternative renewable energy source. This process is quite interesting to develop because it provides two products at the same time, which are expected to be able to answer two critical issues today, namely environmental issues and energy issues.

Aluminum electrodes were used as anodes considering they are relatively stable and capable of producing highly adsorptive Al(OH)₃ coagulants (Saravanan, naje). The reactions that occur in the electrocoagulation process can be written according to equations (1), (2), (3) and (4) as follows:



The research, in general, aims to obtain operating conditions in the form of electrical power, solution pH, and processing time that can provide optimal results in pollutant removal and hydrogen



production [4–6]. Phenol solution is used as a waste model because it is one of the many components of petroleum industry wastewater. This research activity observes 3 systems in stages, including the effect of pH, the effect of electric power, and the processing time, which gives optimal results. Changes in the amount of pollutant concentration and the amount of hydrogen gas produced are parameters for the ability of the electrocoagulation process. Quantitative observations were carried out using a spectrophotometer to measure pollutant concentrations and a gas chromatograph to measure the amount of hydrogen gas. Qualitative observations were carried out using FTIR to determine the type of pollutant that was still present in the liquid waste sample.

2. Methods

The research was conducted on a laboratory scale using a sample solution containing pollutants from various phenol compositions. The anode uses a 1.5 mm (4x8cm) thick Aluminum metal plate and a 1mm (4x8cm) Stainless Steel 316 (SS-316) plate as a cathode. The electrocoagulation reactor was a 500 mL acrylic vessel connected to a direct current power source with a maximum of 50 V. The process of hydrogen gas recovery and conversion of pollutant removal in solution was observed for 4 hours. Observations were made at different initial pH, namely 5, 7, and 10. The pH adjustment was made by adding 0.1M NaOH until it reached the desired acidity level.

Pollutant concentrations were analysed using a UV-Vis spectrophotometer with a Bel UV-M51 Spectrophotometer. Gas chromatography analysed hydrogen production using Shimadzu GC-2014, MS-5A column, and Ar carrier gas. The electrocoagulation process is presented in Figure 1, where the sample solution is put into the reactor (a). Electrodes (d and e) are placed in the reactor while connected to the power supply (g) at 10 - 20 V. Meanwhile, a hot plate with magnetic stirrer (f) is used to ensure the homogeneity of the solution.



Figure 1. The apparatus of electrocoagulation to eliminate phenol solution and produce hydrogen

The total pollutant concentration and the amount of hydrogen production were used as the variables observed during the process, which was carried out within 4 hours. Variable measurements take liquid samples at valve (c) and gas samples at valve (b) every 1 hour. Pollutant concentrations in wastewater were measured using UV-VIS spectrophotometry, while the amount of hydrogen gas was measured using a GC (Gas Chromatograph).

3. Result and Discussion

The performance of the electrocoagulation process using aluminium electrodes has been observed to remove samples of liquid waste containing phenol as a representation of the liquid waste from the petroleum industry. In addition, the ability of the process to produce hydrogen gas was also observed. The process is carried out using a direct current power source with an electric voltage of 10 to 20 volts

for 4 hours for each observation. The independent variables used at this stage are the pH of the sample solution, the electric current voltage, and the processing time. Changes in pollutant concentration and the amount of hydrogen gas produced at intervals of 1 hour were measured. The observation results obtained are as follows:

3.1. The effect of the pH of the solution

In the electrocoagulation process with aluminium electrodes, efficient coagulant formation occurs at a pH range of 6 – 7. The electrocoagulation process can occur at a more comprehensive pH range [7, 8]. Aluminium amphoteric electrodes also help stabilize acidity with their ability to neutralize pH [7]. This results in an electrocoagulation process involving a solution with a low pH that will increase in pH during the process; conversely, if a solution with a high pH is used, there will be a decrease in pH.

Observations were made at low acidity and alkalinity levels, namely at pH 5, 7 and 10, respectively. It is known that the process carried out in the pH range of 4 – 10 does not affect the decrease in the amount of current strength [8]. The observation results are shown in Figure 2, where it can be seen that increasing the pH increases the ability of the process to remove CIP and produce hydrogen gas.

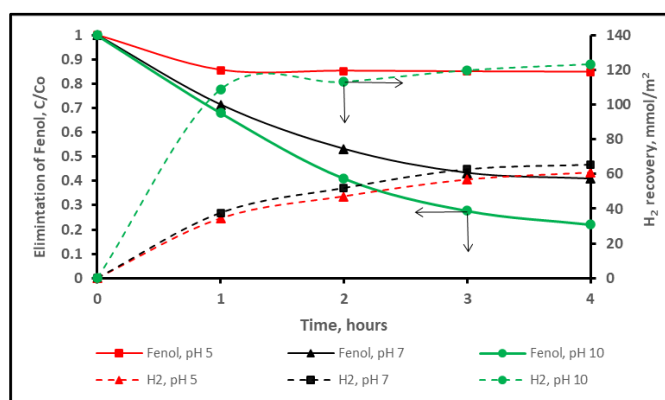


Figure 2. The effect of pH on the ability of the process to remove phenol and produce hydrogen gas (5 ppm phenol solution, 20 volt power supply)

The presence of abundant OH^- ions in a solution with a pH of 10 allows the formation of more $\text{Al}(\text{OH})_3$ coagulants, thus increasing the amount of pollutant that can be adsorbed. Under very alkaline conditions, the greater the occurrence of aluminate ions ($\text{Al}(\text{OH})_4^-$) as a result of excess OH^- in solution according to equation (5) [9],



The presence of aluminate ions makes it possible for hydrogen gas to form because aluminate ions are known to have a good ability to reduce H^+ cations. However, their adsorption capacity is low compared to $\text{Al}(\text{OH})_3$ [8].

3.2. Electric current voltage

The amount of electric current used dramatically affects the ability of the anode electrode to dissolve in the form of ions, which is directly proportional to the amount of coagulant that occurs [4, 6]. The more significant the electric current, the more coagulant occurs until the optimum electric current value is reached. When using an electric current more excellent than its optimum value, there is no significant coagulant addition [10, 11].

The results obtained from observing the effect of the amount of electric voltage on the production of hydrogen gas and the removal of phenol can be seen in Figure 3. The greater the electric current is directly proportional to the number of electrons flowing towards the cathode, the more reaction of forming hydrogen gas on the surface of the cathode occurs. As a unitary oxidation-reduction reaction, an increase in the reduction reaction for the formation of hydrogen at the cathode will affect the oxidation reaction on the anode surface so that the aluminium electrode as the anode is more and more oxidized

to Al^{3+} , which then forms $\text{Al}(\text{OH})_3$ coagulant and increases the effectiveness of dissolved CIP adsorption.

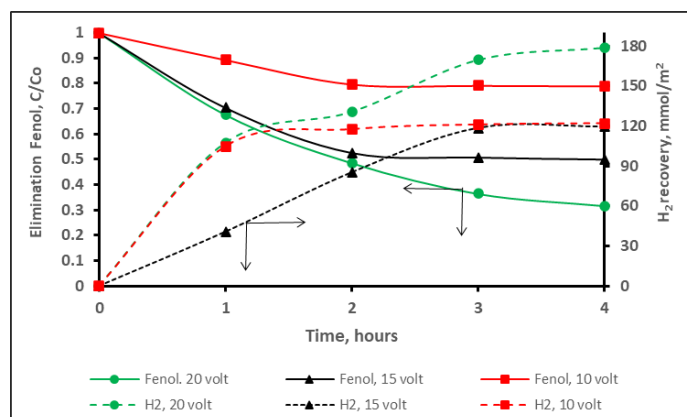


Figure 3. Comparison of the performance of the electrocoagulation process at various values of electric current voltage (phenol concentration 5 ppm, solution pH 10)

The amount of voltage used also affects the processing time, which gives effective results. It can be seen in Figure 3 that the greater the voltage, in addition to providing better performance results, it takes longer time to reach the optimal performance value. A 10-volt mains voltage only takes 1 hour of operation to reach its optimal value. In contrast, at a 20-volt mains voltage for up to 4 hours of operation, there is still a tendency to increase hydrogen production and phenol removal. The operation of electrocoagulation is limited by the optimal time, which at a more extended time does not significantly increase results.

3.3. Times

From the observations shown in Figure 2 and Figure 3, it can be seen that the electrocoagulation process does not take up to 4 hours to achieve optimal results. In observing the pH, it can be seen that when pH 10 is used as the optimal value, the hydrogen recovery reaches a relatively constant value after operating for 3 hours, while the removal of phenol decreases significantly. Meanwhile, when observing the electric current voltage, when using a voltage of 20 volts, the hydrogen gain reached a constant value after operating for 3 hours. The removal of phenol reached a relatively constant value after 3 hours of operation.

It is necessary to determine the optimal conditions, including the solution's pH and the electric current voltage, simultaneously providing maximum results at the suitable processing time range. Based on the results of FTIR observations made on processed coagulants, the results are displayed according to the Fig 4. The appearance of peaks at the wavenumber values 3258.20, 1636.17, 1395.39, 1061.91 cm^{-1} . Based on this, it was confirmed that the presence of an OH group (3258.20 cm^{-1}), the presence of a C=C double bond (1636.17 cm^{-1}), a C-C single bond (1507.82 cm^{-1}), and a C-O bond (1061.91 cm^{-1}). Thus the adsorptive coagulant is proven capable of eliminating phenol ($\text{C}_6\text{H}_5\text{OH}$) dissolved in solution, as evidenced by the presence of groups and types of carbonic bonds that correspond to the bonds in phenolic compounds.

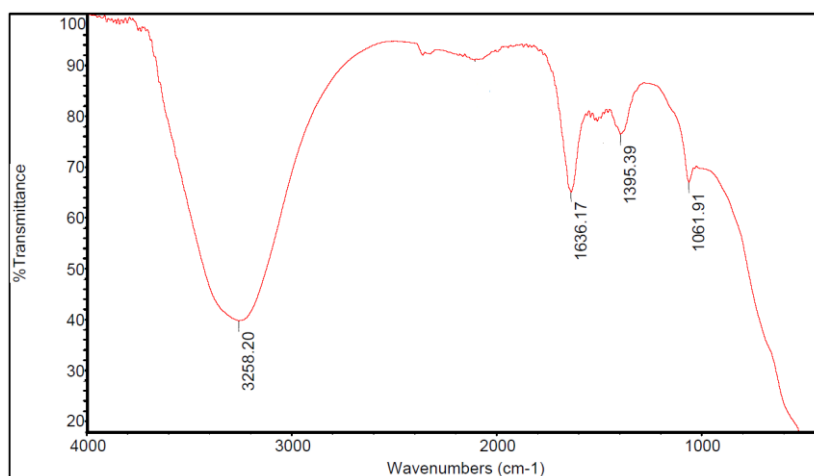


Figure 4. The FTIR spectra of coagulants product from electrocoagulation process

4. Conclusions

Electrocoagulation has been proven to have the ability to remove dissolved phenols as a representation of the petroleum industry's liquid waste. In addition, during the process, hydrogen gas is also obtained, which can be used as an alternative energy source. It was found that the condition of the sample solution at pH 10 and using an electric voltage of 20 volts gave optimal results in removing up to 22% phenol and hydrogen production of 123.31 mmol/m². The optimal gain was obtained at a process duration of 3 hours and did not show a significant increase when the additional time was applied. Further research is needed to improve the ability of the process to remove dissolved pollutants and observe waste samples containing multi-component.

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Electrocoagulation process as an alternative treatment of petroleum industry liquid waste and renewable energy sources

by Fadliah Lia

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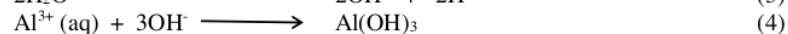
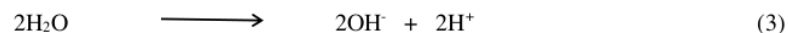
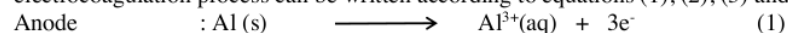
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2. Methods

The research was conducted on a laboratory scale using a sample solution containing pollutants from various phenol compositions. The anode uses a 1.5 mm (4x8cm) thick Aluminum metal plate and a 1mm (4x8cm) Stainless Steel 316 (SS-316) plate as a cathode. The electrocoagulation reactor was a 500 mL acrylic vessel connected to a direct current power source with a maximum of 50 V. The process of hydrogen gas recovery and conversion of pollutant removal in solution was observed for 4 hours. Observations were made at different initial pH, namely 5, 7, and 10. The pH adjustment was made by adding 0.1M NaOH until it reached the desired acidity level.

Pollutant concentrations were analysed using a UV-Vis spectrophotometer with a Bel UV-M51 Spectrophotometer. Gas chromatography analysed hydrogen production using Shimadzu GC-2014, MS-5A column, and Ar carrier gas. The electrocoagulation process is presented in Figure 1, where the sample solution is put into the reactor (a). Electrodes (d and e) are placed in the reactor while connected to the power supply (g) at 10 - 20 V. Meanwhile, a hot plate with magnetic stirrer (f) is used to ensure the homogeneity of the solution.



Figure 1. The apparatus of electrocoagulation to eliminate phenol solution and produce hydrogen

The total pollutant concentration and the amount of hydrogen production were used as the variables observed during the process, which was carried out within 4 hours. Variable measurements take liquid samples at valve (c) and gas samples at valve (b) every 1 hour. Pollutant concentrations in wastewater were measured using UV-VIS spectrophotometry, while the amount of hydrogen gas was measured using a GC (Gas Chromatograph).

3. Result and Discussion

The performance of the electrocoagulation process using aluminium electrodes has been observed to remove samples of liquid waste containing phenol as a representation of the liquid waste from the petroleum industry. In addition, the ability of the process to produce hydrogen gas was also observed. The process is carried out using a direct current power source with an electric voltage of 10 to 20 volts

for 4 hours for each observation. The independent variables used at this stage are the pH of the sample solution, the electric current voltage, and the processing time. Changes in pollutant concentration and the amount of hydrogen gas produced at intervals of 1 hour were measured. The observation results obtained are as follows:

3.1. The effect of the pH of the solution

In the electrocoagulation process with aluminium electrodes, efficient coagulant formation occurs at a pH range of 6 – 7. The electrocoagulation process can occur at a more comprehensive pH range [7, 8]. Aluminium amphoteric electrodes also help stabilize acidity with their ability to neutralize pH [7]. This results in an electrocoagulation process involving a solution with a low pH that will increase in pH during the process; conversely, if a solution with a high pH is used, there will be a decrease in pH.

Observations were made at low acidity and alkalinity levels, namely at pH 5, 7 and 10, respectively. It is known that the process carried out in the pH range of 4 – 10 does not affect the decrease in the amount of current strength [8]. The observation results are shown in Figure 2, where it can be seen that increasing the pH increases the ability of the process to remove CIP and produce hydrogen gas.

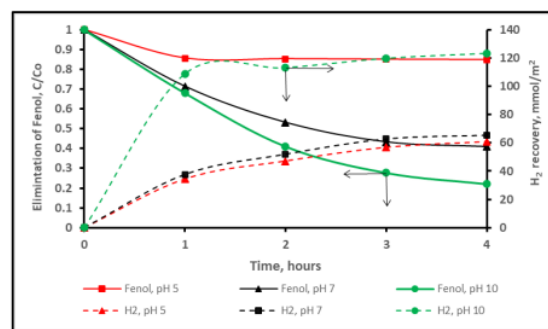


Figure 2. The effect of pH on the ability of the process to remove phenol and produce hydrogen gas (5 ppm phenol solution, 20 volt power supply)

The presence of abundant OH^- ions in a solution with a pH of 10 allows the formation of more $\text{Al}(\text{OH})_3$ coagulants, thus increasing the amount of pollutant that can be adsorbed. Under very alkaline conditions, the greater the occurrence of aluminate ions ($\text{Al}(\text{OH})_4^-$) as a result of excess OH^- in solution according to equation (5) [9],



The presence of aluminate ions makes it possible for hydrogen gas to form because aluminate ions are known to have a good ability to reduce H^+ cations. However, their adsorption capacity is low compared to $\text{Al}(\text{OH})_3$ [8].

3.2. Electric current voltage

The amount of electric current used dramatically affects the ability of the anode electrode to dissolve in the form of ions, which is directly proportional to the amount of coagulant that occurs [4, 6]. The more significant the electric current, the more coagulant occurs until the optimum electric current value is reached. When using an electric current more excellent than its optimum value, there is no significant coagulant addition [10, 11].

The results obtained from observing the effect of the amount of electric voltage on the production of hydrogen gas and the removal of phenol can be seen in Figure 3. The greater the electric current is directly proportional to the number of electrons flowing towards the cathode, the more reaction of forming hydrogen gas on the surface of the cathode occurs. As a unitary oxidation-reduction reaction, an increase in the reduction reaction for the formation of hydrogen at the cathode will affect the oxidation reaction on the anode surface so that the aluminium electrode as the anode is more and more oxidized

to Al^{3+} , which then forms $\text{Al}(\text{OH})_3$ coagulant and increases the effectiveness of dissolved CIP adsorption.

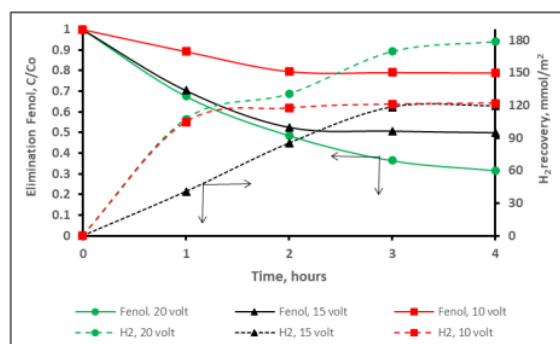


Figure 3. Comparison of the performance of the electrocoagulation process at various values of electric current voltage (phenol concentration 5 ppm, solution pH 10)

The amount of voltage used also affects the processing time, which gives effective results. It can be seen in Figure 3 that the greater the voltage, in addition to providing better performance results, it takes longer time to reach the optimal performance value. A 10-volt mains voltage only takes 1 hour of operation to reach its optimal value. In contrast, at a 20-volt mains voltage for up to 4 hours of operation, there is still a tendency to increase hydrogen production and phenol removal. The operation of electrocoagulation is limited by the optimal time, which at a more extended time does not significantly increase results.

3.3. Times

From the observations shown in Figure 2 and Figure 3, it can be seen that the electrocoagulation process does not take up to 4 hours to achieve optimal results. In observing the pH, it can be seen that when pH 10 is used as the optimal value, the hydrogen recovery reaches a relatively constant value after operating for 3 hours, while the removal of phenol decreases significantly. Meanwhile, when observing the electric current voltage, when using a voltage of 20 volts, the hydrogen gain reached a constant value after operating for 3 hours. The removal of phenol reached a relatively constant value after 3 hours of operation.

It is necessary to determine the optimal conditions, including the solution's pH and the electric current voltage, simultaneously providing maximum results at the suitable processing time range. Based on the results of FTIR observations made on processed coagulants, the results are displayed according to the Fig 4. The appearance of peaks at the wavenumber values 3258.20 , 1636.17 , 1395.39 , 1061.91 cm^{-1} . Based on this, it was confirmed that the presence of an OH group (3258.20 cm^{-1}), the presence of a C=C double bond (1636.17 cm^{-1}), a C-C single bond (1507.82 cm^{-1}), and a C-O bond (1061.91 cm^{-1}). Thus the adsorptive coagulant is proven capable of eliminating phenol ($\text{C}_6\text{H}_5\text{OH}$) dissolved in solution, as evidenced by the presence of groups and types of carbonic bonds that correspond to the bonds in phenolic compounds.

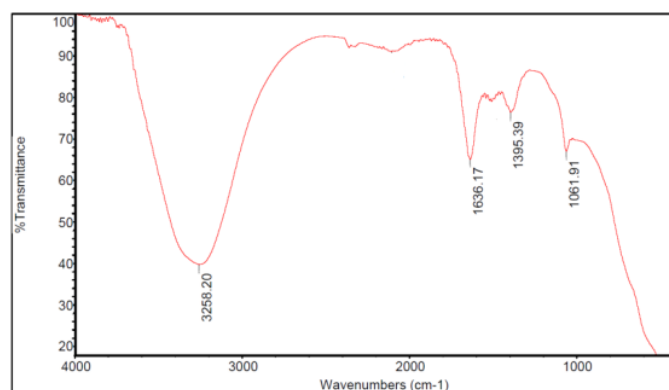


Figure 4. The FTIR spectra of coagulants product from electrocoagulation process

4. Conclusions

Electrocoagulation has been proven to have the ability to remove dissolved phenols as a representation of the petroleum industry's liquid waste. In addition, during the process, hydrogen gas is also obtained, which can be used as an alternative energy source. It was found that the condition of the sample solution at pH 10 and using an electric voltage of 20 volts gave optimal results in removing up to 22% phenol and hydrogen production of 123.31 mmol/m². The optimal gain was obtained at a process duration of 3 hours and did not show a significant increase when the additional time was applied. Further research is needed to improve the ability of the process to remove dissolved pollutants and observe waste samples containing multi-component.

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