Methane Gas Monitoring System (CH$_4$) in Chemical Reactors

Samsul Arifin[1], Suryono Suryono[2], Jatmiko Endro Suseno[2]

[1]*Magister of Physics, Faculty of Science and Mathematics, Diponegoro University, Semarang, Indonesia*
[2]*Department of Physics, Faculty of Science and Mathematics, Diponegoro University, Semarang, Indonesia*

Abstract: Methane gas is one of the gases produced in chemical reactors. Methane gas concentrations need to be measured in order to be safe and controlled to produce optimal chemical processes. In addition, the large concentration of methane gas can be harmful to health. At present it is difficult to obtain a methane gas concentration (CH4) data acquisition system that can store and display data for a long time. Therefore, in this research, an instrumentation system for measuring methane gas concentration that works digitally and computerize is created. This system consists of sensors, computer interface systems and computers as data storage and display. The sensor used for methane gas measurement uses the semiconductor type SnO2 which converts the gas concentration into voltage. The sensor signal is sent to the computer via a computer interface consisting of an Analog to Digital Converter (ADC) and a serial data communication system. ADC and serial data communication systems are built using the Arduino Due microcontroller which is programmed using the C programming language. Computer programs are made to obtain concentration, store data and display it on the monitor. The results of this study indicate that the methane gas concentration can be acquired via a computer, can be stored in data records and can display the conditions of the methane gas concentration during a chemical reaction.

Keywords: methane gas, chemical reactor, sensor, data acquisition, gas concentration

I. Introduction

Methane gas is produced a lot in a chemical process, for example in research on the production of biogas mangosteen peel, it shows that mangosteen peel contains methane gas [1]. This is a research reference to obtain the relationship between methane gas produced when extracting mangosteen peel and the compounds produced.

Another study on mangosteen peel extraction using Ultrasound Assisted Extraction (UAE) by varying physical variables at the time of extraction showed that mangosteen peel contains xhantone compounds. The results of extraction of mangosteen peel with Ultrasound Assisted Extraction (UAE) with the procedure to manage several variations, namely frequency, temperature, and sonication time [2].

Methane gas concentration measurement in the biogas reactor obtained a graph of methane gas concentration (CH4). The graph of methane gas against extraction time was observed to obtain a profile of methane gas produced during the process. Methane gas monitoring data acquisition system needs to be observed with a computer so that it can be seen the increase in methane gas concentration per second.

CH4 gas is the simplest hydrocarbon compound in the form of a colorless and odorless gas with the chemical formula CH$_4$. In addition, other properties of CH4 gas, namely that it can burn at levels between 5-15%, has a molecular weight of 16.04 grams / mol and a specific gravity of 0.554, a boiling point of -161 ° C and has a water solubility of about 35 mg / L at pressure 1 atmosphere. CH4 gas can cause greater global warming, besides that CH4 gas cannot be absorbed by chlorophyll in plant plants so that it is more stable in the atmosphere than CO2 gas which can be absorbed by plants through photosynthesis [3].

II. Theory

2.1 Methane

CH4 gas is one of the greenhouse gases with a global warming potential index of 21 times the CO2 molecule. Because the greater the heating potential, it will cause the earth's temperature to get hotter. CH4 gas emissions can come from natural sources and anthropogenic activities. Natural sources of CH4 gas include wetlands, lakes, rivers, which are fermented by bacteria. Meanwhile, CH4 gas from anthropogenic activities comes from the agricultural sector, livestock, II-8 household domestic waste (septic tanks), reservoirs, temporary (TPS) and final (TPA) processing sites [4].

Anthropogenic activity is estimated to account for approximately 60% of CH4 emissions to the atmosphere [5]. The level of CH4 gas emissions can vary significantly from one country or region to another, depending on many factors such as climate, characteristics of industrial and agricultural production, energy types and uses and waste management. The application of technology to capture and utilize CH4 gas from sources such as landfills, coal

mines and fertilizer management systems affects the level of emissions from these sources (US-EPA, 2011). CH_4 gas is a greenhouse gas that is stronger than CO_2. However, the concentration of CH_4 gas is smaller than other greenhouse gases [6].

2.2 Gas Sensor
The sensor is an electronic device that can convert physical quantities into electrical signal. The sensor can be used as an input device for physical quantities to data processing devices such as microcontrollers, computers, Programable Logic Controllers (PLC), Distributed Control Systems (DCS), and Supervisory Control and Data Acquisition (SCADA). Sensors that are often used in industry include weight sensors, temperature sensors, vibration sensors, electromagnetic sensors, flow and water level sensors, humidity sensors, chemical sensors, capacitive and inductive sensors, and many other types. In an instrument system, there is a part that has a working principle in contrast to a sensor called an actuator, which is a device that converts an electrical signal into other physical quantities. For example, a loudspeaker is an actuator that converts electrical signals into sound pressure [7].

SnO_2 is an n-type wide bandgap metal oxide semiconductor which is widely applied as a base material for gas sensors [8]. The electrical properties of nanocrystalline SnO_2 depend entirely on the crystal size and the surface conditions produced by the gas absorption resulting in space appearance and band modulation [9]. SnO_2 has an ionic bond with 4 electrons. Sn donates 2 electrons each to the O atom. The presence of this ionic bond makes the SnO_2 material more sensitive to changes in adsorbed gas molecules than covalent bonds [10], the ionic bond of SnO_2 can be described by equations (1), (2) and (3) .

$$Sn \rightarrow Sn^{4+} + 4e^- \tag{1}$$

$$O + 2e^- \rightarrow O^{2-} \qquad \text{x 2} \tag{2}$$

$$\overline{Sn + 2O \rightarrow Sn^{4+} + 2O^{2-}} \tag{3}$$

With the response of $SnO2$ to different types of gas, we can divide it into two major groups, the first group consisting of CO, H2, and CH4 which provide reducing properties and easily increase the conductivity of the material. In the second group there are various gases such as NO2 and CO2 which have oxidizing properties and can oxidize the material and reduce the conductivity of the material.

2.3 Data Acquisition System
Sensor data acquisition is a process of collecting information from a number of sensor output signal data, converting it into digital data and sending the data to a computer for storage, processing, and display of data on information display devices or printed on a printer. Sensor data acquisition architecture is shown in Figure 1.
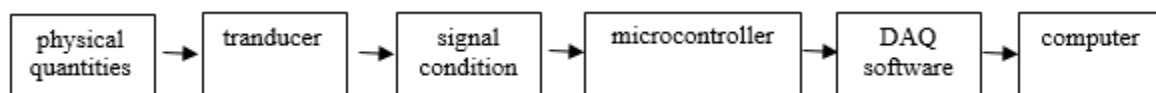

Figure 1. Diagram of the sensor data acquisition system

2.4 Microcontroller
The microcontroller is a chip that can perform digital data processing in accordance with the command program it provides. These commands are in the form of machine code used to run them and must comply with the code provided by the manufacturer. The code can be in the form of a computer program in assembly language or other translational languages that have been created by software companies, for example, C language, basic, or languages from other developers.

The AT91SAM3X8E microcontroller has an Analog to Digital Converter (ADC) facility built into the chip. ADC owned by AT91SAM3X8E has 12 multiplex analog input channels, and has ADC and PWM resolution of 12 bits.

This means that the analog voltage between 0 Volt and 3.3 Volt is coded into one of 4096 binary representations with a susceptible value of 0 to 4095. The conversion result of ADC on the AT91SAM3X8E microcontroller in the Arduino Due system board is determined by the equation:

$$\text{ADC} = \frac{Vin}{Vref} \times (2n - 1) = \frac{Vin}{Vref} \times 4095 \tag{4}$$

Where Vin is the voltage at the selected input pin, Vref is the selected reference voltage, and n is the number of bits.

III. Method

The first step is to build an instrumentation system composed of software and hardware. Hardware used by using the MQ4 gas sensor using a data acquisition system to a computer. The measurements were varied with 5 minutes, 10 minutes, 15 minutes and 20 minutes. The instrumentation system design diagram is shown in Figure 2.
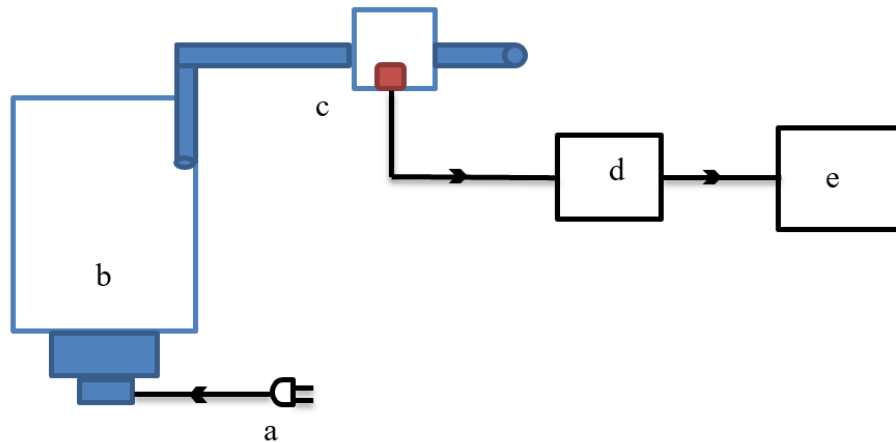
Figure 2. Diagram of instrumentation system design. a: 220 volt AC input voltage, b: chemical reactor, c: methane gas sensor, d: microcontroller, e: computer.

The workings of a chemical reactor are described in the instrumentation system design diagram. When the reactor is turned on, it will produce extraction and produce gas in the description b. then the gas will come out and be measured in Figure c which is used is the MQ4 gas sensor. Instrumentation system to acquire data from methane gas sensors and then displayed on a computer. The data acquisition system stores data in real team with graphic display on the monitor. Methane gas discharge data is stored in the data acquisition system with various measurement times.

The microcontroller used in letter d is Arduino Due which is processed using an interface system to the computer. Before the measurement, characterization and calibration were carried out on the Analog to Digital converter (ADC) and the MQ4 gas sensor. Then carried out several experiments to find the data generated in the chemical reactor.

IV. Results and Discussion

4.1. Characterization and Calibration of Analog to Digital Converter (ADC)

Figure 3 is an ADC test scheme using a potentiometer for ADC characterization, which is done by measuring the ADC input voltage (mV) with the ADC weight value on the monitor serial varied with the potentiometer. Then a linear graph is created to find the gradient which functions to find the x value in the ADC value calibration.
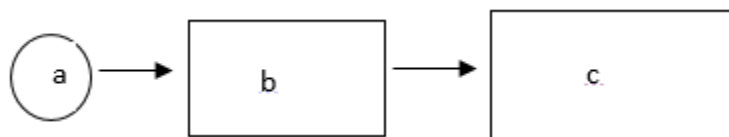

Figure 3. The ADC test scheme using a potentiometer, a: potentiometer, b: Analog to Digital Converter, c: monitor
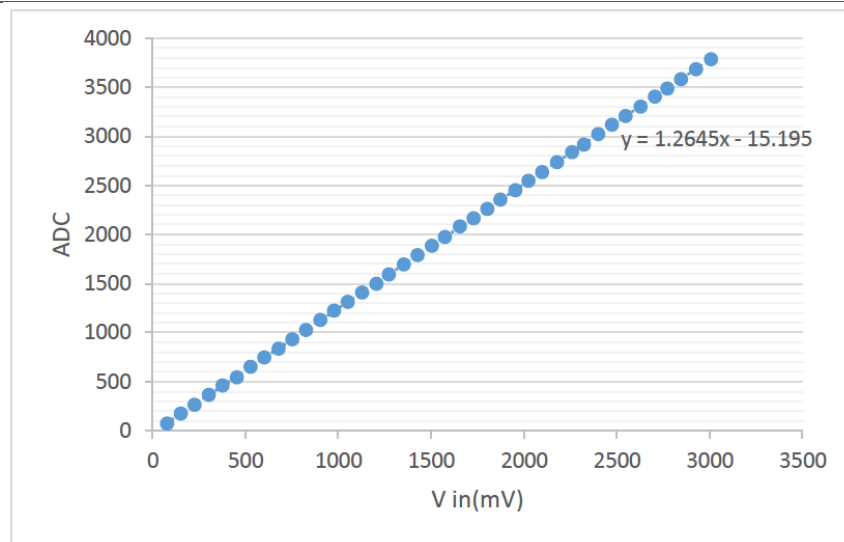
Figure 4. ADC characterization graph

As in the Figure above, the data between the input voltage (mV) and ADC weight is explained, so that the linearity of the data can be obtained with a gradient of y = 1.2645x - 15.195. From this characterization, then calibration is carried out to determine the average error, which can be seen in Table 1.

Calibration aims to determine the error value in the methane gas concentration measuring instrument after characterizing the ADC used. To find the value of the ADC voltage, it is measured by a multimeter which is set with a potentiometer. Calibration results are shown in Table 1.

Table 1. ADC Calibration

No.	Vin (mV)	Reference Vin (mV)	Error (mV)
1	75.5	68.8	6.7
2	151.4	144.8	6.6
3	225.5	220.58	4.92
4	301.5	289.9	11.6
5	374.5	364.1	10.4
6	454	456.64	2.64
7	528	530.8	2.8
8	600	603.08	3.08
9	673	675.22	2.22
10	747	751.44	4.44
11	824	825.14	1.14
12	902	897.48	4.52
13	976	974.76	1.24
14	1057	1060.14	3.14
15	1132	1131.7	0.3
16	1225	1228.28	3.28
17	1317	1318.04	1.04
18	1405	1405.26	0.26
19	1514	1512.28	1.72
20	1690	1690.84	0.84
21	1797	1796.06	0.94
22	1893	1896.16	3.16
23	1990	1989	1
24	2142	2141.08	0.92
25	2244	2242.32	1.68
26	2501	2501.68	0.68
27	2811	2811.24	0.24

28	2992	2990.14	1.86
29	3185	3182.58	2.42
30	3245	3241.16	3.84
Average error			2.987333333

The ADC calibration data in Table 1 describes the average ADC calculation error with a value of 2.987333333 mV.

4.3 Characterization and Calibration of The Gas Sensor
Gas sensor characterization using the datasheet on the sensor. Figure 5 shows a series of gas sensors used to measure the concentration of methane gas. The circuit is based on Ohm's Law for characterization using the sensor datasheet. In free air, we know RS using Ohm's Law.
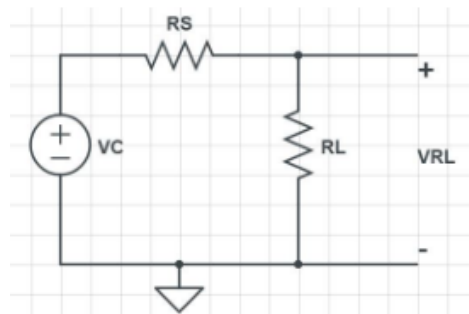

Figure 5. Series of Gas Sensors [12]

On Ohm's law
$I = V / R$
$I = VC / (RS + RL)$
$V = I \times R$
$VRL = [VC / (RS + RL)] \times RL$
$VRL = (VC \times RL) / (RS + RL)$
To find a hospital:
$VRL \times (RS + RL) = VC \times RL$
$(VRL \times RS) + (VRL \times RL) = VC \times RL$
$(VRL \times RS) = (VC \times RL) - (VRL \times RL)$
$RS = [(VC \times RL) - (VRL \times RL)] / VRL$
$RS = [(VC \times RL) / VRL] - RL$
From the graph on the sensor datasheet it can be seen that the resistance ratio in fresh air is constant:
$RS / R0 = 4.4$ ppm
The chart scale is log. This means that on a linear scale, the behavior of gas concentration to resistance ratio isexponential.
$y = mx + b$
Where:
y: the value of X.
x: the value of X.
m: the slope of the line
b: Y interception
For a log scale, the formula is:
$\log (y) = m * \log (x) + b$
Note: log is base 10.
$m = [\log (y) - \log (y0)] / [\log (x) - \log (x0)]$
$m = \log (y / y0) / \log (x / x0)$
$m = \log (0.75 / 2.6) / \log (10000/200)$
$m = -0.318$
$\log (y) = m * \log (x) + b$
$b = \log (y) - m * \log (x)$
$b = \log (0.9) - (-0.318) * \log (5000)$

b = 1.13
x = 10 ^ {[log (y) - b] / m}
x = methane gas concentration (ppm)

4.4 Results of Methane Gas Concentration Measurement
      The results of monitoring methane gas monitoring in the chemical reactor can be seen in Figures 6, 7, 8, 9 and 10. Measurements with the MQ4 gas sensor varied with a time of 5 minutes, 10 minutes, 15 minutes and 20 minutes.
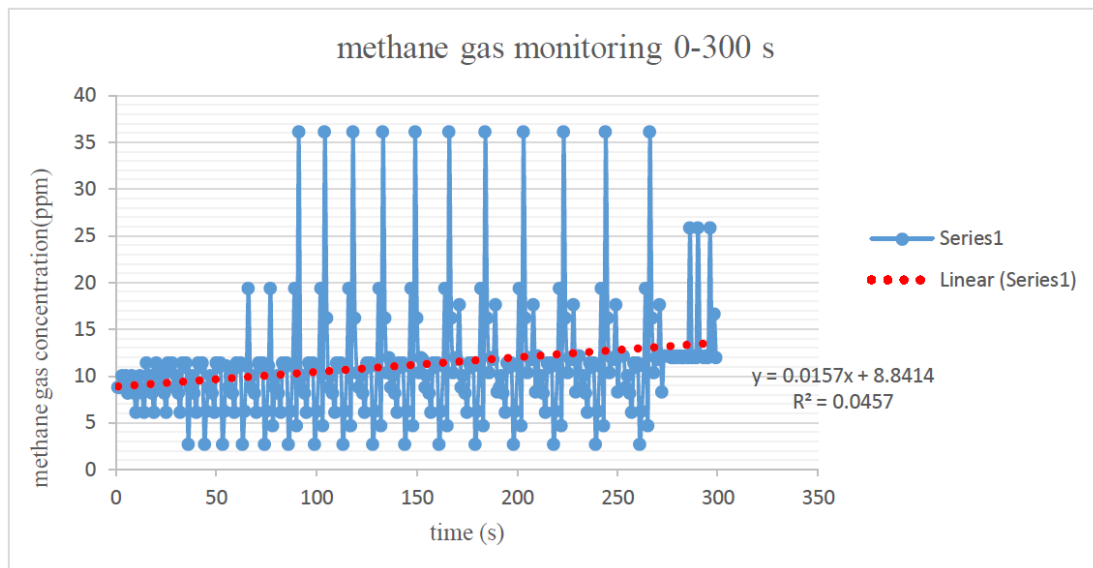

Figure 6. The results of monitoring the concentration of methane gas against a time of 0 to 300 s

      Methane gas concentration measurements in the chemical reactor show a linear line with respect to 0 to 300 s. The concentration data obtained by methane gas on the graph has increased even though it is not stable. This shows the reactor activity against the extracted material. When the methane gas concentration is at the highest or lowest point, it shows the sensitivity of the gas sensor used. From the data, all the results obtained an average linear increase.
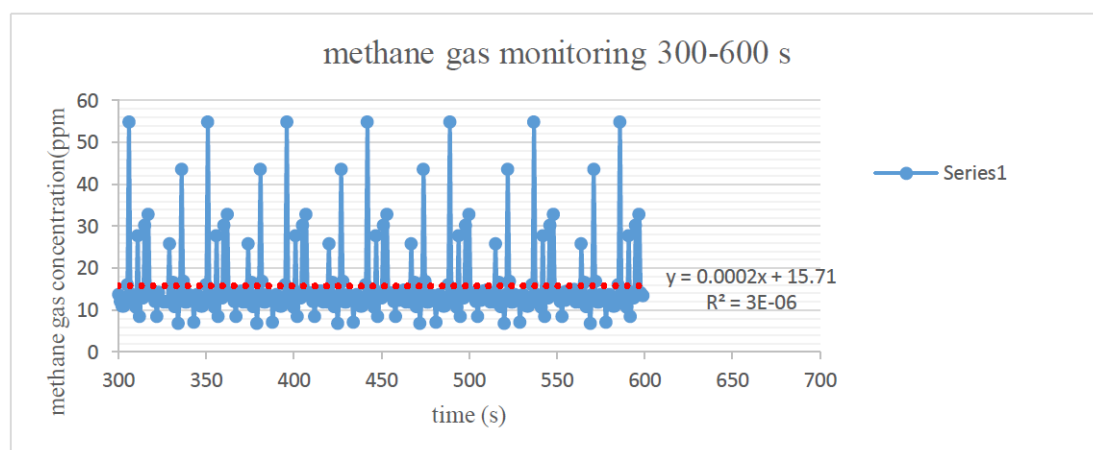

Figure 7. The results of monitoring the concentration of methane gas over a period of 300 s to 600 s

      Methane concentration measurements in the chemical reactor show a linear line over time of 300 s to 600 s. The concentration data obtained by methane gas on the graph has increased even though it is not stable. This shows the reactor activity against the extracted material. When the methane gas concentration is at the highest or lowest point, it shows the sensitivity of the gas sensor used. From the data, all the results obtained an average linear increase.
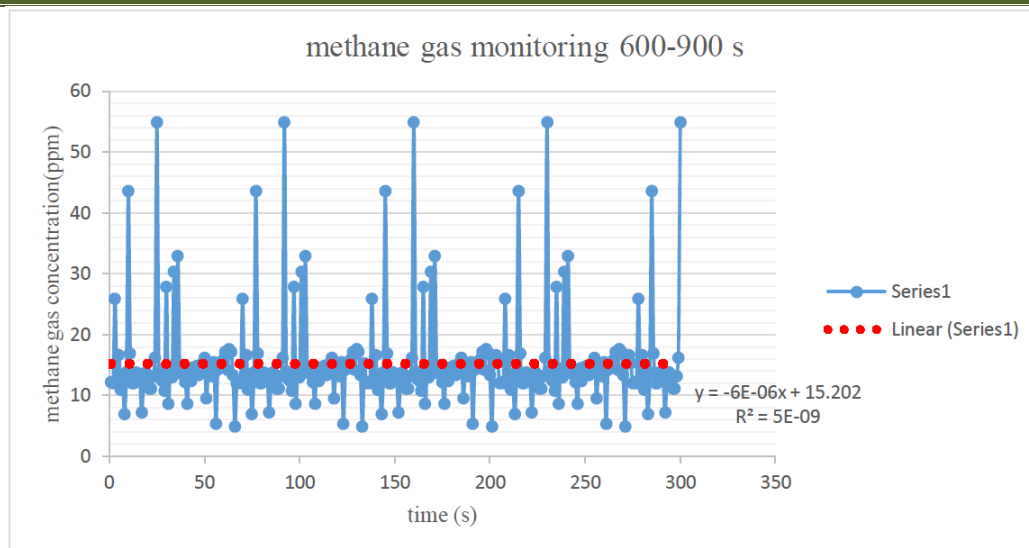
Figure 8. The results of monitoring the concentration of methane gas from 600 s to 900 s

Methane concentration measurements in the chemical reactor show a linear line with respect to 600 s to 900 s. The concentration data obtained by methane gas on the graph has increased even though it is not stable. This shows the reactor activity against the extracted material. When the methane gas concentration is at the highest or lowest point, it shows the sensitivity of the gas sensor used. From the data, all the results obtained an average linear increase.
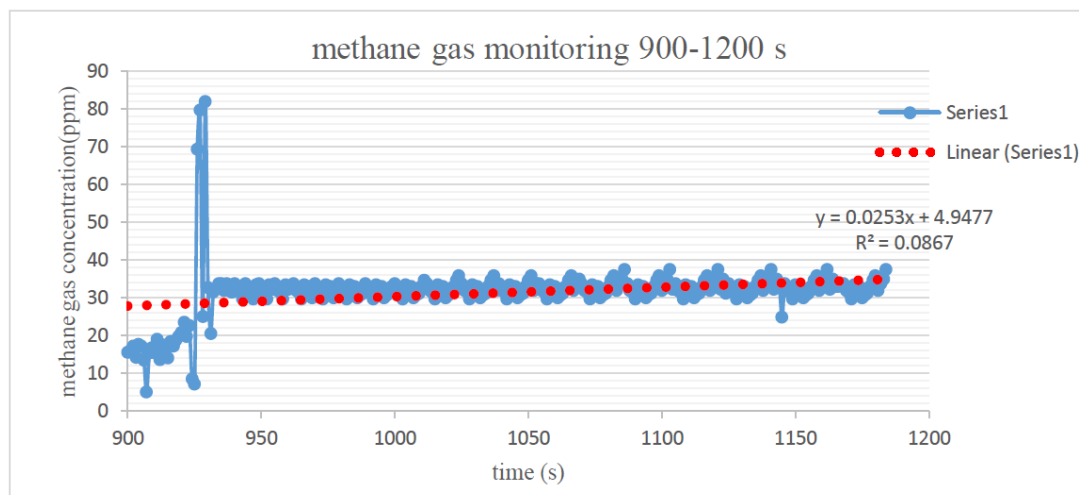

Figure 9. The results of monitoring the concentration of methane gas from 900 s to 1200 s

Methane concentration measurements in the chemical reactor show a linear line over the time of 900 s to 1200 s. The concentration data obtained by methane gas on the graph has increased even though it is not stable. This shows the reactor activity against the extracted material. When the methane gas concentration is at the highest or lowest point, it shows the sensitivity of the gas sensor used. From the data, all the results obtained an average linear increase. Methane concentration data at 900 seconds and above show almost the same value up to 1200 seconds.
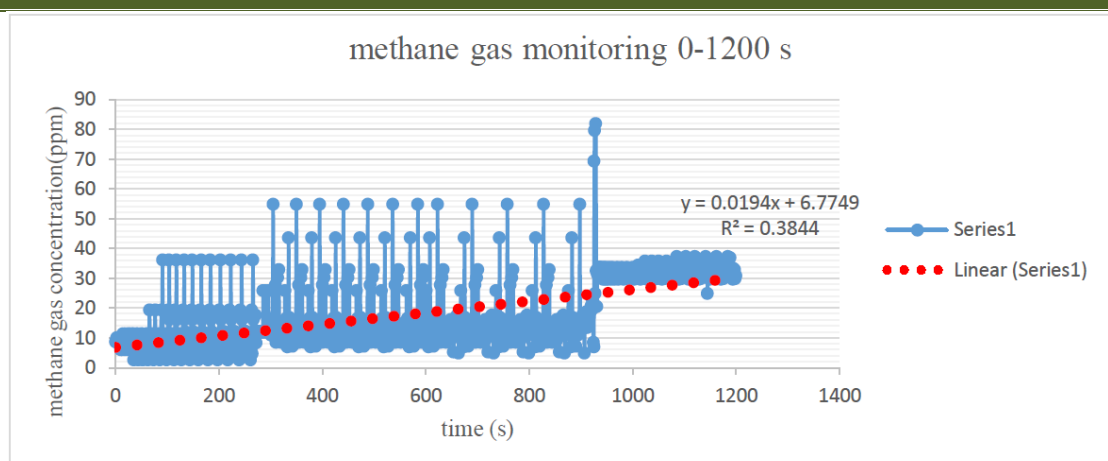
Figure 10. The results of monitoring the concentration of methane gas against a time of 0 to 1200 s

Methane gas concentration measurements in the chemical reactor show a linear line with respect to 0 to 1200 s. The concentration data obtained by methane gas on the graph has increased even though it is not stable. This shows the reactor activity against the extracted material. When the methane gas concentration is at the highest or lowest point, it shows the sensitivity of the gas sensor used. From the data, all the results obtained an average linear increase. From some data, it was found that the increase in the concentration of methane gas with time, at 900 s to 1200 s did not experience a significant increase.

V. Conclusion

From the results of research on measuring the concentration of methane gas in chemical reactors using an instrumentation system based on the Arduino Due microcontroller, there is a conclusion obtained by the researchers, there is a linear increase in the concentration of methane gas with time. Increment data between 8 ppm to 30 ppm. In the extraction conditions, the time from 900 s to 1200 s did not experience a significant increase. This instrumentation system can be used as a measurement reference at the time of extraction in a chemical reactor.

From the research results, there are suggestions from researchers to be further developed. Among them for further research, it is hoped that the characterization of the measuring instrument will be carried out using a calibrator measure the concentration of methane gas in real time. At the time of characterization measurement, it is better to use pure methane gas so that there are no other gases that can interfere with the voltage of the methane gauge that will be generated at the time of characterization. Test the compound content in the resulting chemical reactor extraction.

References

[1]. Laksi, K. M., 2015, *The Effect of Preliminary Treatment of Mangosteen Skin (Garciniamangostana Linn.) With Sodium Hydroxide at Low Temperature on Biogas Production KINANTHI MONDY LAKSI*. (September), 837–842.

[2]. Suryono, S., *Hadiyanto*, H., Yasin, M., Widyowati, R., Muniroh, M., &Amalia, A. 2019, *Effect of frequency, temperature, and time of sonication on xanton content of mangosteen (Garciniamangostana L.) peel extract through ultrasound assisted extraction.* E3S Web of Conferences, 125(201 9), 9–11. https://doi.org/10.1051/e3sconf/201912525006.

[3]. Wahyuni, T. (2016). Methane Emissions from Manure Management in Yogyakarta – Inventory. Environmental Engineering.,FakultasTeknik - UniversitasAndalas Padang - *Universitas*Andalas.

[4]. Slamet, L. (2014). Potential Methane (CH4) Emissions to the Atmosphere Due to Floods. Aerospace News, 15 (1).

[5]. Hauweli*ng*, S.T, Rockman, I, Aben, F, Keppler, M, Krol, J.F, Meirink, E.J, Dlugokenckydan C. Frankenberg. (2006). Atmospheric Constrains on Global Emissions of Methane From Plants. Geophys. Res. Lett. 33. L15821, doi:10.1029/2006GL026162.

[6]. Akorede, M, Hizam, H, Ab-Kadir, M.Z, Aris, I, danBuba, S.D. (2012). Mitigating TheAnthropogenis Global Warming In The Electric Power Industry. Journal of Renewable and Sustainable Energy Review, 2747-2761.

[7]. Suryono, 2018. Teknologi*Sensor: Physical Concepts and Data Acquisition Techniques Based on 32 Bit Microcontroller ATSAM3X8E (ARDUINO DUE)*, UNDIP Publisher, Semarang.

[8]. MOSELEY, P.T., Materials Selection for Semi-conductor Gas Sensors, Sensor and Actuators B, 6, 149-156, 1992.

[9]. Safonofa, O. V., Delabouglise, G., Cheinever, B., Gakov, A.M., Lebeau, M., CO and NO2 gas Sensitivity of Nano-cristaline, Tin Dioxide thin Films Doped with Pd, Ru, and Rh, , 2002.

[10]. Gaskov, A.M., Rumyantseva, M.N., Material for Solid Gas Sensor. Inorganic Material, Vol,36, No. 293-301, 1999.

[11]. Suryono, S., Putro, S.P., Widowati, W., danAdhy, S., 2017, *System on Chip (SOC) Wi-Fi Microcontroler for Multi Station Measurement of Water Surface Level using Ultrasonic Sensor, IOPscience: ISNPINSA* 2017.

[12]. Setiawati, D.A., Dwi Putra, G. M., Sugandi, K.W., 2017. *Performance Test of Biogas Volume Monitoring System Based on Arduino Microcontroller on Floating Drum Type Biodigester*. Agricultural Engineering Scientific Journal and Biosystems Vol.5, No.2.