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HYDROGEN AND METHANE PRODUCTION FROM WHEY

Summary

Decreasing amount of fossil fuels in the world encourages the searching of alternative energy sources. In this time of energetic crisis, the production of hydrogen is an interesting solution. Hydrogen does not produce any contaminating emission. The aim of this study was to build a project installation that produces gas biofuels and define the potential biohydrogen and biogas possible to produce from the waste of a dairy plant. The calculations assume a production of 400 m^3 per day of whey permeate from the dairy plant. The methane fermentation process was carried out according to the modified German standard DIN 38 414/S8 in the eco-technology laboratory in the Poznan University of Life Sciences. The results revealed that, with the assumed quantity of available substrate, it is possible to generate 1 570 960 m^3 of hydrogen per year and 4 749 469 m^3 of biogas with a methane percentage of approx. 49%. Based on these results it could be possible to build a biogas plant of an estimated power of 0,99 MW of electricity and 1,12 MW of heat, as well as the hydrogen fuel cell power of 0,32 MW of electricity.
***Key words**: power engineering, biohydrogen, hydrogen production, biogas, methane*

WODÓR I METAN PRODUKOWANY Z SERWATKI

Streszczenie

Kończące się zasoby paliw kopalnych skutkują sytuacją, w której świat staje w obliczu konieczności poszukiwania nowych, alternatywnych źródeł energii. W czasach kryzysu energetycznego interesującym rozwiązaniem wydaje się być produkcja i wykorzystanie wodoru, który zarówno w wyniku spalenia, jak i wykorzystania w ogniwie paliwowym nie emituje zanieczyszczeń środowiska. Celem pracy było określenie możliwych do wyprodukowania ilości biowodoru oraz biogazu z mleczarskiego odpadu poprodukcyjnego. W obliczeniach uwzględniono umiejscowienie instalacji przy zakładzie mleczarskim produkującym dziennie 400 m^3 permeatu serwatkowego. Wykorzystano ponadto wyniki badań przeprowadzonych w Pracowni Ekotechnologii w Poznaniu uzyskane na podstawie analiz wykonanych zgodnie z obowiązującą niemiecką normą DIN 38 414/S8. Na potrzeby obliczeń posłużono się także danymi zamieszczonymi w najnowszej literaturze przedmiotu. Na podstawie uzyskanych wyników wykazano, że z zakładanej ilości dostępnego substratu możliwe będzie wytworzenie rocznie 1 570 960 m^3 wodoru oraz 4 749 469 m^3 biogazu o procentowej zawartości metanu ok. 49%. W oparciu o te dane obliczono realną moc biogazowni na poziomie 0,99 MW energii elektrycznej oraz 1,12 MW ciepła, a także moc ogniwa paliwowego wynoszącą 0,32 MW energii elektrycznej.
***Słowa kluczowe**: energetyka, biowodór, produkcja wodoru, biogaz, metan*

1. Introduction

The progress of civilization and the increasing population is resulting in continuing growth of the demand for electricity and fuel, which is associated with an increase in environmental pollution. As a result of these events, the world is facing the need to find alternative energy sources and the development of innovative and environmentally friendly technologies [10]. In addition, as a result of the ratification of the climate package by the EU Member States, Poland is committed to produce by 2020, 15% of energy from renewable sources, which forces the Polish authorities to take decisive legal measures to implement this objective. Poland is a country with great potential in biomass energy. In terms of agricultural land it occupies fifth place in the European Union [14]. Intensive agriculture generates large amounts of waste that can be used to produce biogas [7] or compost [4, 16]. Therefore, it is expected a substantial share of biomass of various origins in the production of renewable energy [7]. It is also assumed that the planned increase in the share of renewable energy in the total domestic production of energy will produce new plants using biogas [6, 21].

Nowadays the production and the use of hydrogen as renewable energy technology seems to be interesting. All over the world, researches on hydrogen energy are being developed as well as the possibility of its production, utilization and storage. There are no emissions into the environment when burning hydrogen or using fuel cells, instead, as a result of its conversion, steam is generated, which is presented by the equation 1.

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \quad (1)$$

An additional advantage resulting from the use of hydrogen in a fuel cell, it is also its high conversion efficiency of approx. 45-60% [18].

The aim of this study was to determine the theoretical potential of bio hydrogen and biogas production from the waste generated in a dairy plant. This solution would allow to manage large quantities of whey, which is a waste and a problem for dairies.

2. Hydrogen generation by dark fermentation

The well-known metabolic pathways and hydrogenase enzyme are used in dark fermentation [19]. In the first step, glycolysis, glucose is converted to pyruvate. In the next pyruvate is oxidized to acetyl-coenzyme A (CoA), while reducing ferrodoxin (Fd). The third step consists in the oxidation of ferredoxin through hydrogenase, resulting in molecular hydrogen is obtained [1, 19]. This process is illustrated by the following equations (2-4):

$$glucose \rightarrow pyruvate \quad (2)$$

$$pyruvate + CoA + 2Fd(ox) \rightarrow acetyl + 2Fd(red) + CO_2 \quad (3)$$

$$2\,Fd(red) \rightarrow 2\,Fd(ox) + H_2 \quad (4)$$

During the dark fermentation process the carbohydrates, proteins and fats are converted into volatile fatty acids [1]. Theoretically, one mole of glucose can generate up to 4 moles of hydrogen, 2 moles of carbon dioxide and 2 moles of ethyl [15]. This conversion is presented using the following equation 5.

$$C_6H_{12}O_6 + 2H_2O \rightarrow 4H + 2CO_2 + 2CH_3COOH \quad (5)$$

In fact, according to literature, the achieved amount of hydrogen is much lower, from 1 to 2,7 moles of hydrogen from 1 mole of glucose. This is due to the formation of butyric acid, which lowers the amount of produced hydrogen (equation 6) [12, 15, 19].

$$C_6H_{12}O_6 + 2H_2O \rightarrow 2H_2 + 2CO_2 + CH_3CH_2CH_2COOH \quad (6)$$

The reduction in efficiency of hydrogen production may be due to the process conditions and the used variety of microbial cultures [2, 3, 5, 8, 9, 12, 20].

3. Two-stage hydrogen-methane fermentation process

As a consequence of the increasing prices of the traditionally used substrates for the biogas production in the methane fermentation process and the problems associated with the financial support of renewable energy in Poland, a development of innovative systems enabling the production of high-energy fuels has been observed. In the case of the production of gas biofuels from organic waste, the process is carried out using a two-stage hydrogen-methane fermentation. The first stage of this technology consists in the production of hydrogen in dark fermentation. The resulting waste of the first stage is a substrate for methane fermentation, which is the second step of the process. The scheme technology combining the two-stage fermentation of hydrogen and methane is shown in Figure 1.

4. Methodology

4.1. Charasteristics of the whey industry

The study assumes that the system that produces and uses hydrogen will be located at an existing dairy plant. This solution will allow to use the post-production bio waste (whey) as a substrate from the abovementioned company. The location of the installation, near the plant, will reduce substrate transport costs and will avoid high environmental fees (approx. 160 000 PLN/year) related to the discharge of effluents because of its close location. The average daily production of the dairy plant bio waste is approx. 400 m^3/day.

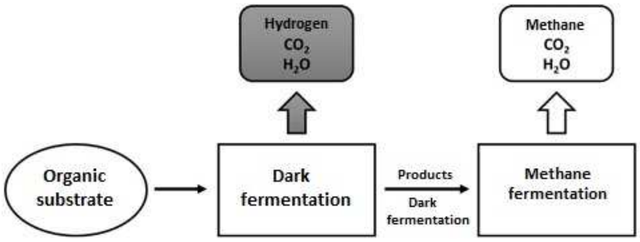

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Fig. 1. Diagram of a two-step fermentation technology, hydrogen-methane

Rys. 1. Schemat technologii dwustopniowej fermentacji wodorowo-metanowej

4.2. Methane and hydrogen production efficiency of the substrate

The study was conducted at the Laboratory of Ecotechnologies – the biggest biogas laboratory in Poland. Laboratory working within the Institute of Biosystems Engineering (Poznan University of Life Sciences). The research was based on modified German standard DIN 38 414/S8 and the standardized biogas guide from the Association of German Engineers in Dresden - VDI 4630.

The inoculum (liquid fraction of the digested pulp) was taken from an agricultural Polish biogas plant working on cow slurry and maize silage (Działyn). The inoculum was stored at room temperature to use it for mesophilic fermentation.

During the experiment the following standard methodology established by Polish Norms (PN) has been used: for dry matter PN-75 C-04616/01, pH - PN-90 C-04540/01, conductivity PN-EN 27888:1999 and organic dry matter PN-Z-15011-3. These parameters were also necessary for the calculation of the efficiency of biogas substrate in units m^3/Mg fresh matter, m^3/Mg d.m. and m^3/Mg o.d.m.

The research on biogas production was realized on 2 dm^3 reactors located in a layer of water at temperature 39°C ±1. Every one of the reactors is part of a 21-chamber fermentation reactor built in the Laboratory of Ecotechnologies (fig. 2). The concentration measurements of methane, carbon dioxide, hydrogen sulphide, ammonia and oxygen in the produced biogas were carried out with the use of the absorption sensors working in an infrared and electrochemical sensor line. The GA5000 gas analyser from Geotech firm was used in the research.

Hydrogen production efficiency from whey, and the average retention time (HRT) was based on literature data [11, 12]. In the paper the theoretical yield of hydrogen production from lactose is 2.7 mole/mole [12]. The calculations include the methane efficiency production from lactose (part of the substrate) in the hydrogen production.

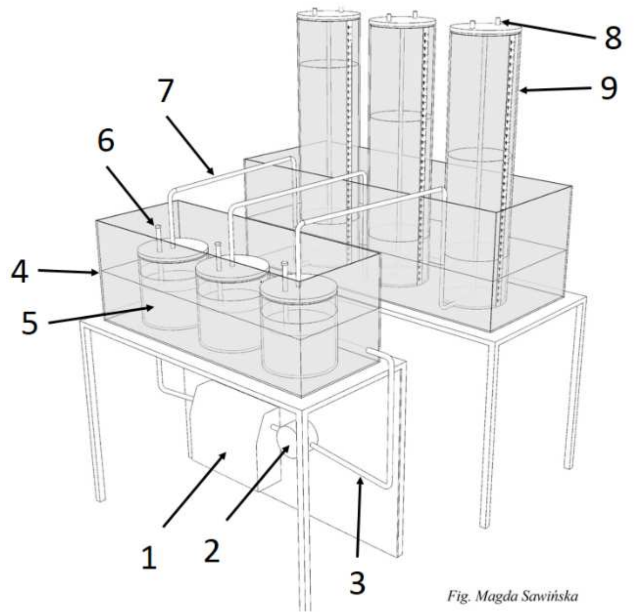

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Fig. 2. Scheme of biofermenter for biogas production research (3-chamber section): 1 – water heater with a temperature in the range of 20-70ºC, 2 – water pump, 3 – isolated hot liquid tube, 4 – layer of water at temperature of 38°C, 5 – biofermentor with the input of 1.4 dm^3 of capacity, 6 – sampling tube, 7 – tube for biogas flow, 8 – security valves (also used for taking biogas samples), 9 – biogas container made of poly (methyl methacrylate)

Rys. 2. Stanowisko fermentacyjne: 1 – ogrzewacz wody z regulatorem temperatury, 2 – pompa wodna, 3 – przewody cieczy ogrzewającej, 4 – płaszcz wodny, 5 – biofermentor o pojemności 2 dm^3, 6 – zawór do pobierania prób wsadu, 7 – przewód odprowadzający biogaz, 8 – zawór do pobierania prób gazu, 9 – wyskalowany zbiornik na biogaz

4.3. Methodology of economic calculations

To determine the mass and volume of available substrate to produce gases, the following formulas and figures were used [17]:

The mass of the substrate introduced into the reactor over a year:

$$M = M_d \cdot 365 \quad (7)$$

where: M – Mass of the substrate [Mg/year], M_d – mass of substrate delivered per day.

The volume of the generated hydrogen:

$$V_H = W_H \cdot M \quad (8)$$

where: $\mathbf{V_H}$ – volume of hydrogen produced [m^3], $\mathbf{W_H}$ – hydrogen efficiency of the substrate [$m^3 \cdot Mg^{-1}$], – mass of the substrate [Mg].

The volume of the generated methane:

$$V_M = W_M \cdot M \quad (9)$$

where: V_M – volume of produced methane [m^3], W_M – methane efficiency of the substrate [$m^3 \cdot Mg^{-1}$], M – mass of the substrate [Mg].

The amount of electricity generated in cogeneration:

$$E_E = V_M \cdot W_{E_{CH_4}} \cdot \eta_E \quad (10)$$

where: E_E – amount of electricity generated in cogeneration [MWh], V_M – volume of produced hydrogen [m³], $W_{E_{CH_4}}$ – energy efficiency ratio of methane - 0,00917 [$MWh \cdot m^{-3}$], η_E – electrical efficiency of the cogeneration plant [-], η_E = 0,36 – 0,44 [-], it was adopted in the calculation of the value of 0.4.

The amount of electricity generated in the fuel cell:

$$E_{FC} = V_H \cdot W_{E_{H_2}} \cdot \eta_{FC} \quad (11)$$

where: E_{FC} – amount of electricity generated in the fuel cell [MWh], V_H – volume of produced hydrogen [m³], $W_{E_{H_2}}$ – energy efficiency ratio of hydrogen – 0,0029559 [$MWh \cdot m^{-3}$], η_{FC} – electrical efficiency of the fuel cell [-], η_{FC} = 0,45 – 0,65 [-], it was adopted in the calculation of the value of 0.6.

The amount of heat generated in cogeneration:

$$E_{T_1} = V_M \cdot W_{E_{CH_4}} \cdot \eta_T \quad (12)$$

where: E_{T_1} – amount of heat generated in cogeneration [MWh], V_M – volume of produced hydrogen [m³], $W_{E_{CH_4}}$ – energy efficiency ratio of methane [$MWh \cdot m^{-3}$], η_T – thermal efficiency of the cogeneration plant [-], η_T = 0,43 – 0,54 [-], it was adopted in the calculation of the value of 0.45.

The amount of heat generated in cogeneration:

$$E_{T_2} = \frac{E_{T_1}}{0{,}274} \quad (14)$$

where: $\mathbf{E_{T_2}}$ – quantity of heat generated, expressed in [GJ], E_{T_1} – quantity of heat generated, expressed in [MWh].

Electric power of the cogeneration plant:

$$P_E = \frac{E_E}{t} \quad (15)$$

where: P_E – electric power of the cogeneration plant [MW], E_E – amount of produced electricity [MWh], t – cogeneration plant operating time [h]. The calculations assumed a value of 8200 hours of working time for a cogeneration plant within a year.

Heat power of the cogeneration plant:

$$P_t = \frac{E_{T_1}}{t} \quad (16)$$

where: P_t – heat power of the cogeneration plant [MW], E_{T_1} – amount of produced electricity [MWh], t – cogeneration plant operating time [h]. The calculations assumed a value of 8200 hours of working time for a cogeneration plant within a year.

Electric power fuel cell:

$$P_O = \frac{E_O}{t} \quad (17)$$

where: P_O – electric power fuel cell [MW], E_O – amount of electricity generated from produced hydrogen [MWh], t – fuel cell operating time [h].

For the calculation of incomes from sales of electricity and heat energy and RES energy production, the following formulas and figures were used [17]:

Income from sales of electricity and heat produced in co-generation:

$$C_E = E_E \cdot \text{Electric energy price} \quad (18)$$

$$C_{GC} = E_E \cdot \text{Green certificate price} \quad (19)$$

$$C_{YC} = E_E \cdot \text{Yellow certificate price} \quad (20)$$

$$C_T = E_{T_2} \cdot \text{Price GJ of heat} \quad (21)$$

$$Income_1 = C_E + C_{GC} + C_{YC} + C_T \quad (22)$$

where: C_E – annual income from electricity sale (cogeneration) [PLN], C_{GC} – annual income from green certificate (cogeneration) [PLN], C_{YC} – Annual income from yellow certificate (cogeneration) [PLN], C_T – Annual income from heat sale (cogeneration), $\mathbf{Income_1}$ – annual income from sales of electricity and heat produced in cogeneration [PLN].

Income from the sale of electricity generated in the fuel cell:

$$D_E = E_{FC} \cdot \text{Electric energy price} \quad (23)$$

$$D_E = E_{FC} \cdot \text{Green certificate price} \quad (24)$$

$$Income_2 = D_E + D_{GC} \quad (25)$$

where: D_E – annual income from electricity sale (fuel cell) [PLN], D_E – Annual income from green certificate (fuel cell) [PLN], $Income_2$ – annual income from sales of electricity and heat produced in the fuel cell [PLN].

Total income from sales of electricity and heat produced in the installation:

$$Income_{total} = Income_1 + Income_2 \quad (26)$$

where: $Income_{total}$ – Total income from sales of electricity and heat produced in the installation [PLN], $Income_1$ – annual income from sales of electricity and heat produced in cogeneration [PLN], $Income_2$ – annual income from the sale of electricity generated in the fuel cell [PLN].

Prices used in the calculation of certificates and sale of electricity and heat were based on the average of the prices of April 2015, according to the Power Exchange and the Energy Regulatory Office and they are summarized in Table 1.

Table 1. Certificate price and sale of electricity and heat (Own elaboration based on data from the Polish Power Exchange and URE – 18.09.2015)

Tab. 1. Ceny świadectw pochodzenia oraz sprzedaży energii elektrycznej i ciepła (Opracowanie własne na podstawie danych z Towarowej Giełdy Energii i URE – 18.09.2015)

	Price	Unit
Green certificate	114,69	PLN/MWh
Yellow certificate	117,51	PLN/MWh
Electric energy	156,49	PLN/MWh
GJ heat	25,00	PLN/GJ

Source: own work / Źródło: opracowanie własne

5. Results and discussion

5.1. Biogas and hydrogen efficiency

The results of the analysis of physical-chemical parameters of whey were collected in Table 2.

Table 2. Physical and chemical parameters for whey and mesophilic inoculum

Tab. 2. Parametry fizykochemiczne dla permeatu serwatkowego i zaszczepki mezofilowej

	pH	Conductivity [mS]	Dry matter [%]	Organic dry matter [% s.m.]
Innoculum	8,03	17,45	3,35	65,72
Whey	4,95	10,10	7,00	90,00

Source: own work / Źródło: opracowanie własne

The parameters were needed to determine the proportion of fermentation mixture, followed by the calculation of the theoretical hydrogen efficiency and the biogas efficiency. The researched substrate was characterized by a dry matter content of 7%.

The average fermentation time of whey was approx. 12 days. The largest peak production of biogas has been observed in the first three days of the research (Figure 1). This is due to intensive degradation of lactose, easily degradable disaccharide, by hydrolysis. A significant decrease in the daily gas production was observed on the fourth day. It was because the process of hydrolysis started to hydrolyze other degradable chemical compounds that were more difficult to degrade.

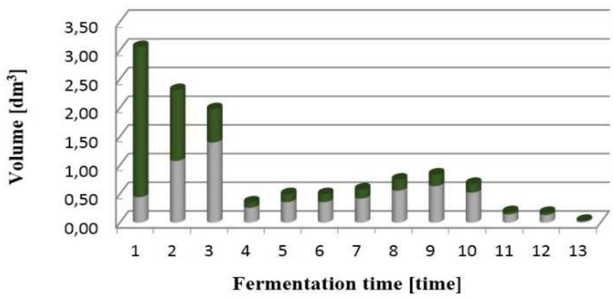

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Fig. 3. Daily biogas and methane production from whey

Rys. 3. Wykres dobowej produkcji biogazu i metanu dla permeatu serwatkowego

The mesophilic fermentation of whey, based on fresh weight of Mg substrate, obtained 49.92 m^3 of biogas it had an average methane content approx. 49% (Table 3).

The whey biogas potential that the research showed is 713.12 m^3/Mg d.m., which is approx. 125 m^3/Mg d.m. higher than the potential of maize silage, commonly used in agricultural biogas plants [13].

During fermenting whey in the methane and the hydrogen-methane fermentation process an appropriate ratio C / N ratio is required in the compounds into the fermentation reactor.

When fermenting whey in the methane and the hydrogen-methane fermentation process is required an appropriate ratio C / N ratio in the compounds into the fermentation reactor.

According to [12], from 1 mole of lactose in dark fermentation it is possible to obtain from 1.5 to 2.7 moles of hydrogen. Different H_2 production efficiency, according to literature data, may be the result of different process conditions and different variety of microbial cultures used. [2, 3, 5, 8, 9, 12, 20]. Based on these results, the theoretical hydrogen-methane efficiency of whey can be observed in Table 4.

Table 3. Biogas efficiency of whey in mesophilic fermentation
Tab. 3. Wydajność biogazowa permeatu serwatkowego w fermentacji mezofilowej

		Fresh matter		Dry matter		Organic dry matter	
Sample	Methane percentage [%]	Cumulated methane [m^3/Mg f. m.]	Cumulated biogas [m^3/Mg f. m.]	Cumulated methane [m^3/Mg d. m.]	Cumulated biogas [m^3/Mg d. m.]	Cumulated methane [m^3/Mg o.d.m.]	Cumulated biogas [m^3/Mg o.d.m.]
Whey	49,01	24,47	49,92	349,50	713,12	388,33	792,35

Source: own work / Źródło: opracowanie własne

Table 4. Theoretical hydrogen-methane efficiency of whey
Tab. 4. Teoretyczna wydajność wodorowa i metanowa dla permeatu serwatkowego

	Fresh matter		Dry matter		Organic dry matter	
Sample	Cumulated hydrogen [m^3/Mg f. m.]	Cumulated methane [m^3/Mg f. m.]	Cumulated hydrogen [m^3/Mg d. m.]	Cumulated methane [m^3/Mg d. m.]	Cumulated hydrogen [m^3/Mg o.d.m.]	Cumulated methane [m^3/Mg o.d.m.]
Whey	10,76	16,30	153,72	261,80	170,80	290,80

Source: own work / Źródło: opracowanie własne

In the results of the hydrogen-methane fermentation, the theoretical production of hydrogen and methane in terms of fresh matter in Mg is respectively 10,76 m^3 and 16.30 m^3. These assumptions include the reduction of methane production by using part of the intermediate metabolites, formed during the decomposition of whey, for the production of hydrogen.

5.2. Economic analysis

Annual production of whey in the selected dairy plant will be approx. 146 000 Mg. Table 5 shows the simplified results of the energy and economic calculations. It does not include installation costs, heating tanks, transport and the purchase of the substrate.

Table 5. Income from sales of electricity and heat produced in the installation
Tab. 5. Przychód ze sprzedaży energii elektrycznej i ciepła wyprodukowanych w instalacji

	Cogeneration unit	Fuel cell
Electric and heat energy		
Electric energy [MWh]	8 536,32	2 790,02
Heat [MWh]	9 603,36	-
Heat [GJ]	35 048,74	-
Power of the installation		
Electric power [MWe]	0,99	0,32
Heat power[MWt]	1,12	
Income		
Electric energy [PLN]	1 335 848,14	436 611,01
Green certificate[PLN]	979 030,12	-
Yellow certificate [PLN]	1 003 102,53	-
Heat [PLN]	876 218,60	-
TOTAL [PLN]	4 630 810,40	

Source: own work / Źródło: opracowanie własne

The annual energy potential of whey produced in the selected dairy plant and used in an innovative installation for the production of hydrogen and biogas is approx. 11 326 MWh of electricity and approx. 9 603 MWh of heat. The estimated amount of electricity generated in the fuel cell is approx. 24,5% of the total energy installation.

The nominal power cogeneration plant installed in the biogas plant is 0,99 MW of electricity and 1,12 MW of heat, while the fuel cell power is 0.32 MW of electricity. It is also possible to locate three or four units in order to avoid suspending production and economic losses caused by engine failure or by the need to replace oil. Total annual income from sales of electricity and heat produced in the biogas plant is 4 630 810 PLN.

6. Conclusions

1. Whey permeate is problematic waste from the point of view of dairies. The high disposing cost of the high BOD and COD load waste water, often exceeds the incomes of the company, it leads to a necessary reduction of the product production in these companies. It is, therefore, necessary to modify the current practice of production and use of the whey as a valuable feedstock for energy production.
2. The construction of an installation for using whey as a source of biomass in the methane-hydrogen fermentation process, will allow to reduce the cost of environmental charges and also to generate additional income.
3. The proposed installation of 0,99 MW of electric energy, 1,12 MW of heat and 0,32 MW of electric energy originated in the fuel cell would be located at the dairy plant producing annually approx. 146 000 Mg of the mentioned waste. Annual incomes for dairy plant would amount more than 4,5 million PLN.

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