Continuously flow microwave pre-treatment for enhanced anaerobic biodegradability of dairy industry sludge

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***Abstract*— *Our research has focused on the investigation of the applicability of continuously flow microwave pre-treatment process for anaerobic digestion of dairy industry sludge. In the continuously flow operation effects of microwave specific process parameters, i.e. specific irradiated energy and microwave power level, were examined on the biogas yield and anaerobic digestion rate. Furthermore, the efficiency of process was characterized based on energetic analysis, as well. Our results show, that depend on the irradiated energy level the microwave power has effect on biogas yield, and determine the biodegradation rate, as well. Higher energy irradiation or higher power has been manifested in enhanced anaerobic digestion, but over a certain value of them led to worsened methane content. Considering the results obtained from energetic analysis can be concluded, that microwave pre-treatment followed by anaerobic digestion is efficient if irradiated energy was kept lower level (100 kJ L^{-1}, in present experiments) with microwave power levels of 536 W, or 700W, respectively. Too high energy intensity at high power level, or too low microwave power, independently from the extent of irradiated energy, led to higher energy demand of pre-treatment, than can be recovered as energy content of surplus biogas.*

Keywords*— *anaerobic digestion, biogas, energy efficiency, microwave, sludge.

I. INTRODUCTION

Wastewater and sludge pre-treatments can be divided three major groups: chemical processes, thermal processes and biological processes. Chemical treatments are efficient, but many times cause corrosion problem in wastewater line, in equipment and tanks, or generate undesirable side-products. Biological processes can be considered more environmentally friendly than chemical treatments, but need longer time and considered more sensitive for the change and fluctuation of process parameters (temperature, pH, ionic strength, presence of inhibiting or toxic components, homogeneity of the material matrix etc.). Heat treatments has numerous advantages over the other processes, such as no formation of undesirable by-product, more flexibility to unsteady capacity, good controllability, high number of pre-designed and modular industrial scale equipment etc. Compared to the conventional heating microwave irradiation has higher heating rate, heating equipment operated by microwaves requires less space, and using well designed system and intermittent irradiation mode the energetic efficiency is higher than conventional method (Cuccurullo et al., 2013).

In high frequency oscillating electromagnetic field, because of the fast change of polarity, the dipolar components of irradiated medium vibrated and oscillated, and, furthermore movement of ionic compounds or charged particles has also been occurred. These effects led to intramolecular friction resulted in internal heat generation. The electromagnetic energy interact directly with the components of materials, the medium absorb the energy and convert it into heat (Mawioo et al., 2016). Considering the specific heat generation mechanisms of microwave irradiation, in real and complex medium, such as the wastewater, composition, dielectric properties of compounds, intermolecular interaction, chemical structure of molecules, physicochemical state, temperature, viscosity and density of continuous phase and their possible alteration during irradiation, furthermore the frequency of electromagnetic field has also effect on the thermal efficiency of heating (Holtze et al., 2006). Another interesting phenomenon of microwave irradiation is the selective heating. If components of a system have different dielectric behavior (dielectric constant, and/or dielectric loss) during irradiation the heating rate of solely components is different. Therefor the temperature difference, occurred by different energy absorption, can led to thermal stress inside of the material structure, or inside of microbial cell, or tissues.

In wastewater and sludge organic matters are partially present in particular form. In many cases the conventionally used wastewater purification processes aim the coagulation and flocculation of soluble organic and inorganic pollutants, transform them insoluble, particulate form. During phase separation processes, these components can be concentrated into sludge. But if the further utilization of wastewater or sludge is planned, for example in anaerobic digestion process, increasing of organic

matter solubility is expediential for enhanced organic matter removal efficiency, biogas production rate and higher biogas yield.

Among sludge management options anaerobic digestion (AD) has been more and more popular at wastewater treatment plant. The main advantage of AD is that sludge stabilization and energy generation can be carried out simultaneously. The output of AD cannot be considered obviously 'final waste', because digestate has a good potential for agricultural utilization, as fertilizer. One of the main problem for application of AD in-line at wastewater treatment plant (WWTP) is the long retention time. On the one hand the slow degradation increase the capital and running costs, and on the other hand present bottleneck for the overall capacity of WWTPs. Therefore, beside the enhanced dewaterability and microbial stabilization of sludge, pre-treatments serve as intensification process for AD. Results from the comparison of conventional heating and microwave irradiation for wastewater and sludge pre-treatment are contradictionally. Detach of non-thermal effects of microwave from thermal effects is difficult, because mainly of high temperature ramp and hot-spot heating effects occurred during microwave irradiation (Sólyom et al., 2011). It can be noticed, that in practice the same condition for conventional- and microwave heating in real material matrix cannot be ensured.

Beyond above mentioned difficulties microwave irradiation has verified positive effect on organic matters of wastewater and sludge, which can be utilizable as pre-treatment followed by AD. Microwave has strong effect on microbial destruction, microwave process need significantly shorter time demand than needed for conventional heat treatments. Depending on the heating rate and final temperature during the processes, with the application of microwave pre-treatments higher disintegration degree can be achieved. Therefore the higher disintegration degree and higher organic matter solubility led to higher biogas yield in AD process. Beside absolute value of biogas production, the biogas production rate presents key issue to evaluate the efficiency of a pre-treatment method. Depending on the material characteristics and heating rate and final temperature, microwave pre-treatments are suitable to increase the degradation rate, therefore accelerate the biogas production (Yang et al., 2013).

Efficiency of microwave pre-treatments depends on the type and condition of anaerobic digestion. Thermophilic digestion suitable to achieve higher biogas product, the effect of pre-treatments are slightly than that of obtained for mesophilic temperature ranged AD tests. It was found that there was no significant difference between the effects of conventional and microwave heating method on biogas yield, but temperature ramp considered as the main influential process parameter in high total solid contented sludge cake processing followed by thermophile anaerobic digestion. On the other hand, beside the temperature ramp has significant effect on biodegradation rate, as well (Koupaie and Eskicioglu, 2016). But in Europe mesophilic AD process can be considered the commonly usable and economic method, therefore the pre-treatment efficiency need to be investigated at this temperature range.

Summarized the experiences of laboratory scale batch microwave wastewater and sludge pre-treatment can be concluded that preliminary results can be considered promising. But extend the investigation for implementation of continuously flow microwave pre-treatments is need to evaluate the applicability of method, find the main influential microwave related process parameters, model and optimization of the process to make suitable the scale up (Boldor et al., 2008). Another key issue for investigation of applicability of sludge and wastewater treatment operated by microwave heating is the temperature range. Considering the disintegration degree as control parameters for pre-treatment solely, pre-treatment over 100 °C cause significantly higher increments in organic matters solubilisations. In multicomponent medium, such as food industry wastewater and sludge, application of elevated pre-treatment temperature led to Maillard reactions, in which refractory compounds are formed from amino acids and reducing sugars, and other polymerization reactions are occurred in the presence of low molecular weighted, easily degradable intermediates (Shahriari et al., 2010). Therefore, change of biogas production due to microwave pre-treatment cannot be intrinsically estimated based on solubilisation ratio alone (Koupaie and Eckicioglu, 2015). Because of evaporation in open vessel system, and polymerization reactions, over a certain value of final temperature and/or temperature ramp during microwaving process the biogas potential of some part of organic matters decrease.

II. MATERIAL AND METHOD

Sludge processed in our experiments has been originated from dairy industry process. Thickened dairy sludge has a TS content of 3.8 ± 0.2 w%, and chemical oxygen demand of 48590 ± 352 mgL^{-1}, respectively. The fresh collected sludge was stored refrigerated at 6 °C before using in closed PP container.

Anaerobic biodegradability of sludge samples was assessed under mesophilic conditions at 37 ± 0.5°C for 30 days. Anaerobic digestion tests were carried out triplicated in 250 mL sealed bottles in a continuously stirred batch laboratory system. For seeding an industrial anaerobic digester sludge of the local wastewater treatment plant was used in 10 w/w% concentrations on dry matter basis. Acclimatization of inoculum was carried out the same temperature as applied for AD tests using pre-treated sludge as substrate.

Blank test was run to determine the own endogenous biogas production of seed sludge, and the blank value was subtracted from the results obtained from real sludge digestion. The initial pH of sludge mixed with inoculum was adjusted to 7.4. To ensure the anaerobic condition for AD tests bottles were flushed by N_2 gas using the double septum of reactor bottles.

Biogas production was measured by the pressure change in the headspace of reactor bottles. Pressure change was detected daily by OxiTop® Control (WTW, Germany) manometric measuring heads attached to the anaerobic reactors. Volume of biogas was calculated by the ideal gas law. Specific anaerobic digestion activity ($mLg_{TS}^{-1}day^{-1}$) was calculated from the first 10 days linear phase of the cumulative biogas production curve. Methane content of produced biogas was measured by a portable biogas analyser (airTOX, Fresenius Umwelttechnik, Germany) after the 30 days digestion. Total solid content (TS) was determined by gravimetric analysis.

Microwave pre-treatment was carried out in a custom made continuously flow equipment. Magnetron equipped in microwave reactor operated at a frequency of 2450 MHz, the power was continuously changeable in the range of 200-700W by adjustment of heating voltage. The volumetric flow rate of sludge through the toroidal type microwave cavity resonator was varied by the revolution of peristaltic pump. Coil tube implemented into the microwave cavity resonator has an inner diameter of 16 mm, and, because of minimal energy loss made from polytetrafluoroethylene (PTFE). To reduce the air and steam bubble forming processed sludge enters up flow from the bottom of resonator. The system has operated in open mode. Tuning screws in waveguide were adjusted to minimal power reflection to achieve high energy efficiency and to protect magnetron from overheating.

Specific microwave irradiated energy (E_s, $kJ\ L^{-1}$) was calculated from the volumetric flow rate of sludge (Q, Lh^{-1}) and the power of magnetron (P_m, W) by Eq. (1).

$$E_s = \frac{P_m}{Q}\ (kJ\ L^{-1}) \tag{1}$$

Considering the calculation method of E_s the same specific microwave irradiated energy can be achieved by the different combination of microwave power and volumetric flow rate. Specific net energy production (NE_p, $kJ\ L^{-1}$) of pre-treatment was calculated based on the volume of produced biogas (V_{bg}, L), methane content of biogas (v%), energy content of methane (H=39 $kJ\ L^{-1}$), microwave power (P_m, W), residence time of sludge in cavity (t, s) and the volume of irradiated sludge in resonator (V_{sl}, L) using Eq. (2) .

$$NE_p = \frac{\left(V_{bg}\ \frac{v\%}{100}\ H\right) - \frac{P_m\ t}{1000}}{V_{sl}}\quad (kJ\ L^{-1}) \tag{2}$$

III. RESULTS AND DISCUSSION

Results of mesophilic AD tests show, that biogas yield from dairy sludge can be improved by the continuously flow microwave pre-treatments. Compared to the untreated (control) sample, which has a biogas yield of 128 ± 6.9 mL g_{TS}^{-1} , microwave pre-treatments could enhance the specific volume of biogas over 400 mL g_{TS}^{-1} (Fig 1).

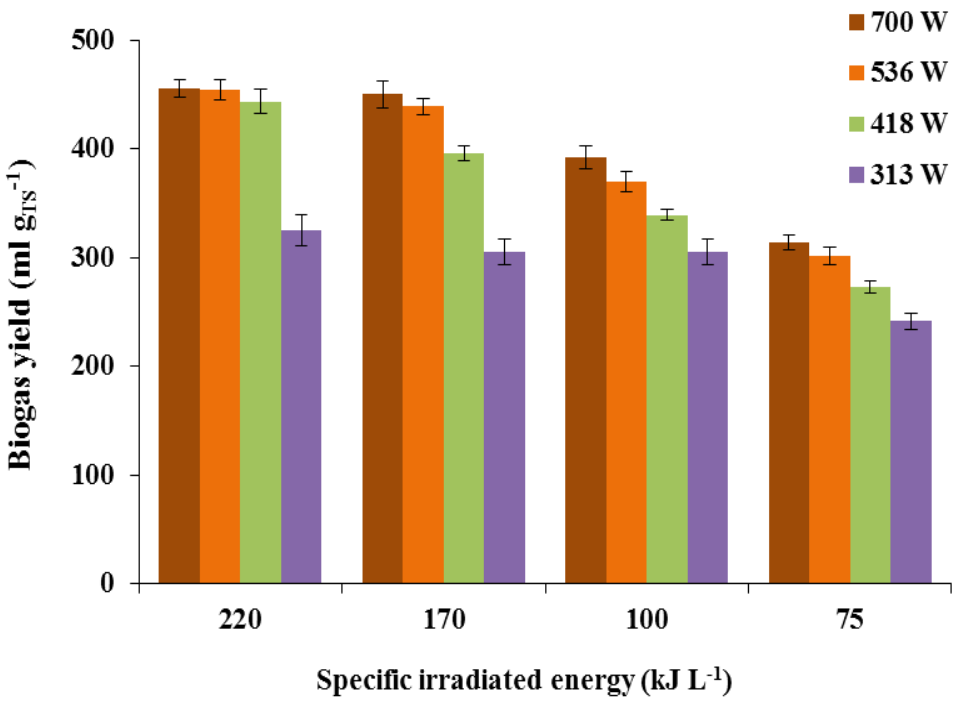

FIG 1— BIOGAS YIELD OF MICROWAVE PRE-TREATED SLUDGE

Our earlier results related to batch pre-treatment (Beszedes et al., 2011) and others studies (Yang et al., 2013) suggested that microwave power level can affect the anaerobic biodegradability of sludge. Investigation of the effects continuously flow microwave pre-treatment process on biogas yield achievable in 30 days digestion verified, that beside the irradiated energy, the microwave power level can be considered as influential process parameter, as well. Applying the highest specific irradiated energy effects of microwave power in the range of 418-700W does not led to significant difference in biogas yield, but by lower energy pre-treatments (170-100 kJ L^{-1}) the higher power resulted in higher biogas production. Maximum biogas yield was achievable by microwaving at power of 536 W and 700 W, and irradiating of sludge with 170 and 220 kJ L^{-1} energy.

From practical purposes, and from the aspects of operating parameters and stability of an industrial scale anaerobic digestion plant, beyond the biogas yield, the rate of anaerobic digestion can provide valuable information to evaluate and appreciate of the efficiency of pre-treatment processes. To quantify the effects of microwave pre-treatments with different energy intensity and microwave power the anaerobic digestion activity was calculated, defined the control parameter as the biogas production rate of linear phase of the cumulative biogas production curve. Specific anaerobic digestion activity is well correlate with the rate of decomposition of organic matters during the fermentation process.

Notwithstanding the high organic matter content, because of the sludge structure, high extracellular polymeric substances (Serrano et al., 2016) and presence of heavily degradable components from disinfection and cleaning process, dairy originated sludge has lower than theoretical biogas production and can be characterized by slow biodegradation rate. In our experiences, the average daily mesophilic anaerobic digestion activity determined for untreated dairy sludge was 8.7 ± 2.1 mLg_{TS}^{-1}. Results of calculation verified, that in addition to biogas yield, the rate of anaerobic digestion can be enhanced by the continuously flow microwave pre-treatment. Similar to the results regard to biogas yield, depend on the specific irradiated energy, digestion activity can be influenced by both the microwave power, and irradiated energy. Using the energy irradiation level of 220 and 75 kJ L^{-1}, varying of microwave power in the range of 418-700W has not significant effect, but microwave power determined significantly the digestion activity if 170 kJ L^{-1} energy was irradiated during pre-treatment (Fig 2). It can be summarized, that increasing of microwave power at the same energy irradiation level can accelerate the anaerobic digestion process, but the actual scale, size and significance of effects was determined by the energy level, as well.

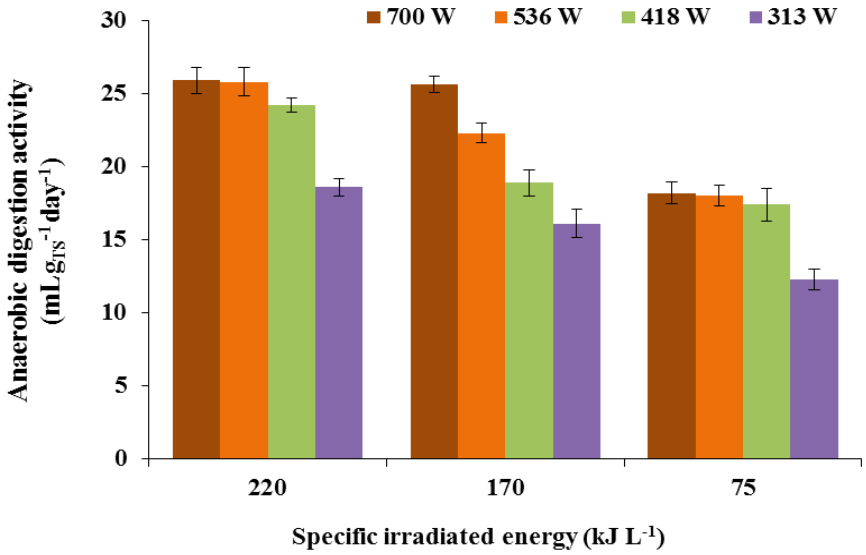

FIG 2— ANAEROBIC DIGESTION ACTIVITY FOR THE FIRST 10 DAYS PERIOD

Although the anaerobic digestion rate has strong effect on the stability of capacity in continuous digesters, because it has influenced by the organic matter removal efficiency, hydraulic retention time and applicable organic matter loading rate; the calorific value of produced biogas is also important parameter for the overall economy of AD technology. For this purpose, the final methane content of produced biogas was also measured at the end of 30 days anaerobic fermentation process.

Raw dairy industry sludge has low methane content (41%) , but as a results of microwave pre-treatment the calorific value of biogas was improved. Beside that microwave irradiation is generally suitable to increase the methane content in produced biogas, it can be noticed, that despite of higher biogas yield and anaerobic digestion activity, pre-treatment with high energy intensity and higher microwave power led to decreasing tendency in the change of methane content. Irradiating energy of 220 and 170 kJ L^{-1} , and increasing of microwave power 700 W from 536 W methane content of biogas decreased (Fig 3). In other energy levels increasing of microwave power led to improvement of methane ratio. Higher biogas yield with lower methane content revealed that microwave treatment was supposed to be suitable to assist in hydrolysis of macromolecules, possibly enhance the efficiency of acidogenesis, but product and/or byproduct of earlier stages of AD process can not be utilizable for microbes of methanogenesis stage (Shahriari et al., 2011).

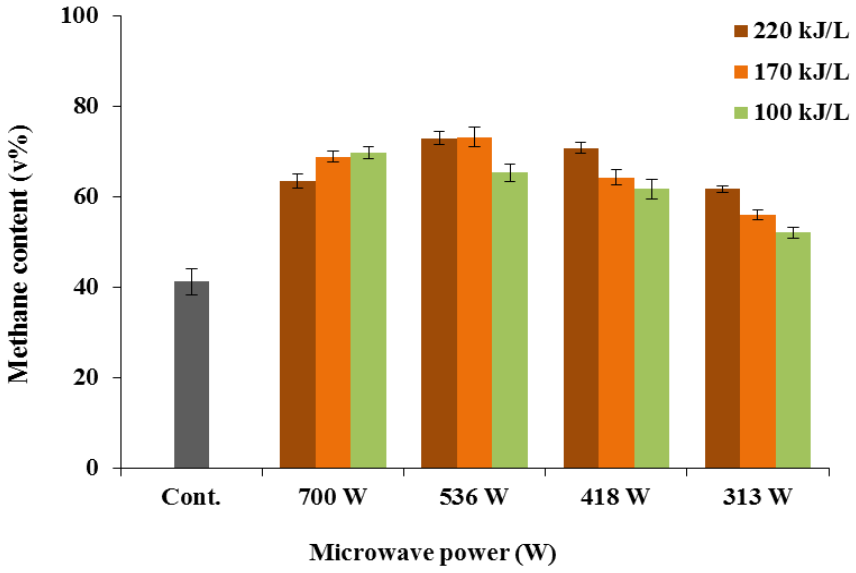

FIG 3— METHANE/BIOGAS RATIO AS FUNCTION OF MICROWAVE POWER AND IRRADIATED ENERGY

Opposite tendencies observed for the change in methane content and biogas yield at higher intensity and higher microwave power necessitates the investigation of energetic efficiency of pre-treatments. One of the simplest methods to quantify the energy efficiency of microwave treatments followed by anaerobic digestion is to calculate the net energy production as the difference between the energy content of the produced surplus biogas and energy demand of microwave process. Results of our calculation indicated that energy content of low power (313 W) microwave pre-treatments, independently from the applied irradiated energy, have not resulted in enough biogas production to compensate the energy demand of microwave process. On the other hand, despite to higher biogas production of sludge processed by high energy intensity and high power microwave irradiation, the energy demand of pre-treatment exceeded the energy content of produced biogas, therefore the process was disadvantageous from energetically aspects (Fig 4).

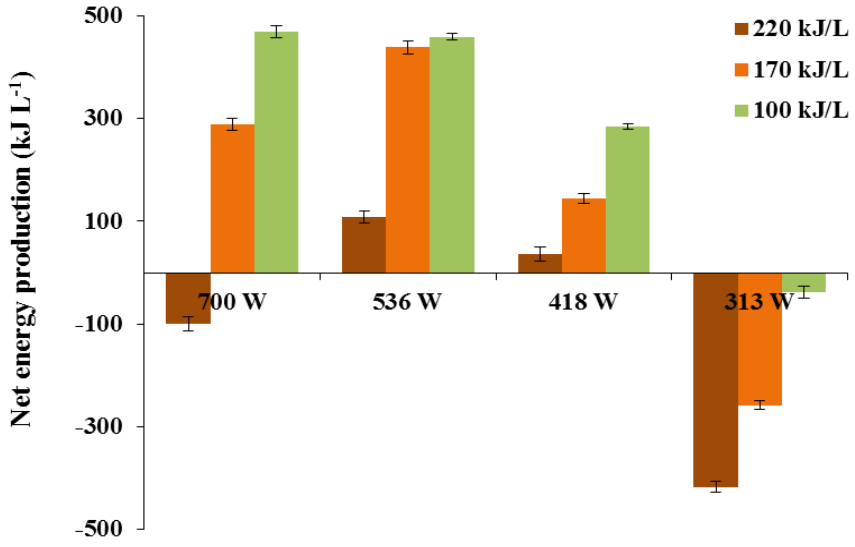

FIG 4— SPECIFIC NET ENERGY PRODUCTION OF PRE-TREATED SLUDGE

Based on the net specific energy production values, from energetically aspects, pre-treatments carried out with 100 kJ L^{-1} energy irradiation at 536 and 700W microwave power, and 170 kJ L^{-1} at 536 W power can be considered the more advantageous for the continuously flow microwave pre-treatment of dairy industry sludge. Further investigation is needed to determine the thermal efficiency of microwave heating to analyze the electrical energy transformation parameters and get more complete information of the overall energy efficiency of microwave sludge pre-treatment process.

IV. CONCLUSION

The main objective of this research effort was to investigate the effect and efficiency of continuously flow microwave irradiation method, as pre-treatment for anaerobic digestion of dairy industry sludge. Our results show, that biogas yield from sludge, and as well as the anaerobic digestion activity of processed biomass, can be improved by microwave pre-treatments. Depending on the extent of energy irradiation into the continuously flow sludge, the microwave power level can have significant effect on the efficiency and rate of anaerobic digestion process. Methane content of biogas was improvable by microwave irradiation, but over a certain value, the high energy intensity and high power microwaving can deteriorate the calorific value of produced biogas.

Results of energy retrieval of microwave pre-treatments during mesophilic anaerobic digestion show, that the energy efficiency of the continuously flow mode process was influenced by the irradiated energy and microwave power, as well. From energetic aspects, the lower energy pre-treatments (100-170 kJ L^{-1}) carried out at higher microwave power level (536-700W) can be considered as favorable. Further investigations are needed to analyze the dielectric behavior of materials during microwave irradiation, and to measure the real power dissipation as a function of flow properties, to get more complete information about the overall energy efficiency and to optimize the continuously flow microwave pre-treatment process.

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