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## Preliminary study of acrylamide monomer decomposition during methane fermentation of dairy waste sludge

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### ABSTRACT

Polyacrylamide (PAM) used in sludge dewatering exists widely in high-solid anaerobic digestion. Acrylamide is registered in the list of chemicals demonstrating toxic, carcinogenic and mutagenic properties. Therefore, it is reasonable to ask about the mobility of such residual substances in the environment. The study was carried out to assess the impact of the mesophilic ( $39 \pm 1^\circ\text{C}$ ) and thermophilic ( $54 \pm 1^\circ\text{C}$ ) fermentation process on the level of acrylamide monomer (AMD) content in the dairy sludge. The material was analysed using high-performance liquid chromatography (HPLC) for quantification of AMD. The results indicate that the process of methane fermentation continues regardless of the temperature effects on the degradation of AMD in dairy sludge. The degree of reduction of acrylamide monomer for thermophilic fermentation is 100%, while for mesophilic fermentation it is 91%. In practice, this means that biogas technology eliminates the risk of AMD migration to plant tissue. Moreover, it should be stressed that 90% of cumulative biogas and methane production was reached one week earlier under thermophilic conditions — the dynamics of the methanisation process were over 20% faster.

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### Introduction

Polyacrylamide (PAM) based flocculants have been accepted and implemented for decades in many applications including pulp, paper, cosmetic and textile industries. Acrylamide flocculants are also used in water treatment, chemical treatment of sludge and in the case of food processing it is recommended for organic fertilisation. The food industry typically generates solids with beneficial fertiliser properties, suitable after appropriate treatment, both to improve the properties of the soils used for agricultural purposes and for effective soil reclamation. A high content of organic matter and a significant amount of beneficial macro- and micro-nutrients have a positive influence on the

physicochemical properties of the soil. In many European countries, national authorities have implemented policies to support the use of sewage from the food industry in agriculture, as it is often considered the best economic and environmental solution (European Commission, 2001). Currently, due to their importance accompanied by many technological benefits, PAM polymers associated with trivalent metal ions, such as iron and aluminium in the form of commercially available coagulants are among the chemicals used in wastewater and sludge management, including the food industry in a large volume (Vanerkar et al., 2013).

According to European Union regulations (EEC, 1774/2002), sludge like other organic residues must be adequately treated

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and proven hygienically safe (Arthurson, 2008). More specifically, the purpose of sludge stabilisation is to reduce pathogens, eliminate offensive odours and inhibit, reduce or eliminate the potential for putrefaction, which is done by biological reduction of the volatile organic fraction or the addition of chemicals. Among various biological, chemical or thermal methods of sewage sludge treatment taken into consideration, before further utilisation in the agro-food sector, anaerobic digestion by converting a part of its organic matter into biogas is currently recommended as the most effective biomass valorisation technology for smaller reactor volumes, lower energy requirements for heating and lower levels of material handling (Duan et al., 2012; Fernandez Rodriguez et al., 2012). Anaerobic digestion of sewage sludge or co-digestion of sludge with the use of several food wastes has been studied recently by several authors (Lee and Shoda, 2008; Wan et al., 2011; Dai et al., 2013; Uma Rani et al., 2013; Di Maria et al., 2014; Buranasilp and Charoenpanich, 2011).

With the identification of environmental and health risks of acrylamide monomer (AMD) used in the production of a major class of water soluble PAM based polymers, concerns related to these topics have been increasingly raised. It is known that AMD adversely affects the peripheral nervous system of organisms as a highly toxic and potent carcinogenic compound (Tareke et al., 2002). Therefore, guidelines for PAM use in variety of applications typically recommend products with <0.05% AMD content only (Sojka and Surapaneni, 2001; Lu et al., 2014).

The twenty year plus period until now indicates that several problems related to the fate of AMD, following application of PAM in cropland, were studied and have been summarised in various articles including the most important topics like PAM degradation mechanism, AMD chemical reactivity and its mobility and accumulation risk in plants (Friedman, 2003). Among others our own study with hydroponically grown head lettuce confirmed that the problem of AMD mobility and its uptake by plants should be an important topic with respect to safe polyacrylamide handling in the agro-food area (Mroczek et al., 2014).

Since degradation of polyacrylamide accompanied with accumulation of its toxic monomer is important to the disposition of biogas residues generically termed as “digestate” (Chu et al., 2003; Zhang et al., 2009; Duan et al., 2012), biodegradation of polyacrylamide by anaerobic digestion under mesophilic conditions in a dewatered sludge system was studied by Dai et al. (2014). The authors concluded that PAM can be consumed as the carbon source by the breakdown of the carbon chain backbone in different groups. These results seem contradictory to those reported by other researchers, which indicated that PAM could be utilised as a nitrogen source rather via microbial amidase activity and transformation in polyacrylates (Kay-Shoemaker et al., 1998). It has been reported that aerobic and anaerobic treatment seems to be a perfect way for biodegradation of many substances (Costa et al., 2014; Lina et al., 2013; Brovelli et al., 2011). On the other hand, Hamelin et al. (2010) reported that the solid fraction from PAM-separated slurry is not suitable to be degraded during anaerobic digestion.

In the present study we focused on the examination of how the selected PAM flocculants, used in commercial thickening operation, affected AMD content during the digestion of sewage sludge collected from a dairy processing plant. Experiments were

conducted under the same analytical conditions, in lab bioreactors, to determine and to compare changes in free residual acrylamide monomer content in digestates following methanogenic fermentation of the raw material under mesophilic and thermophilic conditions. It should be underlined that biogas production based on biomass seems to be one of the most promising directions of renewable energy development in Poland (Cerbin et al., 2012).

## 1. Materials and methods

### 1.1. Materials

The material tested was PAM treated sludge collected from a local dairy processing plant generating typical milk waste and cleaning wastewaters from the facilities of the factory. The sludge samples were obtained from dairy effluent treatment plant characterized by flow about 600 m<sup>3</sup>/day and Population Equivalent about 23,500. The characteristics of the influent wastewater of the plant making drinking milk, yoghurt, kefir and quark and working without whey protein recovery system were as follows: pH was 5.5, total COD was 5000 mg/L, total nitrogen was 30 mg/L and total phosphorus was 7.1 mg/L. Chemicals used during wastewater and sludge treatment included alum, iron chloride, iron sulphate and selected PAM flocculants. Following technological procedure, the highly charged, very high molecular weight cationic product in the form of emulsion with residual acrylamide monomer content below 1000 mg/L was employed for sludge thickening in dosage of 3.5 kg/Mg dry matter (DM). As inoculum — the liquid fraction of digested pulp from a local agricultural biogas plant was used, which is common practice in laboratory scale experiments (Djelal and Amraneb, 2013; Chen et al., 2015).

The study was conducted in a biogas Laboratory of Ecotechnologies working within the Institute of Biosystems Engineering at Poznan University of Life Sciences. The research was based on modified German standard DIN 38414/S8, while chemical and physical analytical methods were based on Polish Standard system (Dach et al., 2014; Boniecki et al., 2013). Analytical procedures were developed within several scientific projects financed by the EU 6th Framework Program and national funds realised in the years 2006–2012.

### 1.2. Digestion under meso- and thermo-philic conditions

#### 1.2.1. Methane production set-up

The experiment of biogas production was conducted through anaerobic digestion in the set of multichamber biofermentors (Fig. 1). This biofermentor is commonly used for testing biogas efficiency for large amount of biomass samples.

The batch experiment with sewage sludge methane fermentation was carried out according to the German norms: DIN 38414/S8 and VDI 4630. Those methodologies are commonly used in German biogas laboratories as well as in other countries in central Europe. At the beginning of the process 300 g of sludge were placed in reactors and 700 g of inoculum (mesophilic or thermophilic) were added to initialize fermentation process. Aerobic digestion experiments of analysed sewage sludge samples were carried out in glass reactors constructed in the

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