1 Introduction

Greenhouse gas emissions of municipal solid wastes (MSW) are in the focus of legal authorities and numerous environmental scientists. Especially, industrial nations are in the middle of forcefully reducing their annual amount of emissions. In most of these nations, landfill guidelines provide standards to comply to. In Germany, landfill guideline transition was defined in 1993 which state that landfilled waste has to comply with new standards from June 2005 onwards [1]. An important impact factor for tightening the regulations are greenhouse gas emissions of landfills which contribute to about 13 % of the global anthropogenic methane emissions [43]. Monitoring compliance with these new standards revealed that treated waste scarcely meets the required stability values. For waste pre-treatment, mechanical-biological waste treatments (MBT) as well as thermal waste treatment (incineration) are typically applied in equal measure. In the MBT process, the high-calorific fraction is removed by sieving and then sent to incineration. The remaining fraction is further mechanically and biologically treated and leachate contamination and gas emissions are significantly reduced. The mechanical stability was examined and can be granted if an adapted way of construction is applied [39]. The biological stability is in most cases considered insignificant when gas production values of MBT residual waste meets the requirements of the German waste storage ordinance [1]. However, MBT residual waste cannot be considered as biologically inert material; it still holds a gas emission potential when residues are landfilled and exposed to environmental conditions [60].

Within the past years of the collaborative research project SFB 477 *Life Cycle Assessment of Structures via Innovative Monitoring*, experimental and theoretical investigations were carried out to gain knowledge about emission behaviour of landfilled MSW and to create modelling networks to predict decomposition and landfill gas production by using different modelling approaches. Within the project B5 of the SFB 477, the chemical and biological reaction processes in municipal landfills are analysed to facilitate the development of new models for long-term landfill behaviour. Knowledge about these reaction processes allows the prediction of landfill gas emission as well as the evaluation of adequate monitoring concepts. Conventional landfills are operated to minimise the amount of moisture infiltrating into the waste to inhibit landfill gas emission. Hence, this is considered as a dry cell concept. If water infiltration cannot be foreclosed, e.g. due to cracks in the top covers, biodegradation will be initiated even after decades of inactivation. An alternative concept, combining emission controlled landfill operation and waste treatment, are landfill bioreactors. They are designed and operated to enhance biodegradation processes by an increased

moisture content due to water addition and/or recirculation of leachate [59]. In this sense, whatever concept of waste treatment is used, either MBT or landfill bioreactors, moisture content is regarded as one of the most crucial factors in landfill operation with regard to biodegradation enhancement.

1.1 MBT waste

The process of the MBT plant of the Hanover waste treatment centre is shown schematically in Figure 1-1.



Figure 1-1: Schematic process in MBT plant in Hanover [16].

Input material is municipal solid waste collected within the area around Hanover. Contraries are removed before shredding and metal removing. A screen drum sieves out coarse particles with a high calorific value for incineration. The through fraction containing most of the organic material suitable for biological treatment enters a ballistic separator. Here, the through fraction is separated into a coarse fraction for incineration and a fine fraction. The fine fraction enters the biological treatment plant and is liquefied with steam for pumping. By adding recycled fermentation residue, the organic material in the waste is efficiently converted into biogas in fermentation tanks. After re-oxygenation in air tunnels, the residual waste is piled in composting windrows. The MBT residual waste is then landfilled.

1.2 Motivation and Objective

As a novel waste, MBT residual waste is used for investigations on emission behaviour at different environmental conditions. The focus of this work is on the evaluation of biochemical parameters which are known to be crucial for degradation of organic matter under anaerobic conditions [47]. Since the organic matter content in MBT residual waste is markedly reduced by the intensive biological treatment, the composition of metabolic intermediates and products during anaerobic degradation is expected to be very different compared to conventional MSW. For assessing the remaining biodegradation potential, it needs to be investigated if the acid buffering intensity in MBT residual waste is sufficient to stabilise the pH for methanogenic activity. This issue is addressed by different experimental investigations to uniformly quantify acid buffering intensities and to reveal the influence of different adjusted pH conditions on anaerobic metabolic processes in MBT residual waste. Also, metabolic intermediates and products need to be investigated with regard to their microbiological availability. The waste matrix is expected to highly influence the sorption behaviour of products and substrates of anaerobic degradation. Hence, this work also addresses the issue of the sorption behaviour of four different surrogates for small carboxylic acids which are produced and converted during acetogenesis and methanogenesis.

Encouraged by academic discussions about the influence of moisture content on anaerobic degradation efficiency, a concept for experimental investigations was developed which focuses on the impact of the aforementioned reduced organic matter content in MBT residual waste. Furthermore, the moisture content distribution in landfilled MBT residual waste is investigated. Due to a reduced particle size and the removal of larger bulking material, the hydraulic behaviour of landfilled MBT residual waste is different compared to conventional MSW. Preferential moisture flow pathways develop during emplacement and settlement which affect the flow of water through the waste. Low permeability due to high density emplacement may lead to a stagnated flow and/or surface run-offs [56]. Therefore, detection and monitoring of heterogeneous moisture distribution in a landfill body is critical for mechanical landfill stability and landfill gas emission control. In this work, a non-destructive technique was investigated on laboratory scale to assess moisture content distribution in MBT residual waste. Potentially, this technique can be used to quantity *in-situ* moisture content and monitor moisture distribution in MBT landfills.

2 Theoretical background

2.1 Biodegradation of organic matter

In landfills, anaerobic degradation predominates due to high moisture content at emplacement, subsequent compaction and infiltrating precipitation [6]. The entire process of anaerobic degradation passes several reactions, whereas, usually four main pathways are distinguished (Figure 2-1).



Figure 2-1: Overview of main pathways of anaerobic degradation of organic matter [8].

As substrates for biodegradation, high-molecular and water-insoluble polymers are hydrolysed to low-molecular water-soluble substances by enzymes outside of the microorganisms. These hydrolysed (smaller) substances can be transported into the microorganisms for further degradation. Two different ways for decomposing hydrolysed substances are distinguished. During aerobic degradation, organic matter is converted to carbon dioxide, water and mineral components, e.g. nitrate, sulphate and phosphate, in the presence of oxygen. In the absence of oxygen, microorganisms convert the organic matter under anaerobic conditions. With regard to the released energy (ΔG_R°), aerobic degradation of organic matter (Eq. (1)) is more favourable than anaerobic degradation (Eq. (2)).

$$C_{6}H_{12}O_{6} + 6O_{2} \rightarrow 6CO_{2} + 6H_{2}O$$

$$\Delta G_{R}^{\circ} = -2870 \text{ kJ}$$

$$C_{6}H_{12}O_{6} \rightarrow 3CO_{2} + 3CH_{4}$$

$$\Delta G_{R}^{\circ} = -418 \text{ kJ}$$
(2)

Depending on the availability of electron acceptors, organic matter is either fully degraded to carbon dioxide and water or incompletely decomposed to carbon dioxide, methane and water. During acidogenesis, the hydrolysed substances are converted into organic acids and alcohols as soluble products as well as carbon dioxide and hydrogen as gaseous products. These substances are further converted into acetic acid during acetogenesis. During these degradation pathways, the pH usually decreases markedly due to the organic acids produced. Therefore, the extent of the pH decrease, or acidification phase, represents the activity of anaerobic degradation which depends on the acid buffer intensity of the waste.

A great amount of energy is bound to methane when anaerobic decomposition occurs incompletely due to a lack of electron acceptors. During biodegradation of organic matter, several inorganic substances are successively used as electron acceptors (Figure 2-2). Redox potentials depend on the current pH, that is, redox potential decreases by 59.1 mV when pH decreases by one unit [48].



Figure 2-2: Microbiologically catalysed redox reactions and the associated redox potentials at pH 7, redrawn after Schwoerbel [67].

Landfill gas emissions were structured by Farquhar and Rovers in 1973 [14] by developing a typical pattern (Figure 2-3). After emplacement, the present oxygen is depleted and anaerobic degradation proceeds which is indicated by increasing volumetric carbon dioxide content (phase I and II). After consumption of electron acceptors, organic matter is incompletely degraded to carbon dioxide and methane (phase III). As anaerobic degradation proceeds, stable methanogenic phase is reached (phase IV).