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Mathematical Modeling and Experimental Investigation of the Pyrolysis of Waste in Rotary Kilns

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1 Introduction

Pyrolysis processes are used in the field of the thermal treatment of waste e.g. as a pretreatment step for waste or as a processing unit in combination with gasification, combustion for specially prepared waste fractions, e.g. the Siemens-KWU process in Japan or the Con-Therm process in Germany.

A mathematical model can provide an approximation for the setting of operating parameters for the optimization of existing processes, e.g. with respect to the maximum possible throughput with the same quality of the pyrolysis products, or for the setting of operating parameters for an unknown starting material. Constructive parameters for newly drafted plants can also be determined and tested with the help of a model.

This paper gives an overview of existing mathematical approaches and current development in the field of mathematical modeling and related experimental investigations of pyrolysis processes of waste materials in rotary kilns. In addition to the ~~combustion~~ pyrolysis of waste in rotary kilns related results and findings in the other fields, such as combustion of waste and cement processes, are also included.

2 General Principles

In order to be able to describe the pyrolysis process in a rotary kiln using a mathematical model, the transport of the solid and the specific conversion processes dependent upon the constructive parameters such as diameter and length of the rotary kiln as well as operating parameters (main influencing parameters) such as angle of inclination, rotational frequency, throughput and temperature profile over the length in the rotary kiln must be described. For process models, which describe such processes in a reactor, it can usually be distinguished between a reactor model and a so-called basic model. The behavior of the solid in the reactor (residence time behavior) is described using the reactor model and the material and heat transfer as well as the conversion process with the help of the basic model.

The approaches to process modeling can be roughly categorized into balance models (see **Figure 1**) and maximum-gradient models (**Figure 2**) or models divided into sections as the case may be.

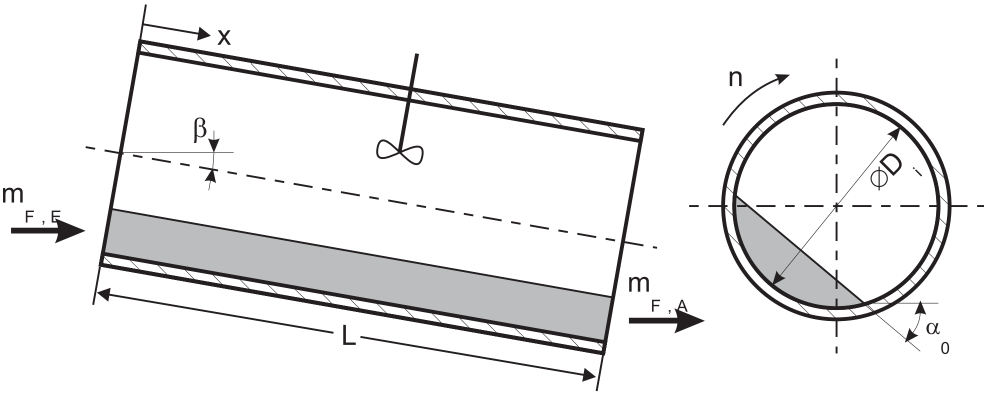

Fig. 1: Balance Model

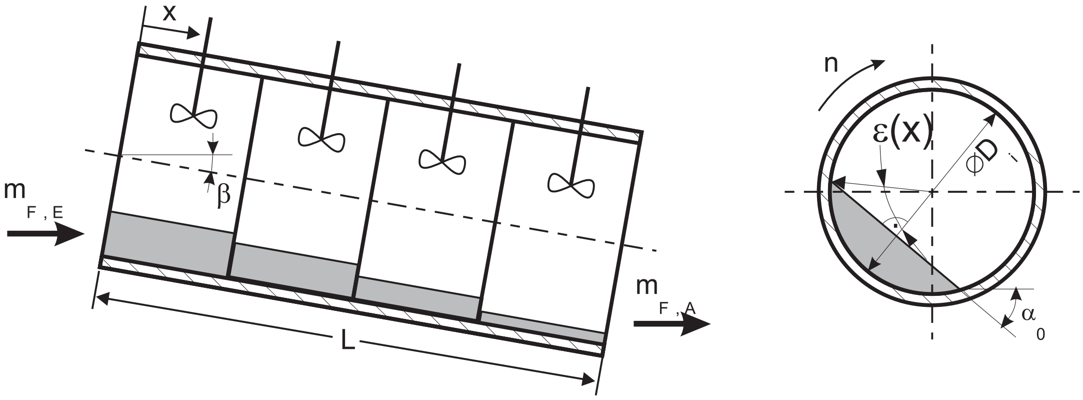

Fig. 2: Maximum-Gradient-Model

Balance models can generally be traced back to the basic element „stirred vessel“ (abbreviated as SV in the remainder of this paper). Maximum-gradient models are obtained through the connection of several SV elements or zones (sections) in series. In the following sections the state of the art regarding reactor and basic models and kinetics will be introduced before calculated and experimental results are discussed using an overall model.

3 Reactor models

The behavior of the solid, also referred to as the residence time behavior or as reactor behavior in connection with the apparatus, has a significant influence on the course of the conversion over the reactor length. Therefore, the residence time behavior and the main influencing parameters for the control of the residence time behavior are of great importance for the optimization of the overall process. The description of the residence time behavior also forms a fundamental basis for the mathematical modeling of the solid conversion. In rotary kiln systems constructive parameters, such as rotary kiln radius R and kiln length L, and operating parameters, such as mass flow m_F, angle of inclination β, and rotary kiln speed n, are important with regard to the residence time behavior (**Figure 3**).

The residence time behavior is also influenced by the characteristics of the starting materials, e.g. composition, bulk density ρ_F, angle of repose α_o, particle-size distribution, etc.

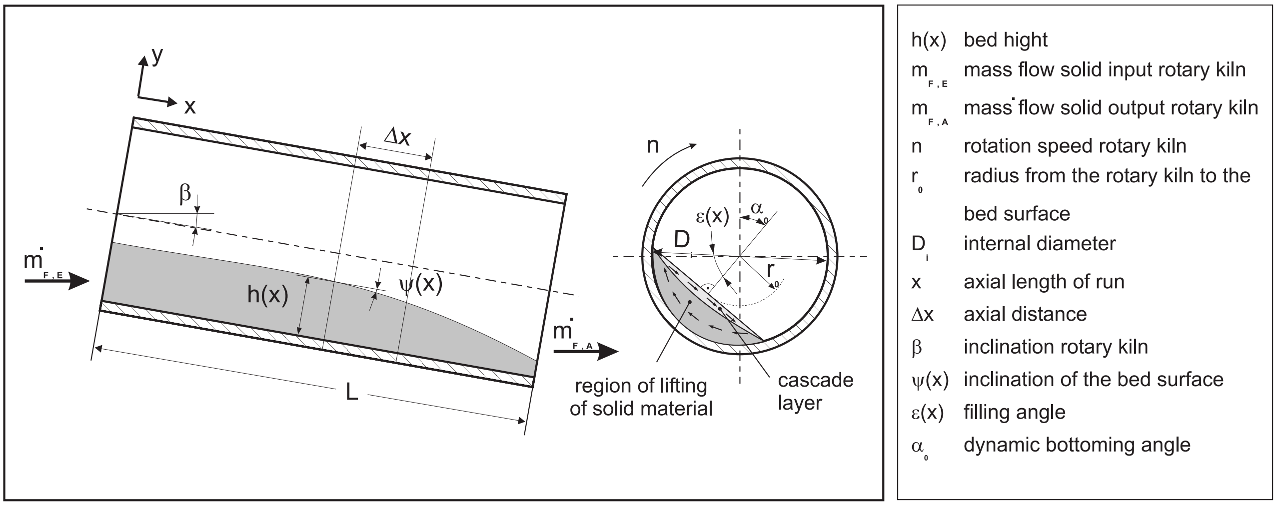

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Fig.: 3: Main influencing parameters for the material transport

Generally, the residence time behavior is characterized through the residence time distribution E (Θ) [1] and the derived average residence time τ. In rotary kiln systems in particular, the residence time behavior can be described using path-dependent values such as filling height h (x), angle of bed inclination ψ (x) and the filling angle ε(x).

3.1 Balance models

Balance models or global models and black box models do not go into the details of the solid transport in the reactor but specify the overall characteristic values such as average residence time or axial dispersion generated from geometrical or mechanical considerations and/or experimental results. For this reason balance models can be characterized by the basic element SV. According to the corresponding approaches presented in [2], the basic equation for the determination of an overall average residence time for rotary kiln systems can be defined as follows:

$$\tau = \frac{k_1 \cdot L \cdot f_1(a_0)}{n \cdot D_i \cdot f_2(\beta)} \qquad (3\text{-}1).$$

The diameter D_i is inversely proportional and the length L directly proportional to the residence time τ for a constant mass flow. In addition, the inclination of the rotary kiln β, the rotational frequency n and the dynamic angle of repose α_0 are also taken into consideration. The equation is only valid for small degrees of filling ϕ, e.g. less than 15 to 20 %.

3.2 Maximum-Gradient Models

A series of experiments to investigate the solid transport were carried out by Khodorow [3], Saeman [4], [5] and others until the 1950s. For a process control optimally adapted to the procedural operating sequence the previously used empirical residence time equations (balance models) were no longer sufficient. Through the connection of several basic „stirred vessel“ elements in series the attempt was made to calculate a defined average residence time for each process segment in the rotary kiln (e.g. drying, heating, combustion and post-combustion zones). The introduction of the traversal contact times for solid-gas and solid-wall and the inclusion of the change in cascade layer height led to an increase in the accuracy of the models.

The approached developed by Austin [6] and Rogers and Gardener [7] in the late 1970s are counted among these models.

The residence time model delivers the following parameters for the total process model:

The course of the filling angle ε(z) (**Figure 3**) with the derived values of filling height h(x), degree of filling ϕ (x) and the corresponding portions of the area covered with solid, the surface of the solid facing the gas and the free wall area, which is necessary for the heat transfer as well as the average residence time τ, as well as individual residence times Δt_i for each SV element i.

The course of the filling angle for an unknown starting material can be determined according to the model from Hogg, Shoji and Austin with few cold investigations of the residence time using the following equations:

$$\left(\frac{M\cdot z}{R}\right)=\int_{\varepsilon(z=0)}^{\varepsilon(z)}\frac{M^2\cdot\sin\varepsilon\cdot d\varepsilon}{\sin^3\varepsilon-\left[\sin^6\varepsilon-M^2\cdot\left(1-\frac{2\cdot C}{M}\cdot\sin^3\varepsilon\right)\right]^{0,5}} \qquad (3\text{-}2),$$

$$\text{with } M=\left(\frac{2\cdot\dot{m}_S}{K}\right) \text{ and} \qquad (3\text{-}3),$$

$$C=\left(\frac{\tan\beta}{\cos\alpha_0}\right)\ (3) \qquad (3\text{-}4),$$

$$\text{as well as } K=2/3\cdot B\cdot\rho_s\cdot 2\cdot\pi\cdot n\cdot R^3\cdot\cot\alpha_0 \qquad (3\text{-}5),$$

m_S	mass flow of the solid [kg/h]
β	angle of inclination of the rotary kiln [°]
α_0	angle of repose [°]
ρ_S	bulk density of the solid [kg/m^3]
n	rotational frequency of the rotary kiln [min^{-1}]
ε	half of the filling angle [°rad]
z	variable over the length of the rotary kiln, starting at the discharge [m]
B	material factor, must be determined experimentally [-].

The equations (3-2) to (3-5) are only valid for low degrees of filling. The material factor B in this equation is an experimentally determined dimensionless material parameter which physically describes the ratio of the residence time of a particle on the surface to the total residence time for a cycle in the bed and on the surface. For sand a material factor B was determined to 0,624.

When considering the heat transfer, the residence time is incorporated in the heat transfer coefficients as can be seen from the work by Frisch [8] and Mollekopf [9]. According to Vogel

[10], the time that the particle resides in the bed must be determined in order to calculate the contact time solid-wall. For the calculation of the contact time solid-gas the simplified assumption was made that the particle travels at a constant speed on the bed surface. The friction was neglected.

Zengler [11] describes the discharge of a particle out of the cascade layer with the help of a spiral. This characterizes the point at which the particle is released from the material collective as a result of a greater centrifugal force versus gravity. The particles travel along parabolas through the rotary kiln. In this approach the friction between the particle and the material bed is accounted for through the friction coefficient. The approach is limited by the fact that the centrifugal force is not necessarily greater than the gravitational force for all particles.

Another residence time model is proposed by Mu and Perlmutter [12]. In this case the solid bed is divided into N cells which differentiate between the lifting region and the cascade layer. A possible reverse mixing is taken into account through the reverse flow fraction and an unmixed relayed fraction through a so-called bypass. The parameters required for this model formulation are determined using a zonal-cylindrical solid transport model developed by Mellmann [13]. The total residence time distribution in the rotary kiln can then be calculated with the help of Laplace transformation.

4 Basic Models

In rotary kiln reactors the heat transfer to the bed takes place either

- directly through a combustion gas or
- indirectly through an external heater.

The type of energy input depends upon the respective process carried out. For example, the heat for the endothermic calcination of raw material is transferred through a hot gas. For process under inert atmosphere, such as pyrolysis, the energy is usually introduced from the outside, e.g. electrically or via internal gas-heated radiating pipes.

The partial processes radiation, convection and conduction are coupled with one another in the type of heat transfer. The material conversion processes also play a role in the energy balance. In order to couple the material conversion processes with the heat transfer and for the determination of heat transfer surfaces for gas, wall and solid, information concerning the solid transport and the residence time are required. A classification of the model into different categories according to approach is carried out in a similar manner as for the solid transport models.

4.1 Balance Models

Balance models with the basic element „stirred vessel“ (global models) describe the heat transfer mechanism in a rotary kiln in its entirety, i.e. the fundamental equations for the heat transfer are based on the entire rotary kiln without the differentiation into individual sections or zones. Heiligenstaedt [14] described the heat transfer in rotary kilns for various types of drum designs. His approach provides the equations for the convective heat transport and the radiation. The objective is to determine the heat flow transferred to the material.

The model developed by Biardi [e.g. 15] for the waste combustion includes solid mass and energy balances for the entire rotary kiln in addition to the overall solid residence time according to Equation 3-1. The waste conversion is determined by the transport coefficients from Sherwood functions and stoichiometric coefficients, among other things. The model delivers reference values for the dimensioning and operation of large-scale plants as a function of varying input values. The simplified approaches make it possible to provide a sufficiently accurate description of complex circumstances.

4.2 Maximum-Gradient Models

The model according to Gardeik [16] and the rotary kiln model according to Wocadlow [17] can be classified as maximum-gradient models. Gardeik's work deals with the heat transfer in internally heated, adiabatic rotary kilns. He developed two model approaches. In the first approach gas, solid and wall are considered as homogenous, in the second, the ~~instationary~~ non steady state behavior of the wall during a rotation is also included. Model II can then be verified through an examination of the limiting values of Model I. Analogous to the recuperator equations, the effective transport coefficients are used to describe the axial gas temperature profile. Additionally, a so-called storage factor, which accounts for the intensified influence of the rotary kiln wall on the heat transfer from the gas to the solid, is defined. In order to estimate the influence of practical parameters, the model must be expanded to include heat radiation and heat loss. Similar approaches have been suggested by Frisch [18].

In the model according to Wocadlow, the axial temperature profile and concentration in a pyrolysis rotary kiln were estimated mathematically for the soil remediation by the Deutschen Babcock AG and the Ruhrkohle Westfalen AG. Thereby, the soil output temperature, a measure for the quality of the decontamination, is of particular interest. The penetration model according to Mollekopf is used to describe the heat transfer from the wall at the bottom (underside) of the bulk material. Through the modification of this model the heat transfer at the bulk material surface and bottom can be described. Since the gas temperature in pyrolysis processes is less than in combustion processes, the gas radiation can no longer be assumed to be gray. The calculation of the entire heat quantity absorbed and effectively transferred by the rotary kiln wall, the solid and the gas is carried out using the model from Adams and Brimacombe [19], [20].

The multi-dimensional heat transfer model according to Mellmann similar to Ghenda [21] incorporates the course of the axial filling angle in the rotary kiln and therewith changing cascade layer height from zone to zone. The mass and energy balances for each zone form the foundation of this model. The calculation of the heat transfer from the wall to the material and from the gas to the material is based on work by Frisch (penetration model) [8], [22], [23], [24], [25], Lehmberg, Hehl and Schügerl [26], [27]. In comparison the zonal-cylindrical model from Bauersfeld does not regard the changing cascade layer [28].

The rotary kiln is differentiated into gas, bed and wall zones. The bed zone is further divided into the cascade layer and the lifting region. The calculation of the gas flow in the flame-heated rotary kiln can be carried out using comprehensive mathematical models from Bundrock [29], [30]. Thereby, the differential equations from the movement, continuity and enthalpy equations are solved numerically. Input flows over the rotary kiln length due to secondary air or resulting from chemical reactions in the solid bed can be incorporated. It also includes the prediction of various flows, the variation of geometrical boundary conditions and the determination of the configuration of heat transfer surfaces and can also be coupled with the partial models for heat transfer, solid transport and chemical reactions. In the operating procedure described, each axial solid section can be balanced according to the energy. A boundary condition is given by the product input temperature at the start of the rotary kiln. In order to minimize the amount of calculation required, such a zonal breakdown according to the variation of the model parameters should be chosen.

For the modeling of the incineration of low calorific solid waste in rotary kilns Ruppert [31] developed a model that describes the chemical carbon burnout of single particles as well as the movement of the particle in the rotary kiln. Therefore, numerical (maximum-gradient) models of a porous body and a non-porous body with a surrounding ash layer were combined. The models were verified by experimental results.

The basic model developed by Gehrmann [32] is based on the mass- and energy balances (equation 4-1 and 4-2). The description of the solid transport gives rise to a maximum gradient model, i.e. only the change over the length of the rotary kiln is considered, not over the bed height. A small stirred vessel element with a volume dV from the point of view of an "observer riding along" is regarded for the conversion processes. The stirred vessel element is situated at a certain position z along the longitudinal direction of the rotary kiln after a certain residence time dt. At this position, the stirred vessel element undergoes material and energy exchange with the gas atmosphere flowing above and energy exchange with the wall of the rotary kiln. Dependent upon the temperature, the mass dm is transferred from the surface to the gas flowing above it in a certain time period dt in the stirred vessel element dV. The changing of the solid's enthalpy is equal to the sum input and discharge as well as formation/consumption.

$$\frac{dH_S}{dt} = \frac{dm_S * c_{S,0S} * (T_S - T_0)}{dt} + \frac{d(T_S - T_0) * m_S * c_{S,0S}}{dt} = \dot{Q}_{WS,Sch} + \dot{Q}_{WS,\varepsilon} - \dot{Q}_{SG,\alpha} - \dot{Q}_{SG,\varepsilon} - \frac{dm_S}{dt} * \left|\Delta h_R\right| \tag{4-1}$$

$Q_{WS,Sch}$	Heat flow from the wall on the side of the bulk material into the solid due to radiation and conduction,
$Q_{WS,\varepsilon}$	Heat flow from the wall opposite the solid due to radiation under consideration of the secondary radiation of the gas,
$Q_{SG,\varepsilon}$	Heat flow from the solid into the gas due to radiation under consideration of the secondary radiation of the opposite wall,
$Q_{SG,\alpha}$	Heat flow from the solid into the gas through convection,
$dm_S/dt*\Delta h_R$	consumption of heat due to material conversion.

The gas mass flow is assumed to be as steady-state:

$$\frac{dH_G}{dt} = 0 = \dot{H}_{G,x-\Delta x} + \dot{H}_{SG} + \dot{Q}_{SG,\varepsilon} + \dot{Q}_{SG,\alpha} + \dot{Q}_{WG,\alpha+\varepsilon} - \dot{H}_{G,x} \qquad (4\text{-}2),$$

with

$H_{G,x-\Delta x}$	enthalpy flow from the last SV,
$H_{G,x}$	enthalpy flow from the actual SV,
H_{SG}	enthalpy flow from conversion,
$Q_{WG,\alpha+\varepsilon}$	Heat flow from the wall into the gas due to radiation and conduction.

With the reactor model from Austin and the described basic model a calculation for the solid temperatures for sand at 300 °C and 500 °C wall temperature was performed.

Figure 4 shows the calculated and measured solid temperature profiles for sand and 300 °C and 500 °C wall temperature.

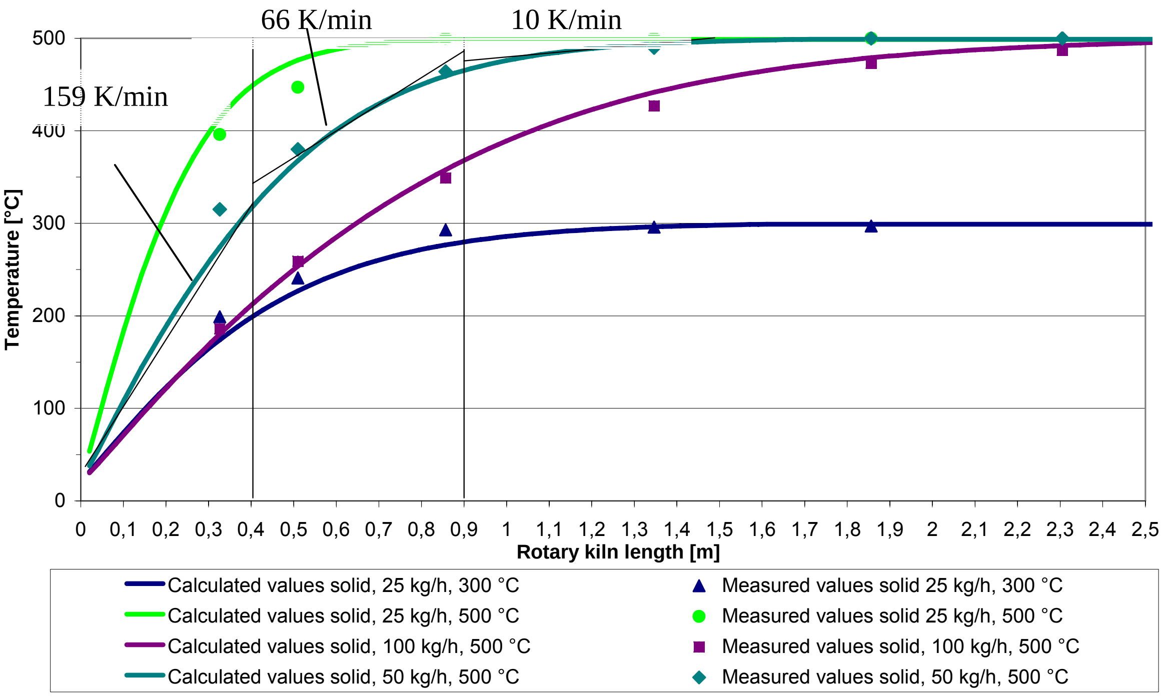

Fig. 4: Solid temperature profile (Δm/Δt = 0)

The gas flow at the rotary entrance is about 1 Nm^3/h.

The courses of calculated and measured temperatures show a good accordance.

5 Description of the Kinetics

The investigation of the kinetics can be differentiated into purely empirical investigations of the product spectrum dependent upon the pyrolysis conditions and into investigations of the product formation with respect to the kinetics.

Numerous investigations, including those by Berghoff, Bilitewski, of the product distribution in dependence upon the pyrolysis conditions have been carried out for various waste materials and reactors. In addition to these purely empirical investigations kinetic approaches also exist.

In [33] an overview of the field of applications and the fundamentals of pyrolysis as well as the dependence of product composition and distribution upon the significant main influencing parameters, such as e.g. composition of the starting material, temperature and residence time, are presented (Biletewski, Härdtle and Marek).

Heil [34] carried out investigations of waste materials with different compositions under variation of the temperature and residence time with respect to product distribution and composition in gas, coke and tar in a parallel-flow shaft furnace. The heavy metal and pollutant content of the products, such as e.g. H_2S, NH_3 and HCN, were of particular interest.

Similar investigations were carried out by R. Berghoff [35] and H. Federle [36].

A general overview of the utilization of plastic waste through pyrolytic degradation was presented by Sinn and Kaminsky [37]. The objective of these investigations was to point out to what extent pure plastic and plastic mixtures can be thermally utilized through pyrolysis in a fluidized bed reactor. Following a structured planning of the experiment, the yield of the individual components in the gas during the pyrolysis of PE, for example, was approached through regression equations depending upon temperature, throughput capacity and fluidized bed gas flow rate. With the help of these equations optimization calculations could be carried out with regard to the methane output, for example. In addition to the profitability considerations for a large-scale plant, individual investigations of the dependence of the pyrolysis of PE, PVC and PTFE on the residence time, temperature and catalytic influences were carried out. Thereby, it was determined that during the pyrolysis of PVC approximately 90 % of the bound hydrogen chloride could be recovered as pure hydrochloric acid through the use of activate carbon adsorbers.

Investigations in the field of pyrolysis of the light-weight fraction of shredder waste were carried out by C. Pasel [38] and M. Hamm [39].

The investigation and ~~modelling~~modeling of the drying and pyrolysis of fir and birch wood spheres in a thermobalance was undertaken by Staudinger and others [40], [41] with the goal of simulating the material and heat transfer processes in a fluidized bed. The model only takes into account one dimension of the particle. Differential mass and energy balances as well as thermodynamic equilibrium correlations and kinetic reaction equations are drawn. With the help of the Arrhenius function, the product formation during the pyrolysis of wood is described. The simultaneous solution of the differential equation is done numerically.

Similar investigation and model approaches were carried out by Wiest [42] and Schinkel [43] for the pyrolysis of biomass, maize in particular, in a rotary kiln.

During the analysis of the pyrolysis of PVC and plastic mixtures by Bockhorn [44] using a thermogravimetric mass spectrometer, the kinetic parameters of the decomposition and the product formed were determined. The decomposition rate or the degree of plastic conversion, as the case may be, is calculated using a degressive approach with the help of the Arrhenius function. The kinetic parameters can be determined through the logarithmic representation of the equations. For example, the decomposition of PVC occurs in two steps: In the first step, the sample loses around 60 % of its initial weight between 220 and 375 °C at the maximum rate of decomposition. The calculated apparent reaction order is given to be 1.49. Between 375 and 530 °C, a further 30 % of the mass is lost in a second step. The decomposition rate is thereby relatively small. On the basis of the TG and DTG curves an apparent reaction order of 1.57 can be determined.

A mathematical approach for the ~~modelling~~modeling of the combustion of waste in rotary kilns is presented by Rovaglio, Manca, Biardi and Falcon [15]. The model accounts for mass, material and energy balances for solid and gas phases as well as the solid residence time. For the solid conversion, transport approaches, for example, are drawn up with the help of Sherwood functions. In comparison with experimental investigation, the global mathematical approach has proven itself as debitable and practical.

For the model of Gehrmann [32] the conversion processes proceed dependent upon the process conditions (temperature, concentration) in the SV element~~.~~. The determination of kinetic data for mixed waste materials is not recommended for the mathematical modeling due to the ~~heterogenous~~heterogeneous and frequent changes in composition. The use of so-called effective kinetic data is more suitable. In Gehrmann [~~31~~32] the course of conversion for a waste material or mixture of wastes is determined through experiments using a thermobalance. The heating function $\vartheta = f(L)$, important for the pyrolysis processes under inert atmosphere, is assumed at first and then underlies the corresponding residence time behavior as the heating function $\vartheta = f(t)$ for the investigation of each waste or waste mixture using a thermobalance.

The results from the thermogravimetric investigations are inserted into the model and a corresponding heating function $\vartheta = f(L)$ over the length of the rotary kiln results. If this does not coincide with the curve specified by the thermogravimetric investigations, then the procedure must be repeated. For a specification of a first heating function following steps has to be done:

An axial temperature distribution in the solid is calculated for the starting material using the reactor model (Fig. 4, without reaction at first, then with an approximation of the course of the conversion).

This serves as first step for the investigations using the thermobalance. When the curve is divided into 4 sections, a heating rate can be determined for each section which can then be carried out in the thermobalance. However, if ~~homogenous~~homogenous materials are considered, the kinetic approach, similar to the investigations mentioned above [44], can also be used for the ~~Gehrmann~~ model [32]. For example for a continuous heating rate of about 10 °K/min the curve of decomposition of polyethylene (PE) (**Figure 5**) was measured.

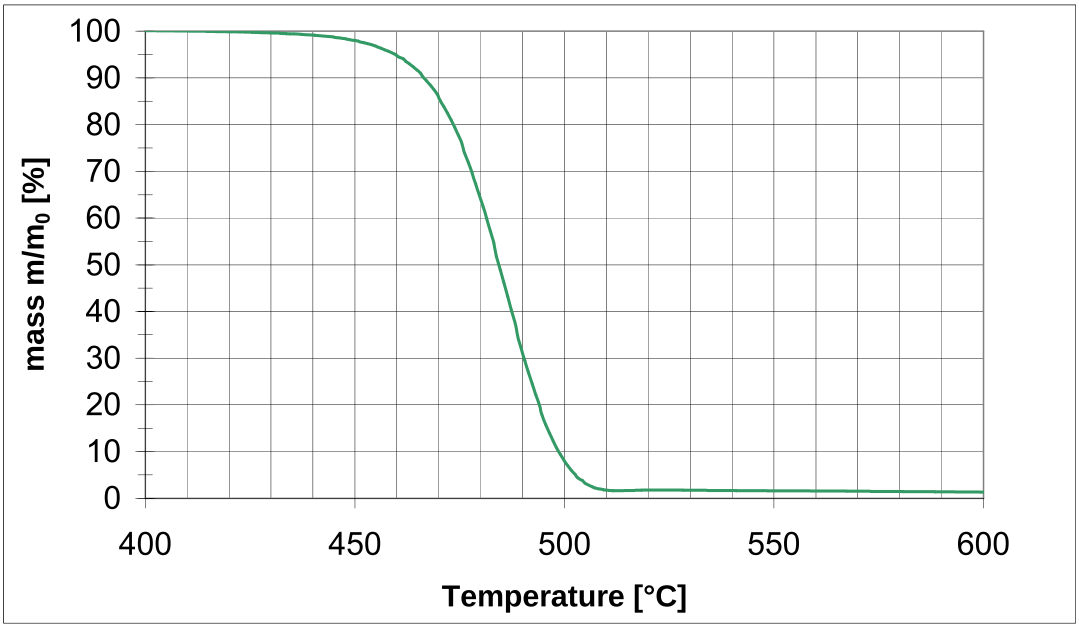

Fig. 5: Result from an investigation with the thermobalance with PE at a heating rate of 10 K/min

These curve could be approached by these function:

$$\frac{d\alpha}{dt} = +k_{\infty} * \exp\left[(-\frac{E}{R*T}) \right] * (1-\alpha)^n \text{ [43]} \qquad (5\text{-}1),$$

$$\text{with } \alpha = \frac{m_0 - m(t)}{m_0 - m_{\infty}} \qquad (5\text{-}2).$$

The determined values from Fig. 5 are:

n = 0,76, E = 262 kJ/mol, k_{00} = 9,10*10^15 1/s.

m_0 mass in the first stirred vessel at the time t = 0,

m(t) actual mass at the time t and

m_{00} mass in the last stirred vessel.

Then follows with the mass flow m_0 as actual mass flow into a stirred reactor element and m_{00} as mass flow out of the rotary kiln:

$$\frac{\Delta m_S}{\Delta t} = (\dot{m}_0 * \Delta t_0 - \dot{m}_{\infty} * \Delta t_{\infty}) * k_{\infty} * \exp\left[(-\frac{E}{R*T} \right] * \left[1 - \frac{(\dot{m}_0 * \Delta t_0 - \dot{m}_S(t) * \Delta t)}{(\dot{m}_0 * \Delta t_0 - \dot{m}_{\infty} * \Delta t_{\infty})} \right]^n \qquad (5\text{-}3)$$

With regard to the equations (4-1) the courses of the solid and the gas during conversion of PE 10 kg/h and sand 90 kg/h could be calculated as it is shown in **Figure 6**.

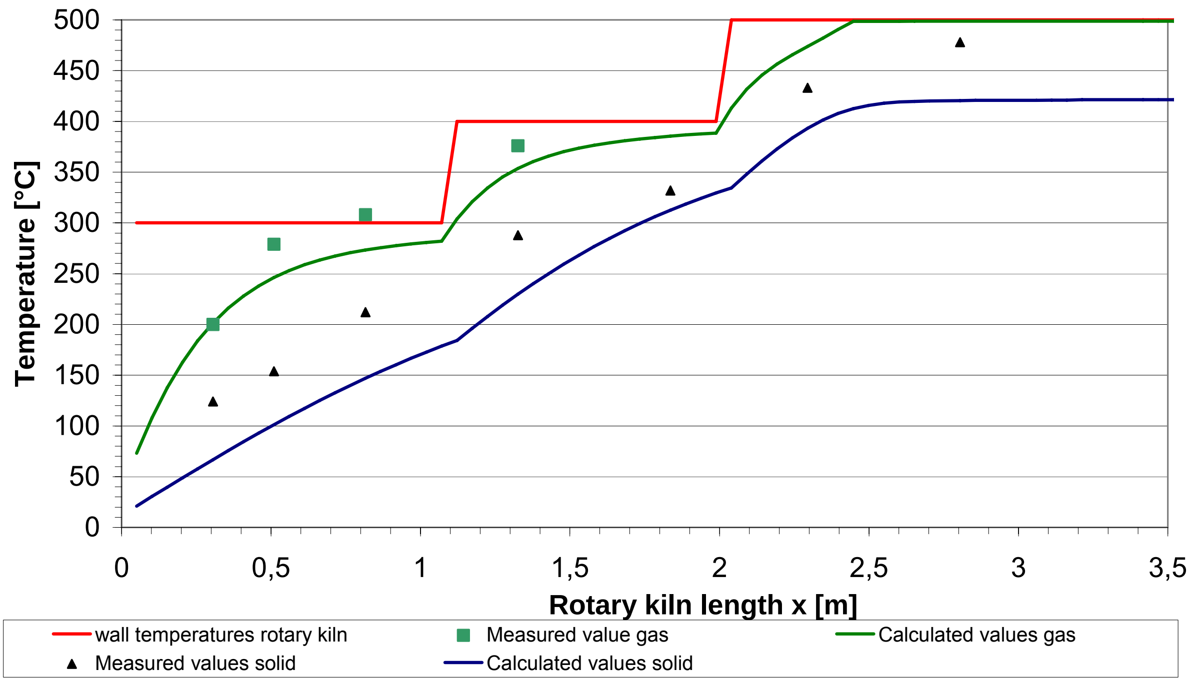

Fig. 6: Calculated and measured temperature profiles of solid and gas phase with a staging of the wall temperatures of 300/ 400/ 500/ 500 °C with 90 kg/h sand and 10 kg/h PE

The gas flow (nitrogen) at the rotary entrance is about 0.8 Nm^3/h. The reaction enthalpy $\Delta_R h$ was assumed with 4.600 kJ/kg, $\Delta t_0 = 12$ s, $\Delta t_{00} = 3$ s for 100 SV and a length about 5,1 m.

The results show that the model can describe the residence time behavior and heat transfer (Figure 4) as well as the thermal conversion of homogenous material already plausible in comparison with experimental results. With this mathematical model for the description of pyrolysis processes a determination of the dependence on the main constructive and operational parameters in a rotary kiln is possible.

The modeling of the gas composition and the validation for heterogeneous waste material is currently under development.

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