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Municipal Waste Pyrolysis: Comparison of Laboratory Scale and Pilot Scale Results

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1 Introduction

Substitute, among other, fuels are implemented in order to save fossil fuels in the raw material industry (e.g. cement production) and in power plants or for the operation of decentralized waste treatment plants with relatively uncomplicated flue gas purification systems. These substitute fuels must be tailored for each individual plant and process technique with respect to the material characteristics [1], (e.g. chlorine heavy metals). If these substitute fuels are derived from waste (e.g. municipal waste), then even a pollutant release is very difficult with a mechanobiological treatment with corresponding expenditure since the pollutants exist in nearly all material (paper, metal, plastic, textile, etc.) and particle fractions [2]. A thermal pretreatment through pyrolysis or gasification is a possible alternative to the mechanobiological treatment for the preparation or supply of substitute fuels. The implementation of a pyrolysis in a rotary kiln before a power plant reactor (ConTherm, [3]) and of a fluidized bed gasificator before the calcinator in a cement calcination process (Rüdersdorfer Cement Plant [4]) have been mentioned as an example. Basic thermal processes (pyrolysis, gasification, and combustion) are therefore now meaningful as not only as a part of a waste treatment process but also as process components for the treatment or preparation processes in the raw material industry and in power plants [5]. In both cases, the development and optimization of primary measures for pollutant breakdown or binding is still an important field of research and in industry.

The first experiments for the investigation of primary measures are carried out on the laboratory scale. Further investigations are then continued on the pilot plant scale and must finally be transferred into the practice. Large uncertainties often occur when transferring the results from one scale to another,

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making even an approximation impossible. In this paper the pyrolysis process in a rotary kiln is examined.

In order to utilize the products resulting from the pyrolysis, mainly the hot pyrolytic gas and the coke, energetically and/or physically with low pollutant loads, additives which bind chlorine and heavy metals in a water-soluble form during the pyrolysis can be introduced to the process. These compounds can then be washed out of the coke. The coke can then, after an additional separation of metals and impurities, be either incinerated directly or sold as dried substitute fuel.

Laboratory and pilot plant scale experiments showed that variable leaching rates for the chlorine compounds out of the coke depend upon the geometry and operating parameters. In order to systemize the influences, qualitative and quantitative experiments to determine the residence time-behavior with and without material conversion were carried out in addition to the hot experiments. Significant results are, for example, considerations about the scale-up factors, which make a basic transferability of results from the laboratory to the pilot plant scale possible. Therefore, the fundamental independent variables for the pyrolysis will be discussed here. The corresponding plants will then be described and the individual results will be discussed in connection with the considerations for the scale-up factors.

2 Main Influencing Parameters of Thermal Treatment

The main influencing parameters for the thermal waste treatment plants can be roughly divided into material characteristics, amount of oxygen available, reaction gases, temperature, pressure, reactor properties, material input, residence time, and additives as shown in Figure 1.

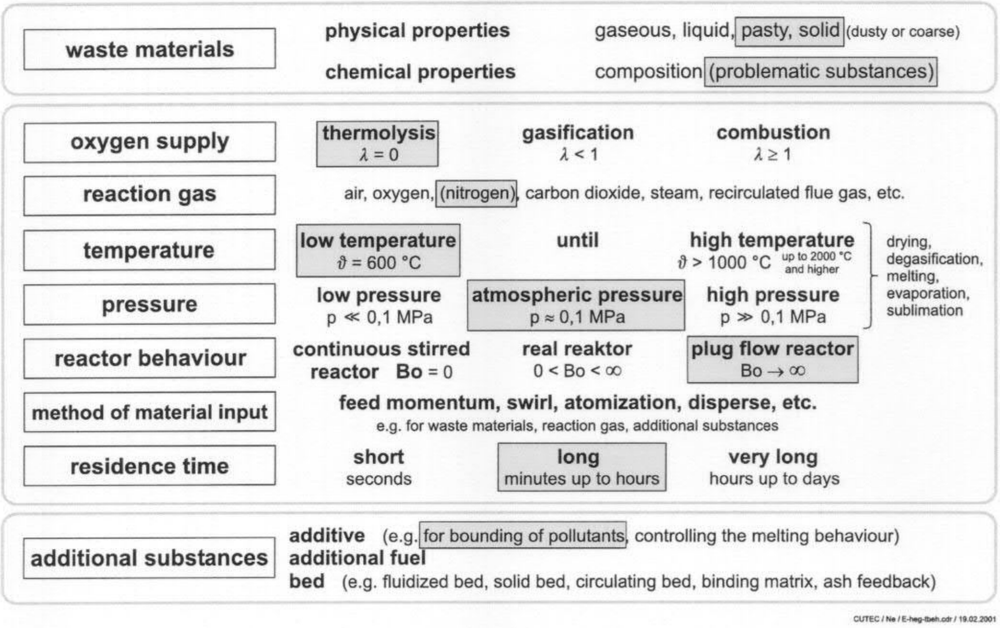
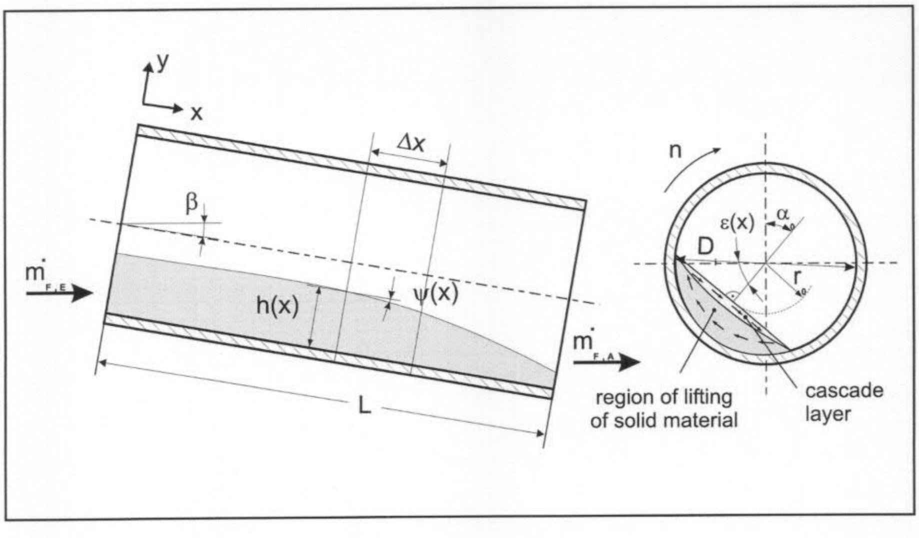


Fig. 1: Main Influencing Parameters of Thermal Treatment

The main independent variables with individual characteristic values for the pyrolysis of waste in rotary kiln systems presented here are labeled in Figure 1. The solid is treated under exclusion of air at ca. 500°C and atmospheric pressure in a solid bed or rotary kiln. The reaction behavior can be approximated as a plug flow reactor (Bo → ∝) lengthwise along the rotary kiln and described as a stirred reactor (Bo → 0) along the rotary kiln cross-section in a segment Δx (see Fig. 2, Bo-number characterizes the mixing conditions). The residence time amounts to ca. 30 minutes to 1 hour. Additives can be introduced with respect to the binding of pollutants or to influence bed characteristics. The material conversion process in the bed, in other words the pyrolysis or the coke formation as well as the pollutant release and binding, are dependent not only upon the values of the primary independent variables but also upon their time course or the course over the length of the process. Basically, the main influencing parameters can be divided into fundamental reaction parameters and into operational parameters. Fundamental reaction parameters are e.g. temperature, stoichiometric ratio of additive, gas atmosphere and residence time. Operational parameters depend on the apparatus used for the process. The residence time behavior of the apparatus has a great influence upon the reaction course. In the case of the rotary kiln, parameters influencing the residence time distribution (RTD)- behavior are the mass flow, rotation speed, inclination etc. (see below). In order to transfer the results from laboratory scale experiments to pilot scale and industrial practice, the so- called scaling factors or scaling laws are very important. While on the pilot scale as well as on the laboratory scale, temperature, stoichiometric ratio and gas atmosphere can be adjusted, this is difficult for the residence time. For pilot rotary kilns, the RTD- behavior is an additional parameter to consider in relation to the above-mentioned operational parameters. On the other hand, the RTD- behavior (reactor behavior) influences the conversion in the reactor. For the evaluation of this influence, a process analysis for the RTD is necessary.

As already mentioned, the process in the rotary kiln can be regarded as plug-flow over the length and as a stirred reactor along an individual segment Δx, in other words above the bed height. For the material conversion processes in the rotary kiln and the corresponding main independent variables, one must differentiate between reactions which occur along the apparatus (plug-flow) in the bed and the processes in which the reactions take place over the bed height in the rotary kiln section (stirred reactor), for example at certain temperatures. This differentiation is with respect to the choice of experiment conditions on the laboratory scale or the evaluation of the results very important.

Pyrolysis reactions are particularly sensitive to temperature and heating rate in the individual rotary kiln sections. The coke formation is carried out along the entire apparatus length. The residence time significant for this process is the residence time along the apparatus. During the investigation of certain reactions which for example occur within a definite temperature interval and therefore in a corresponding kiln section, the bed height or the residence time over the bed height is then the significant variable. The release of chlorine from the waste and the corresponding binding with the help of additives is a good example. This process occurs within a narrow temperature range [6] and will be discussed in detail in Section 4. Due to the thorough mixing conditions in the individual rotary kiln sections (stirred reactor elements) and the resulting approximate plug-flow characteristics along the kiln length, a relatively steep temperature gradient occurs across the length of the rotary kiln and a less pronounced temperature gradient over the height of each individual section. The parameters which influence the residence time along the kiln length and those influencing the residence time over the bed height must therefore be regarded separately. The explanations for the two cases are derived according to Figure 2.



$h(x)$	bed hight
$\dot{m}_{F,E}$	mass flow solid input rotary kiln
$\dot{m}_{F,A}$	mass flow solid output rotary kiln
n	rotation speed rotary kiln
r_0	radius from the rotary kiln to the bed surface
D_I	internal diameter
x	axial length of run
Δx	axial distance
β	inclination rotary kiln
$\psi(x)$	inclination of the bed surface
$\varepsilon(x)$	filling angle
α_0	dynamic bottoming angle

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Fig. 2: Main influencing parameters of the material transport in rotary kilns

The main independent variables for the residence time in the lengthwise direction include, in addition to the geometric dimensions such as kiln radius R or the kiln diameter D_i and length L, the material specific variables of bottom angle α_0 (x) and the operating parameters inclination ß, number of revolutions n and the mass flow m_F (x). Furthermore, the residence time behavior can also be described through the path-independent values such as the bed height h (x), the bed inclination ψ (x), and the filling angle ε (x). For a constant mass flow, the diameter D_i as well as the number of revolutions n and a function of the inclination are inversely proportional to the residence time τ whereas the length L as well as a function of the bottom angle α_0 are directly proportional to the residence time. The following equation then results for the average residence time τ [i.e. 7]: $\tau = \frac{k_1 \cdot L \cdot f_1(\alpha_0)}{n \cdot D_i \cdot f_2(\beta)}$ (1). The constant k_1 in Equation (1) generally reflects number values for internal fittings and weirs. Zengler [8] derived, through extensive experimental investigations, a semi-empirical equation for the determination of the axial solid residence time in rotary kilns:

$\tau = \frac{t \cdot L}{\sqrt{D \cdot g}}$ with $t = 0{,}5 \cdot Fr_p^{-0{,}5} \cdot (\beta + 0{,}014)^{-1} \cdot \phi^{0{,}02} \cdot \sin\alpha_0 \cdot \Delta^{0{,}1} \cdot K^{-0{,}03}$ (2)

and thereby confirmed the fundamental dependencies contained in Equation (1). In addition, the filler content ϕ, the dimensionless particle diameter Δ, and the dimensionless wall roughness K are taken into account in Equation (2).

As previously mentioned, the residence time above the bed height is the significant independent variable for the release or binding of pollutants. This residence time is directly proportional to the corresponding bed height h(x) in each kiln section Δx. The bed height is primarily dependent upon the kiln diameter D_i or the radius R and the filler content ϕ. The filler content can be expressed through the filling angle ε (x) according to Fig. 2: $\phi = \frac{(2 \cdot \varepsilon - \sin(2 \cdot \varepsilon))}{2 \cdot \pi}$ (3).

The maximal bed height h_{max} can be described through the following equation $h_{\max} = R \cdot (1 - \cos\varepsilon)$ (4). The bed height h´ = f(R, φ) at a certain kiln radius R and angle φ ($0 \leq \varphi \leq \varepsilon$) can be expressed according to the following equation: $h' = [\sin(90° - \varepsilon - \varphi)] \cdot R \cdot \frac{\cos(\varepsilon - \varphi) - 1 + h_{\max}}{\cos(\varepsilon - \varphi)}$ (5).

The maximum bed height results from $\varphi = \varepsilon$ and Equation 5 then yields Equation 4. The maximum bed height is however not the most important variable for the binding of chlorine. The height distribution plays a much greater role in dependence upon the filling angle and the radius. When h´ is plotted against the height according to h´- Δh´ (φ-$\Delta\varphi$), the mass content which depends upon the radius at a constant filler content can be described for a certain binding rate. With the help of these considerations, the results from different reactors at constant filler contents can be compared.

3 Equipment: Laboratory and Pilot Scale Plants –Analyses

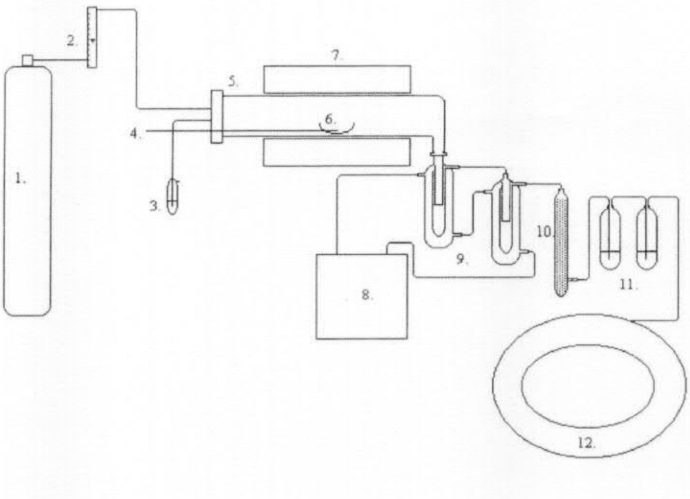
The experiment plants on the laboratory scale, the solid bed, and the batch rotary kiln and the pilot scale plants as well as the analysis methods will be elucidated here.

3.1 Laboratory Scale Plants

Test were performed in a fixed bed (see Figure 3) and a batch rotary kiln (see Figure 4).

Pyrolysis of 15 g of a mixture of 3 g PVC with 12 g limestone in a fixed bed was carried out under nitrogen flow at 500 °C for 25 minutes with a heating rate of 35 °K/min.

Pyrolysis of 110 g mixture of wood chips / PVC (10/1) with and without limestone (Ca/Cl = 3:1) in a batch rotary kiln was carried out under nitrogen flow at 500°C for 1 hour with a heating rate of 10 °K/min.



1. Bonbonne d'azote
2. Rotamètre
3. Sécurité surpression au Hg
4. Tige poussoir
5. Réacteur en pyrex
6. Nacelle en pyrex
7. Four électrique
8. Cryostat
9. Pièges à huile refroidis à -20°C
10. Piège rempli de laine de verre
11. Pièges à eau
12. Chambre à air de récupération des gaz

Figure 3: Fixed bed, ULB Brussels

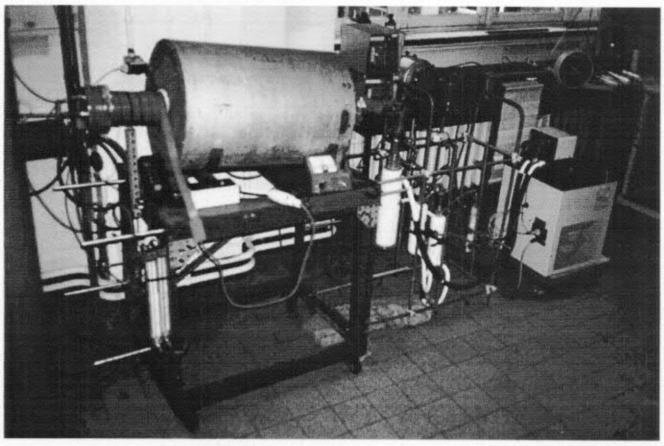


Figure 4: Batch Rotary Kiln, ULB Brussels

3.2 Pilot Scale Plant

Figure 5 shows the flow chart of the pilot plant rotary kiln used for pyrolysis.
The rotary kiln plant consists of the main components

- pyrolysis rotary kiln ([1] in Figure 5),
- washing column for purifying the pyrolytic gases ([2] in Figure 5) and
- combustion chamber for the postcombustion of the purified pyrolytic gases ([3] in Figure 5).

The material to be treated is delivered from the feeding shaft flooded with the inert gas nitrogen into the kiln via a conveyor worm. The rotary kiln has a diameter of 0.3 m and a overall length of 5.8 m. The conveyed amount can be regulated over revolutions of the worm. Depending on the input material, a maximum mass flow of about 15 to 70 kg/h can be achieved. Four individually adjustable heaters with a total electrical power of 40 kW heat the input material. The heated length of the kiln is 3.6 m. The reaction chamber of the rotary kiln is sealed off from the outside by two heat-resistant rotating mechanical seals, which are flushed with nitrogen. The maximum temperature in the kiln is not to exceed 1000 °C. At the end of the kiln, the pyrolytic coke is discharged through an electrically heated delivery chamber.

The pyrolytic gas is directed through the delivery connection piece from the kiln into a three-staged washing column filled with Raschig rings ([2] in Figure 5). Here, the hydrocarbon-loaded gas is cooled to about 30 °C and the medium and heavy-volatile fractions are washed out. The washing fluid consists of soda lye diluted in water, which is pumped through a circuit with two series-connected settling tanks. The purified pyrolytic gas arrives in the succeeding combustion chamber ([3] in Figure 5) where it is burned, along with natural gas as flame support.

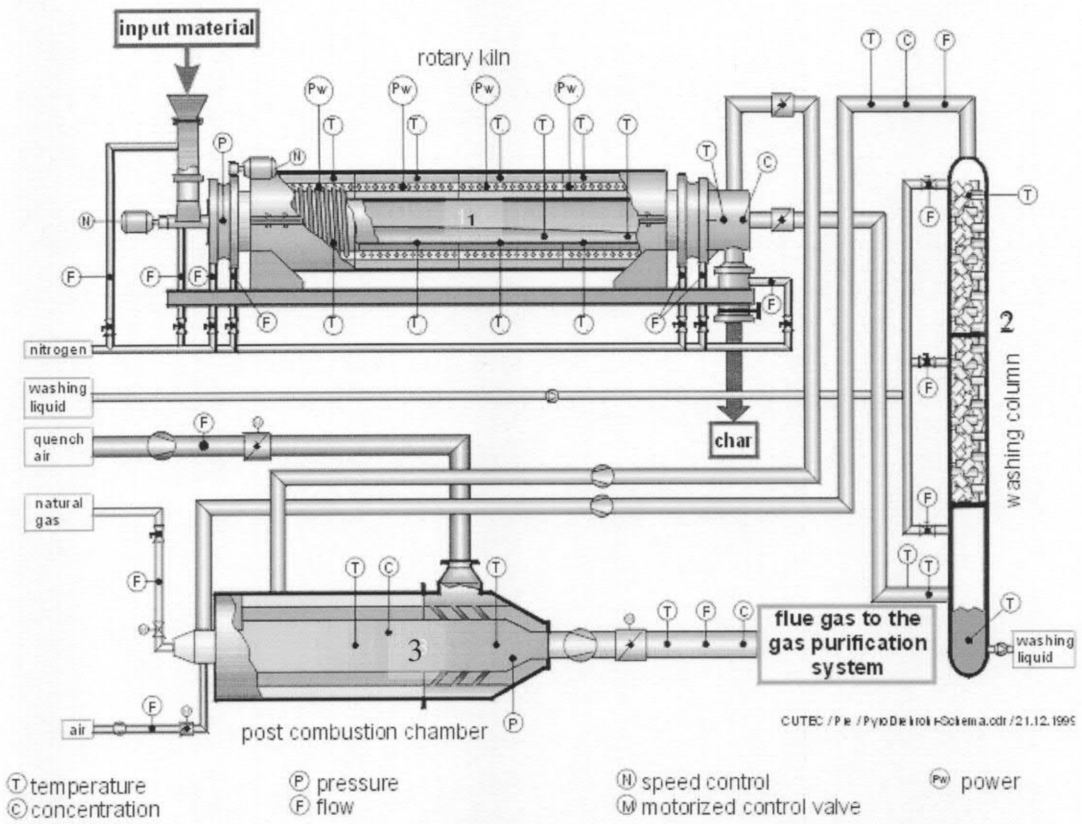


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Figure 5: Schematic presentation of the pilot plant rotary kiln, Cutec, Clausthal

All measured quantities essential for evaluation and modelling (mass, temperatures, pressures, concentrations) are recorded and processed continuously with spreadsheet software. The material flows entering or leaving the plant (input material, purified pyrolitic gas, condensate and pyrolytic coke), are sampled discontinuously in order to determine compositions and material-specific properties (e.g. calorific value or loss of red heat) of these flows.

3.3 Analyses

Leaching tests were performed on all residual solids: approximately 3 g of sample in 50 ml of water for 1 h at 50°C. Chloride anions were titrated in all the solutions with silver nitrate (potentiometric titration).

Minerals present in the char before and after washing were determined by X-ray powder diffraction (XRD) with Philips PW 1729 equipment.

Ashes were analysed using a SEM Jeol JSM 820 coupled with an energy dispersion microanalysis device (EDAX 9100). The accelerating voltage was 20 kV.

4 Chlorine Capture

The following section presents a short summary of the significant reaction pathways during the release and binding of chlorine and heavy metals during the pyrolysis and also an introduction and discussion of the results of the laboratory and pilot plant scale experiments.

4.1 Reaction Mechanism

The pyrolysis of PVC can be separated into two sections [6]. In the first section between 220 and 375 °C, ca. 64 % of the total mass reacts to a gas consisting mainly of HCl and some aromatic compounds. At ca. 380 °C, the formation of HCl finished. In the further course of the reaction up to ca. 530 °C, aromatic hydrocarbons are formed. The entrapment rate of the volatile HCl out of the PVC into the char reflects of the amount of adsorbed and chemically-bound chlorine. The chemically-bound components can be leached to a certain degree (so-called leaching rate).

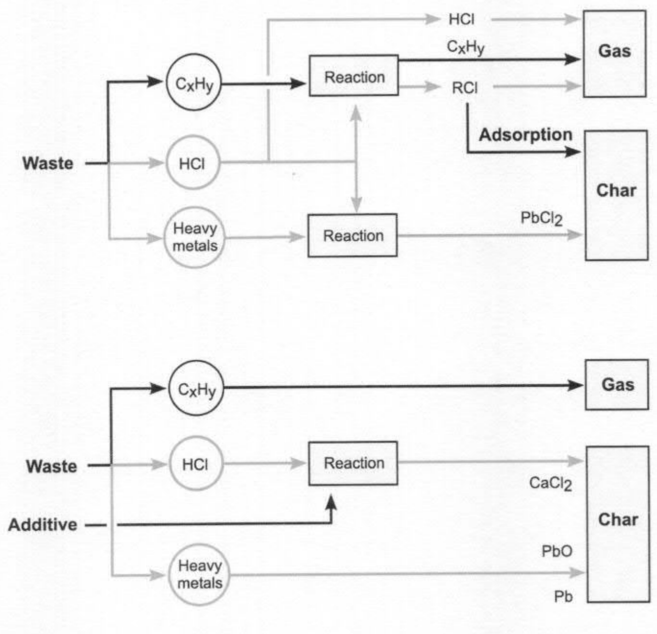

Fig. 6: Reaction scheme for the pyrolysis of waste materials without and with additives

The introduction of alkaline additives to chlorine or heavy metal-containing waste leads to a nearly chlorine-free pyrolytic gas with the simultaneous possibility to leave the heavy metals in their reduced form in the residue while the chlorine is leachable out of the residue as compounds such as calcium chloride (bottom graphic in Figure 6 [9]).

Without the introduction of additives, the chlorine remains either mainly as hydrogen chloride HCl or as a residual chloride RCl in the pyrolytic gas or is adsorbed onto the coke in a unleachable form. The heavy metal components form, among other compounds, heavy metal chlorides which are only partially leachable (upper graphic).

The residence time in a relatively narrow section of the rotary kiln above the bed is significant for the release and entrapment of chlorine, not the residence time across the length of the kiln.

4.2 Laboratory Scale Results

In all cases, less than 0.5% of the chlorine input is present in the pyrolytic gases as HCl. In the fixed bed 75 % of the chlorine input is present as leachable calcium chloride in the char. For the batch rotary kiln it was shown that in the presence of limestone 24% of the chlorine is present as leachable calcium chloride in the char, while in absence of limestone the chlorine is mainly entrapped in the char (only 1.6% leachable) [9].

4.3 Pilot Scale Results

As for the lab-scale experiments, less than 0.5% of the chlorine input is present in the pyrolytic gases as HCl on the pilot scale. The pyrolysis of wood chips at 22 kg/hr of course results in no chlorine in the char, (see Table 1, No. 1).

No.	wood (%)	PVC (%)	PbO (%)	CaCO3 (%)	Ca(OH)2 sludge (%)	mass flow input (kg/h)	mass flow output, char (kg/h)	Temperature distribution (zone 1 to 4) (°C)	Ca/Cl (molar)	Cl leachable (% of input)	Pb leachable (% of input)
1	100,00					22,0	3,8	500/500/500/500			
2	90,00	10,00				22,0	3,5	500/500/500/500		2	
3	63,63	6,36		30,00		22,0	9,9	500/500/500/500	3/1	59	
4	45,45	4,54			50,00	**11,0**	3,3	**700/600/550**/500	2/1	**86**	
5	45,45	4,54			50,00	22,0	6,8	500/500/500/500	2/1	60	
6	98,00		2,00			22,0	3,7	500/500/500/500			0,0
7	88,34	9,90	1,79			22,4	4,8	500/500/500/500		7	16,0
8	62,78	6,28	1,35	29,60		22,3	8,4	500/500/500/500	3/1	43	0,0
9	45,05	4,50	0,91		49,54	22,2	7,3	500/500/500/500	2/1	56	0,4

Table 1: Results from Pilot Scale Experiments

In the test with a mixture of wood chips and PVC (10:1) at 20 kg/hr only 2% of the chlorine input can be leached out by char washing. However, examination of the char after leaching by scanning electron microscope coupled with energy dispersion analysis confirms the presence of chlorine in non-leachable form (see Table 1, No. 2). In the pyrolysis of the same wood-PVC mixture with the addition of limestone, about 60% of the chlorine input can be leached out by char washing. Examination of the char (SEM + EDAX) confirms the presence of chlorine associated with calcium before leaching (see Table 1, No. 3). In the pyrolysis of the same wood-PVC mixture with the addition of hydrated lime containing sludge, about 60% of the chlorine input can also be leached out by char washing (see Table 1, No. 5). Calcium is also associated with chlorine in the char. The test with the same mixture but with the half of the mass flow and pre- heating produces a much better result; about 86 % of the chlorine input can be leached out by char washing (see Table 1, No. 4). After the pyrolysis of wood contaminated with lead oxide, the lead is mainly present as PbO in the char (see Table 1, No. 6). When the same input contains PVC, the lead is mainly present as PbO, with 16% in leachable $PbCl_2$ (see Table 1, No. 7). Addition of limestone to the last input causes lead to be mainly present as PbO and 43 % of the chlorine input is leachable from the char (see Table 1, No. 8). The pyrolysis of the same mixture with the addition of hydrated lime containing sludge instead of limestone generates no leachable lead but 56 % of the chlorine input can be leached out by char washing (see Table 1, No. 9).

With limestone additions to the wood-PVC-mixture, the HCl reacts preferentially with calcium to form calcium chloride. At 500 °C, the 60 % leachable chlorine from the char obtained in the pilot scale is less than the 75 % in the laboratory scale fixed bed. In batch rotary kiln only 24 % leachable chlorine was found. The behavior of the lead is closely related to that of chlorine. In absence of sorbent, part of the lead oxide reacts with HCl to form lead chloride. With limestone additions, the lead oxide remains in the char ashes.

5 Residence Time- Behavior and Scale-Up-Factors

In the following sections, the results of the residence time-behaviors without material conversion (cold investigations, Section 5.1) along the pilot rotary kiln as presented and discussed for Plexiglass. The scale-up factors are determined with the help of dimensional analysis. The considerations for the similarity during the material conversion in the lengthwise direction and cross-section of the rotary kiln are discussed in Section 5.2.

5.1 Residence Time-Behavior without material conversion

In this section, the determination of the residence time along the length of the rotary kiln will be discussed. Investigations of the measurement of residence time distributions (RTD) are described and the measured values are compared.

A Dirac impulse in the form of a defined amount of color-labeled solid particles m_0 are introduced to the stationary solid mass flow via a conveyor worm in order to examine the residence time behavior. Samples were taken after the time t_i at regular intervals Δt_i (measured after addition of the tracer into the rotary kiln) at the rotary kiln output. A representative sample distribution followed. The concentration of labeled solid particles (m_i) was then determined through enumeration and weighing. The average residence time τ can be determined from the time-dependent measured data: $\tau = \frac{\sum_{i=1}^{N} t_i \cdot \frac{m_i}{m_0} \cdot \Delta t_i}{\sum_{i=1}^{N} \frac{m_i}{m_0} \cdot \Delta t_i}$

[10] (6) The deviation σ is then calculated using the residence time distribution and average residence time $\sigma^2 = \frac{\sum_i (\frac{m_i}{m_0} \cdot \Delta t_i \cdot t_i^2)}{\sum_i (\frac{m_i}{m_0} \cdot \Delta t_i)} - \tau^2$ (7). The dimensionless deviation σ_Θ can be converted to

$\sigma_\Theta^2 = \frac{\sigma^2}{\tau^2}$, (8) and the Bo-number $Bo = \frac{2}{\sigma_\Theta^2}$ for Bo > 100 or determined implicitly from $\sigma_\Theta^2 = 2 \cdot Bo - 2 \cdot Bo^2 \cdot (1 - \exp(-Bo))$ for Bo < 100 (9).

The experiments were carried out with Plexiglass granulate at a constant kiln rotational frequency n of 3 r/min. The mass flow m_F and the inclination ß were varied. The measured residence times and bed heights with the corresponding operating parameters are listed in Table 2. Additionally, the residence time calculated according to Equation (2) (Zengler) plotted against the experiment number with the corresponding operating parameters is shown in Figure 7.

Test number	Material-parameter	Geometry-parameter	Operational-parameter			Measured values		
plexiglas	static bottom angle: 35°, Settled apparent density: 700 kg/m³ equivalent particle diameter: 1,3 mm	Length of rotary kiln: 5,1 m, Diameter of rotary kiln: 0,3 m	β (°)	n (u/min)	m_m (kg/h)	τ (min)	h_E (cm)	h_A (cm)
1			0	3	47	**94**	14	4
2			-0,65	3	46	**71**	10	4
3			-1,3	3	50	**50**	9	4
4			-1,95	3	48	**40**	7	4
5			0	3	28	**125**	13	4
6			-0,65	3	29	**79**	8	3
7			-1,3	3	30	**56**	7	2
8			-1,95	3	30	**42**	3,6	3

Table 2: Measured residence time and bed heights for experiments with Plexiglass

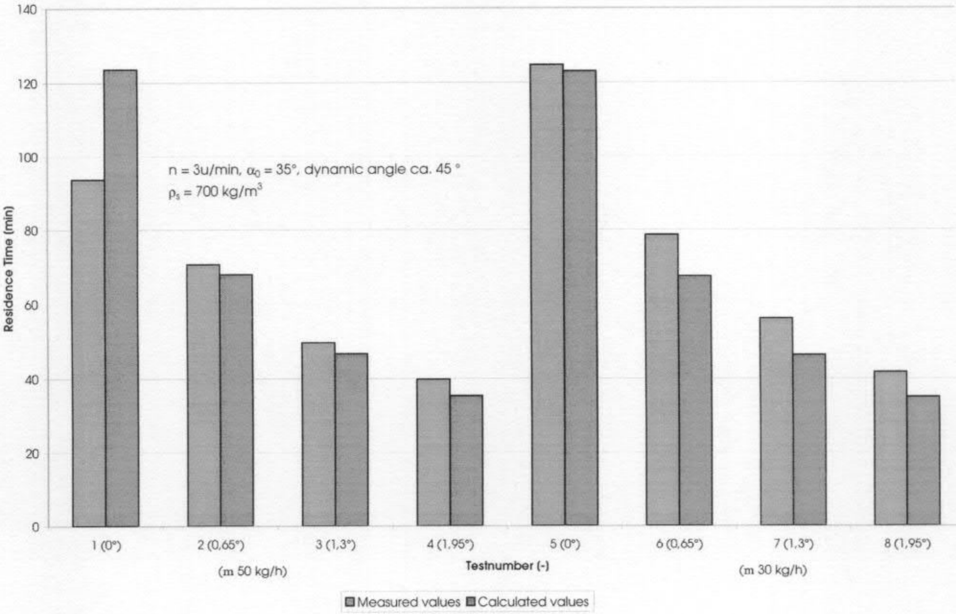

Fig 7: Measured and according to Zengler calculated residence time

The measured values are labeled with green bars, the values calculated using Equation (2) with blue bars. The computed results were adjusted to fit the measured values through the roughness parameter K for the calculation of the average residence time according to Zengler. Since the experiments were carried out in a metal rotary kiln, the roughness can be assumed to be nearly zero (10^{-6} mm). The calculated results only deviate significantly from the measured values in Experiment 1. It is however noticeable that the influence of the mass flow has little effect on the calculated residence times at constant inclination angles. The experiments confirmed that, dependent upon the input material and mass flow, the residence time is shortened with increasing inclination (see Figure 7). At constant inclination and rotational frequency, the residence time depends upon mass flow (see V1 and V5, Table 2) for an average degree of filling above ca. 25 %, which represent an input bed height of 12 cm and output height of 4 cm. The residence time increases then with decreasing mass flow at constant material volume (the input and output bed height are approximately equal in experiments V1 and V5).

However the residence time show no significant dependence upon the mass flow (experiments V3 and V7 or V4 and V8) for average degree of filling below ca. 25 %. The filling heights also fall with decreasing mass flow.

When the deviation is determined using the measured values m_i (t_i), Δt_i and the Bo-number derived according to the Equations (4 to 7), Bo-numbers of 130 to 262 result for experiments 1 to 7. A deviating Bo-value of 50 was determined for experiment 8. The Bo-numbers lie in the range for the plug-flow conditions. The results obtained in a specific rotary kiln of defined length and diameter and with certain material and operating parameters can not be directly applied to smaller or larger rotary kilns. The corresponding scale-up factors are necessary to transfer the results. The scale-up factors can be derived from the similarity principle [11]; two systems are completely similar when all the dimensionless characteristic values which describe the process have the same value. The characteristic values which describe the axial solid transport without material conversion are the Froude-number $F_R = \frac{n^2 \cdot D}{g}$ (10), the Bo-number $Bo = \frac{L^2}{D_{ax} \cdot \tau}$ (11), the degree of filling ϕ, the dimensionless residence time $\theta = \frac{\tau}{L} \cdot \sqrt{D \cdot g}$ (12) or τ_1/τ_2 according to Equation (1), and the angle ratio of the filler angle α_0 to the inclination β (sin a_0/β) (13). The influence of the particle diameter d and the dimensionless wall friction K are ignored. The influence of the particle diameter is discussed in more detail in Section 5.2. The above-mentioned characteristic values were confirmed for the most part by the experiments done by Zengler. With the scale-up factors λ_D and λ_l under the conditions $D_2 = \lambda_D \cdot D_1$ (14a) and $L_2 = \lambda_1 \cdot L_1$ (14b) from the angle relation, the inclination β_1 must equal β_2, assuming that the same materials are used. The equation $F_{R1} = F_{R2}$ results from the Froude-number $n_2 = \frac{1}{\sqrt{\lambda_D}} \cdot n_1$ (15). From $\theta_1 = \theta_2$ the equation $\frac{\tau_1}{\tau_2} = \frac{1}{\lambda_L} \cdot \sqrt{\lambda_D}$ (16a) or $\frac{\tau_1}{\tau_2} = \frac{k_1 \cdot L_1 \cdot f_1(\alpha)}{n_1 \cdot D_1 \cdot f_1(\beta)} \cdot \frac{n_2 \cdot D_2 \cdot f_2(\beta)}{k_2 \cdot L_2 \cdot f_2(\alpha)} = \frac{\sqrt{\lambda_D}}{\lambda_L}$ (16b) (with $f_1(\alpha_0) = f_2(\alpha_0)$ and $f_1(\beta) = f_2(\beta)$ as well as $k_1 = k_2$) results. From the Bo-number $Bo_1 = Bo_2$ or $\frac{D_{ax2}}{D_{ax1}} = \lambda_L \cdot \sqrt{\lambda_D}$ (18) can be derived. Furthermore, the degree of filling for both rotary kilns $\phi_1 = \phi_2$ must be equal. If the ratio L/D can be assumed to be constant, then $\frac{L_1}{D_1} = \frac{L_2}{D_2} = \frac{\lambda_L \cdot L_1}{\lambda_D \cdot D_1}$ and $\lambda_l/\lambda_D = 1$.

5.2 Residence Time-Behavior with material conversion

A complete similarity cannot be maintained with regard to the material conversion. This can easily be shown by the example of the influence of the particle diameter:

During the material conversion, the particle size decreases from that of the input material due to mass loss in dependence upon the time and the surrounding conditions. Characteristic properties of the particle then also change, such as radius, pore radii and length, internal and external surfaces, the related conditions for material and heat transfer as well as the chemical reactions. When different particle diameters are examined in a corresponding rotary kiln, different gasification behaviors, for example, must exist. Therefore, a complete similarity regarding the residence time behavior with material conversion is abandoned and a partial similarity is assumed.

For material conversion processes mainly occuring across a broad temperature range and therefore generally carried out along the length of the rotary kiln, the relation for the scale-up derived in the previous section can be of used. The relevant parameters can then be adapted so that the residence time and the accompanying heating rate on the laboratory scale correspond for example with the conditions in the pilot rotary kiln. The parameters and similarities discussed in Section 5.1 in connection with the axial solid residence time are in this case only valid for the corresponding lengthwise section Δx in which all material and geometric parameters can be assumed to be constant. Each of these sections Δx can then be regarded as a stirred reactor element or as a batch rotary kiln. This makes the representation of the continuous process in a rotary kiln a cascade of stirred reactor elements.

If material conversions mainly occur within a certain temperature interval and thus in a small section Δx of the rotary kiln, then the residence time over the bed height is the significant variable as explained above. An example for such a conversion process is the entrapment of chlorine with the help of additives (the entrapment rate represents the amount of chlorine which is leachable out of the coke). Based on the experiments in the laboratory solid bed (gram scale), a laboratory batch rotary kiln (100 g scale), and a pilot rotary kiln (several kg/h scale) described in Section 4, the question now arises, how the experimental conditions must be coordinated to make a transfer of the results possible. The following points should be taken into account when attempting a qualitative evaluation or transfer of the results from one scale to the next.

When the normal course of the entrapment rate for the entrapment reaction is examined in dependence upon the residence time (Figure 8), then one can generally determine a nearly linear increase in the entrapment rate in the range of lower residence times (range c). The entrapment rate begins to approach its limit asymptotically with increasing residence time (range a). The corresponding reactor behavior is important when discussing the shape of the curves in Fig. 8. A uniform release of HCl from the PVC in the entire volume of the packing material occurs by very thorough mixing (stirred reactor characteristic). However, with unmixed packing material, a progressive heating of the packing material must take place over the height (plug-flow characteristic). In addition to the well-known reaction technological aspects for the evaluation of stirred reactor and plug-flow characteristics, the influence of the demixing, which leads to an accumulation of the specific lighter PVC granulate on the bed surface, should be mentioned here. The HCl released from the PVC directly at the bed surface would have no contact to the additive in this case. The reaction technological aspect of the reactor characteristic and the demixing should therefore be considered when comparing the results of the experiments in the laboratory solid bed with the laboratory batch rotary kiln. The reactor behavior can achieve partial similarity using the correlation presented in Section 5.1. However, the maintenance of the similarity with respect to the residence time over height is difficult. If a certain residence time or height $h_{1,P}$ is assumed in the packing of the rotary kiln (Fig. 9), then a degree of entrapment corresponding to the minimum packing height $s_{1,P} = h_{max} - h_{1,P}$ can be determined from Fig. 8.

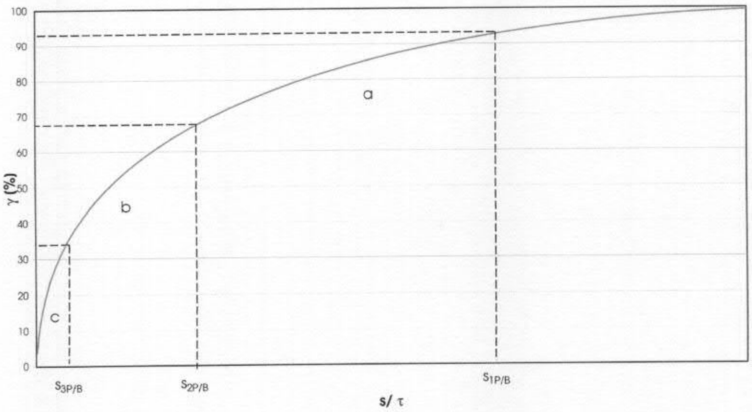

Fig. 8: Theoretical entrapment in dependence upon the packing height or the residence timet

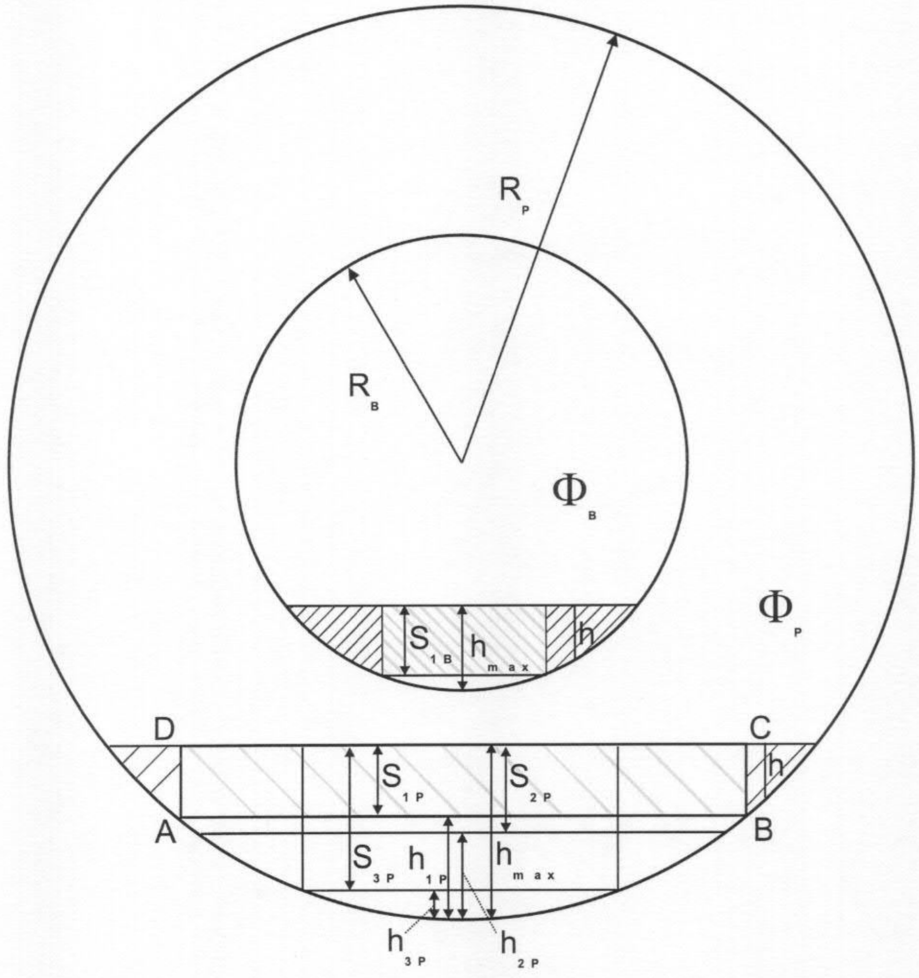

Fig. 9: Geometry at rotary kilns with different diameters

The packing height required in order to make a certain chlorine entrapment rate possible is defined as $s_{1,P}$. All layers below a height $h_{1,P}$ have a longer residence time and therefore a higher entrapment whereas the residence time of the layers above this height is shorter resulting in less entrapment. With this minimum packing height $s_{1,P}$ between the bed surface and the rotary kiln wall (segment AD or BC), a surface element is cut out of the packing. Multiplication of this surface element with the length element Δx yields the volume element ΔV which, multiplied with the density, produces a mass element Δm. For this mass element, the bed height h between the rotary kiln wall and the bed surface is equal to or greater than the minimum packing height $s_{1,P}$. The bed heights for the mass elements to the left of segment AD or to the right of segment BD are less than the minimum packing height and have shorter residence times than the previously-mentioned case. It is now important which section of the curve (Fig. 8) is considered. A reduction of the residence time or the minimum packing height s in the left section (c) causes a significantly stronger reduction in the degree of entrapment than a reduction in the range of higher residence times or bed heights (sections a and b). Three sections (a to c) will be discussed here as examples:

a) Asymptotic section: If $s_{1,P}$ lies in the asymptotic range in Fig. 8, then the decrease in the residence time in the ranges $h < s_{1,P}$ have little effect of the total entrapment rate.
b) The mass element with $h < s_{2,P}$ increases for a packing height $s_{2,P}$. At the same time, a reduction in the residence time due to the shape of the function in Fig. 8 leads to a total entrapment rate less than in case a).
c) If a packing height $s_{3,P}$ is assumed, then the sections with $h < s_{3,P}$ dominate and the entrapment rate changes linearly with the bed height or residence time in these sections.

When experiments in the rotary kiln with different radii are observed, then the same degree of filling should be maintained from the view of similarity with regard to the residence time over the bed height (see Section 5.1). At a certain degree of filling, the bed height $h = h'$ is a function of the radius R and the filling angle ε according to Equation (5). In general, $R_B < R_P$ (Fig. 9) holds true for a batch rotary kiln with index B for batch rotary kiln and index P for pilot rotary kiln. Due to the requirement for the equality of the minimum packing height $s_{1,B} = s_{1,P} = s_1$ (where $s_1 = h_{max} - h_{1,B}$), the mass element for h <

s_l in a batch rotary kiln with a smaller radius is significantly greater than that in a pilot rotary kiln with a larger radius. This would lead to lower entrapment rates for the batch rotary kiln.

The experimental results from the solid bed, the batch rotary kiln and the pilot rotary kiln will now be discussed with respect to the above-mentioned aspects of the residence time over the bed height.

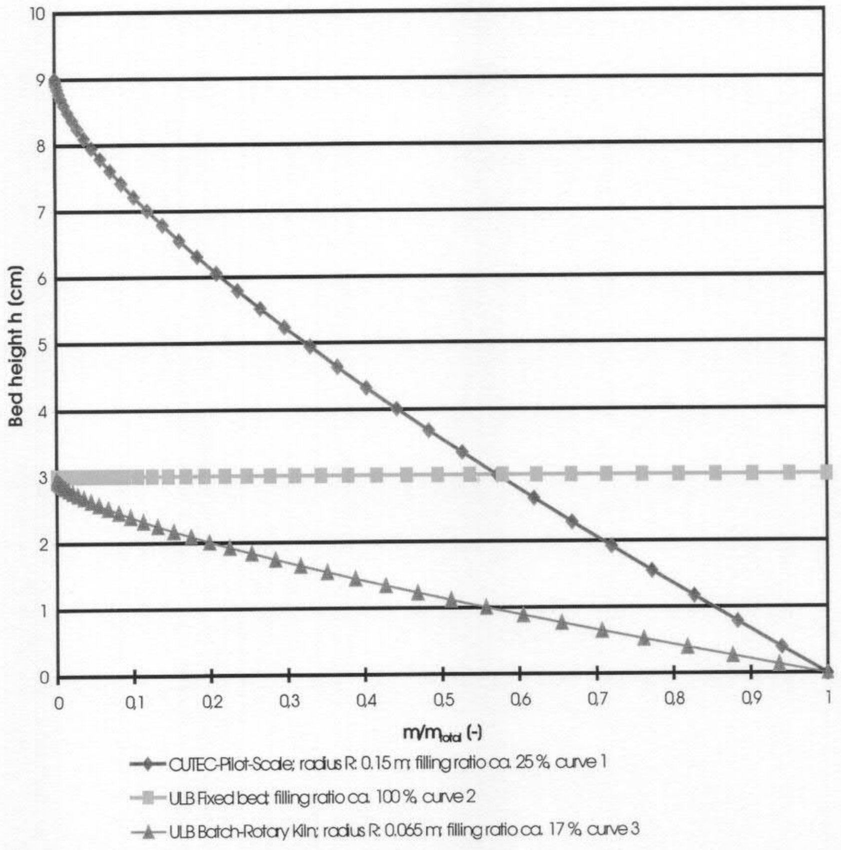

Fig. 10: Bed height as function of the mass ratio

For the solid bed, in which 100 % of the mass occurs at the same bed height of ca. 3 cm (Fig. 10, curve 2), the highest leaching rate of chlorine compounds is obtained with 75 % of the chlorine input. Due to the geometric conditions in the batch rotary kiln, a ca. 17 % degree of filling, corresponding to a maximum bed height h_{max} of ca. 3 cm (Fig. 10, curve 3), results. This explains the low leaching rate of 24 % in comparison with the solid bed. A maximum bed height h_{max} of ca. 9 cm (Fig. 10, curve 1) is achieved in the pilot rotary kiln due to the geometric conditions and a degree of filling of ca. 25 %. From Fig. 10, curve 1, one can see that ca. 44 % of the total mass is found at a bed height of less than h = 3 cm. In a comparison to the conditions in the batch rotary kiln, approximately 100 % of the mass is found at a bed height of less than h = 3 cm. This offers an explanation why the leaching rate for chlorine compounds of 60 % of the chlorine input in the pilot rotary kiln must lie distinctly above that for the batch rotary kiln.

The leaching rates of the solid bed and batch rotary kiln cannot be compared directly due to the different reactor behaviors and the mixing characteristics. The different leaching rates of the batch and pilot rotary kilns can however refer to the above-mentioned influence of the residence time over the bed height under the condition of fundamental similarity concerning the degree of filling.

In summary, the experiments confirmed that fundamental dependencies with respect to the stoichiometery of chlorine and additives, temperature, heating rate, and the residence time, etc could be achieved in the solid bed. The influence of the mixing and the reactor characteristics as well as the related residence time behavior must be investigated in a batch rotary kiln, whereby one must consider that a minimum height must be maintained in the batch rotary kiln for the transfer. A corresponding course of procedure is required for the scale-up to a pilot scale rotary kiln.

6 Symbols and Indices

t	dimensionless residence time [-],	τ	average residence time [s],
L	rotary kiln length [m],	D	diameter rotary kiln [m],

g	gravitational acceleration [m/s^2],	Fr:	Froude-number ($D n^2$)/g [-],
n	number of revolutions [u/s],	β	rotary kiln inclination [°],
ϕ	degree of filling [-],	α_0	static packing angle [°],
Δ	relative particle diameter (d/D) [-],	λ_l	scale-up factor for the length [-]
d	(equivalent) particle diameter [m],	λ_D	scale-up factor for the diameter [-]
K	relative wall roughness (κ/d) [-],	κ	roughness height [m].
R:	rotary kiln radius [m],	L:	length of the rotary kiln [m],
ρ_F:	packing density of the solid,	$h_{E,A}$:	bed height input, output
$\varepsilon_{I,A}$:	filling angle input, output,	φ, $\Delta\varphi$:	angle, angle difference [°]
h, h´, Δh´:	bed height, bed height difference [m]	P, B:	pilot/batch rotary kiln
s:	minimum packing height [m]	Bo:	Bo-number ($L^2/(D_{ax}\,\tau)$) [-]
D_{ax}:	axial diffusions-coefficient [m^2/s]		

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