The Influence of Varying Operational Parameters on Both the Combustion Efficiency in and the Emission of Pollutants from Fluidized Bed Plants 1)

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1. Methods and Apparatus

Our method of determining the influence of different operational conditions on fluidized bed plants consists in a stepwise alteration of one single operational parameter while maintaining the others as constant as possible (1). It is well known that this is easiest on a laboratory scale, whereas with increasing plant size the procedure becomes more and more onerous. If beyond operational parameters also the design concept and the size of a plant are varied, one obtains useful hints how to generalize and scale-up the results achieved.

Present findings were obtained using several types of laboratory equipment with thermal performances between 2 and 20 kW as well as from a semi-technical plant of 300 kW. Figure 1 is a schematic drawing of the shapes and dimensions of fluidized bed reactors used. Apparatus no. I is a tube reactor of 6 cm diameter and 60 cm height on top of which has been arranged a freeboard of approx. 35 cm hight and 10 cm diameter. Apparatus no. II is a tube reactor of 6 cm diameter and about 120 cm high. Here the ash is retained by an integrated inertial separator. Apparatus no. III represents a two-stage secondary air reactor with the following dimensions:

lower section:	6 cm diameter, 60 cm high,
upper section:	10 cm diameter, 80 cm high,

integrating an inertial separator. Unit no. IV is a pressurized reactor allowing combustion pressures up to 10 bar. Its reaction tube has a diameter of 6 cm and a height of 1 m, and incorporates an inertial separator. An early version of the pressurized reactor, operated at 4.5 bar, was of a similar shape and size as apparatus no. I. The reactor space provided by the semi-technical plant, finally, has a cross-section of 40 by 80 cm, a height of approx. 1 m, with a freeboard of 80 by 80 cm cross-section and approx. 2 m height.

The coal is fed pneumatically, along with all of the combustion air, to the electrically pre-heated laboratory units, whereas in the semi-technical plant coal is fed with a small fraction of the total air from one side into the fluidized bed. The atmospheric laboratory units, due to their high surface-to-volume ratio, are equipped with a heat insulation allowing to maintain a combustion temperature as high as approx. 950 °C. The pressurized unit, however, requires a variable heat exchanger for thermal discharge since in this case the heat release rate is higher by a factor of 10. As to the atmospheric semi-technical unit, it also needs heat exchangers which are immersed in the fluidized bed.

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With regard to the similarity particularly of the laboratory equipment to bigger plants, one had to compromise on this. On the one hand a contact time between gas and solids comparable to that of a bigger fluidized bed plant had to be attained which requires an adequate height of the fluidized bed. On the other hand the thermal performance was to be kept low, i.e. within the limits of a laboratory unit. As a compromise between these requirements resulted an elongated reactor shape which, seen under the aspect of flow mechanics, due to its high length/diameter ratio can at first view not be compared with a bigger plant since it tends to aggregative fluidization and pulsations. In order to be able to use these easy to be handled reactors and to obtain reliable results nonetheless, elongated wire spirals were introduced in the reactor spaces. This helped to avoid the formation of big bubbles and strong pulsations and to bring about a more particulative fluidization (2).

An essential design difference of the laboratory units consists in the substitution of the enlarged cross-section of the free board by an inertial separator. The objective of this constructional modification is to determine the functionality and need of such a high-volume free-board.

2. Results

The results were obtained from an evaluation of analysis on the feed materials, flue gases, ash, and from the material balance of throughputs.

Figure 2 is a schematic summary of the variations of the main operational parameters, including their range and direction of variation as well as relevant standard values plus qualitative effects on: specific heat release rate, C-loss, CO-, SO_2-, and NO_x-concentrations in the flue gas.

Depending on the slope and inflexion of the arrow indicating the direction of parameter variations of a given component, such variation has a stronger or weaker influence on throughput and emission; a horizontal arrow stands for invariance in respect of the independent parameter. The specific heat release rate, expressed as MW/m^2, goes up along with both increasing fluidizing velocity and pressure, i.e. along with those parameters determing the throughput of air and also of coal. The performance drops along with rising excess air, i.e. in a situation where an increasing proportion of the air throughput is no longer utilized. The other parameters, however, hardly exert any influence. The dependence of the specific heat release rate on the apparatus design, therefore, is negligible and will be -- with an excess of air $\lambda = 1.3$ (5 % O_2 in the flue gas) -- approx. 1.2 to 1.5 MW/m^2. This correspondends to the values which in the meantime have been observed also at demonstration plants (3). Most of the arrow constellations revealing a sizeable influence are backed by measuring data plotted on diagrams, a selection of which is given hereunder.

2.1. C-Loss and CO Emissions

The C-loss is a critical factor for the economics of fluidized bed plants, whereas keeping the CO content in the flue gas within admissible limits generally does not pose any problems. As can be taken from figure 2, the two data sets are of a striking parallelity. The reason for this is that the more CO will be generated at reduced temperatures within the local and thermal transition zone between reactor zone and flue gas duct, the more carbon passes through this transitional zone as char carry over. Tars and volatile hydrocarbons were not observed. On being introduced into the hot ash of the fluidized bed, the coal will be dispersed immediately and exposed to the excess air whose oxygen reacts first with the volatile matter.

Diminishing C-loss along with pressure rise is related to an increased O_2 concentration, whereas diminishing C-loss along with rising temperature is attributed to higher reaction velocity. Increasing fluidizing velocities reduce the residence time of the coal in the reactor and, thus, cause higher C-losses. If one found a means of extending this residence time -- be it by appropriate plant design or/and by coal preparation -- the specific heat release rate could be improved proportionately to the fluidizing velocity.

A longer residence time of the coal by means of increased bed height will diminish C-losses, too.

C-loss may be influenced also by coal preparation measures. As shown on figure 3, the C-loss will, when fueling closely sized coal fractions, pass through a maximum as soon as the particle diameter approches the elutriation cut point. Coarse coal grains will remain in the bed up to the moment where they are burnt down to a size allowing their elutriation or preventing them from being recycled by the inertial separators. With sufficiently small fractions (coal dust) the reaction time is apparently shorter than the residence time in the reactor space so that the coal particles are almost completely burnt up. As far as industrial plants are concerned, the logical conclusion from this is to separate coal dust from the coarser fractions and blow the dust pneumatically into the fluidized bed from below, in order to allow a maximum residence time of the dust and avoid erosion in the feed ducts, whereas the coarse fractions, being introduced from above, are allowed sufficient time to spread over the bed while being burnt up. In this case it can be taken from the diagrams, e.g. figure 3, where the preparation cut points for each specific plant are viz. which granular fraction should be separated and/or further comminuted.

Examination of the carbon carry-over by means of screening for its size distribution does not yield accurate results since char aggregations will disintegrate. The results of figure 3 are, however, reconfirmed by this approximative evaluation. Moreover it can be verified from fly ash separation in two subsequent cyclones that the fine dust from the second cyclone is very low in carbon, whereas the "coarse dust" of the first one will always contain the bulk of the unburnt carbon.

Apart from the determinable and adjustable operational parameters, C-loss is also a function of the specific plant parameters. Measuring data can best be reproduced in laboratory equipment. When doing so, one observes other and so far not measurable operational conditions which bear on the results. Among these have to be considered the size distribution of the fluidized bed ash particles which changes during operation, or changing fluidity and cohesive properties of the bed ash when adding various gradations of limestone.

A comparison of the C-loss measured in reactor no. 1 with that of the semi-technical plant V (figure 5) -- the cross-section of the free-board has been enlarged in both units -- shows good coincidence. It should be borne in mind, however, that here varying particle gradations and bed heights compensate each other in a way not clearly identified so far.

As was expected, there are differences also in the C-losses for the different laboratory reactor types since inertial separators are not optimized. This is not disturbing as long as feed materials, viz. types of limestone and coal, are compared by measurements in one reactor only. As soon as it comes to scaling up results, however, one has to know about the reasons and influences of specific operational conditions.

2.2. SO_2 Emission and NO_x Emission

Statements on the pressure-dependences of SO_2 and NO_x emissions (figure 2) are so far based on measurements of two pressure levels (1.1 and 4.5 bar). When moving to the higher pressure SO_2 and NO_x emissions will be diminished by more than 50 %. The qualitative evaluations of early orientation tests on the new pressure apparatus IV where measurements at several pressures between 1 and 10 bar are to be carried out, do reconfirm this improvement.

Figure 10 shows the strong dependence of SO_2 emissions on the size of the limestone (at one fixed Ca/S ratio). It is, however, striking and so far not explainable (figure 5) that varying sizes of one same type of limestone lead to different temperature dependencies. Dolomite shows a similar behaviour (figure 6). When adding coarse material, SO_2 emission will slump with rising temperatures, whereas the opposite happens when material of small grain sizes is added.

Excess air (figure 7) has a weak influence on SO_2 emission, while its effect on NO_x emission is strong since in this case the oxygen concentration is decisive for the conversion of that proportion of fuel-nitrogen which is transformed to NO. (Due to the low temperatures in a fluidized bed plant, 10 to 30 % only of the fuel-nitrogen and no nitrogen from the air is converted to NO.) A lack of O_2 favours rather the competing reaction which yields molecular N_2.

The dependencies of SO_2 and NO_x emissions are mostly active in opposite directions (figure 2). A good insight into the conditions leading to NO formation is possible with the secondary air reactor. When plotting the NO_x emission against different incremental primary air/secondary air ratios, a NO_x minimum is met at a distribution of primary versus secondary air of 50 : 50. This effect was most obvious in reactor no. 1 (4), whereas it was less pronounced in the bigger reactor no. III where, at the same time, the maximum emission values were reduced. The lowest values were abserved in the semi-technical reactor (figure 8); they were in the same order of magnitude which prevails also in other larger plants. From this may be concluded that the NO emissions measured in the laboratory reactors are atypically high. The formation of coarse bubbles in big reactors bring about a certain distribution of O_2 as high as into the upper bed zones. Such by-pass effect can be compared with the feed of secondary air. Consequently, the way how oxygen acts on the coal and, possibly, the removal of reducing volatile coal components are main determinants for NO_x emission. Emissions from the semi-technical plant may be considered typical for NO_x emissions from full-scale plants (figure 8), with 1 g NO_2/kWh being approximately equivalent to 200 ppm NO. So, even though the numerical values obtained from laboratory measurements are exaggerated in respect of their absolute value and do not allow any generalization as to NO_x emissions, one may nonetheless derive certain tendencies (e.g. dependence on excess air) which can be scaled-up to bigger plants.

Typical figures for SO_2 emissions cannot be identified since the main influential factor on SO_2 emission will be the amount of limestone added. SO_2 emission, therefore, is only to a small extent typical for a given plant. Figure 9 shows a dependence on the limestone/sulphur mole ratio when feeding different coals high in ash and with varying sulphur contents, but adding one same limestone at pressures of 1.1 and 4.5 bar. It is the spontaneous desulphurization of coals rather than their sulphur content which brings about the difference in SO_2 emissions, -- emissions which on a laboratory scale can be reduced to zero. To find out the limestone with optimum sulphur capturing efficiency (i.e. accomplishing the desired desulphurization with admixture of the possible minimum sorbent amount), some three dozens of limestomes of different geological formations, deposits, and trade marks were tested (5). Geologically young and porous limestones appear to be best suited.

Similar differences as to sulphur capturing properties can be observed also among dolomites. In this case only the $CaCO_3$ proportion acts as a sulphur capturing medium. Optimum desulphurization is a function of the size of limestone particles (6). As can be seen on figure 10, limestone dust < 10 µm is an excellent sulphur capturing medium due to its big surface and thorough distribution in the fluidized bed and this notwithstanding its short residence time. Unlike this, the residence time of coarser fractions with a more reduced total surface is too short as to allow adequate reaction with SO_2. Those particles, however, which are not elutriated and, therefore, accumulate in the fluidized bed on having been fed continuously to it, provide again a very good sulphur capturing efficiency. As soon as particle sizes increase further, however, this beneficial effect is lost again. Such loss of efficiency along with increasing particle size is less pronounced with dolomite due to the fact that here the percentage of magnesium carbonate enlarges the pore volume during combustion and this volume does not get blocked by sulphate formation.

2.3. Halogen Emission

At the temperatures prevailing in a fluidized bed plant, fluorides and chlorides as mineral components of the coal are released as HF and HCl also in the presence of lime. Early results have shown that on condition of low temperatures in the flue gas duct, HF and HCl can be bound by lime-containing fluidized bed flue ash. Trials on an optimization of these bonding conditions have been initiated.

3. Summary

The experiments on a laboratory scale and in the semi-technical plant have releaved a considerable development potential for fluidized bed plants (7) as well as the fact that tests on a smaller scale may sizeable contribute to this end so that results from the different plants are appreciated as being complementary to each other.

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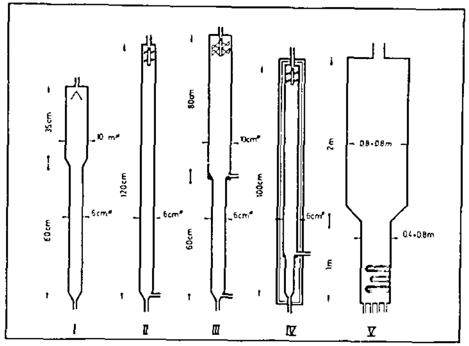

Figure 1: Shapes of the fluidized bed reactors used

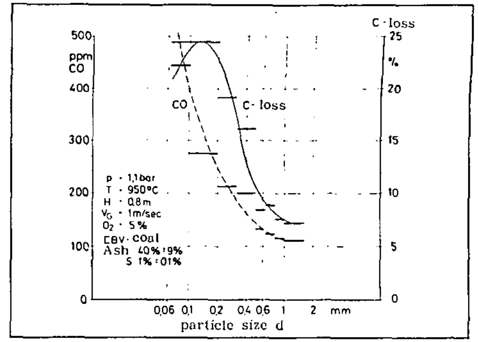

Figure 3: C-loss and CO emission as a function of grain size

Parameter Range (Standard)	$\frac{MW}{m^2}$	%C-Loss	$\frac{gCO}{kWh}$	$\frac{gSO_2}{kWh}$	$\frac{gNO_2}{kWh}$
Pressure 1 → 4,5 bar (1,1 bar)	↗	↘	↘	↘	↘
Temperature 750 → 950°C (850°C)	→	↘	↘	↗	↗
Excess air in the 0,5 → 12% O_2 flue gas (5%)	↘	↘	→	→	↗
Fluidizing velocity 0,3 → 1,5 m/sec (1m/sec)	↗	↗	→	↗	↘
Bed height (fluidized) 0,2 → 1,2 m (0,8m)	→	↘	↘	↘	→
Secondary air 0 → 66% (0)	→	↘	↘	↘	→
Ca/S 0 → 3 mol/mol (1)	→	→	→	↘	→
Particle size of coal 0 → 2 mm (0,1-1mm)	→	↘	↘	→	→
Particle size of limestone 0 → 2 mm (0-0,1mm)	→	→	→	↗	→

Figure 2: Variation of operational parameters in fluidized bed reactors on laboratory scale

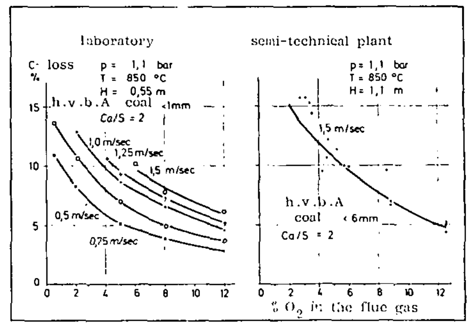

Figure 4: Carbon loss as a function of excess air

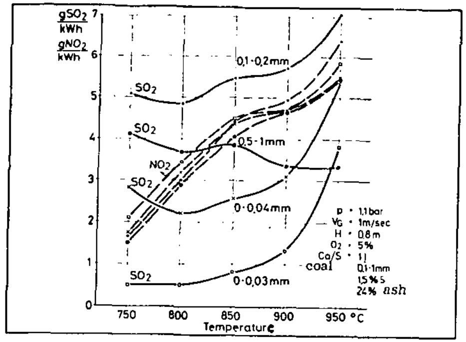

Figure 5: SO_2 and NO_x emissions for different sorbent sizes of cretaceous limestone as a function of temperature

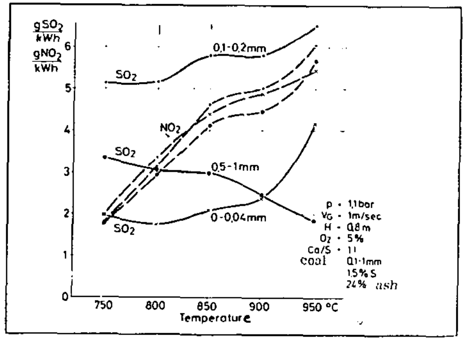

Figure 6: SO_2 and NO_x emissions for various sorbent sizes of dolomite as a function of temperature

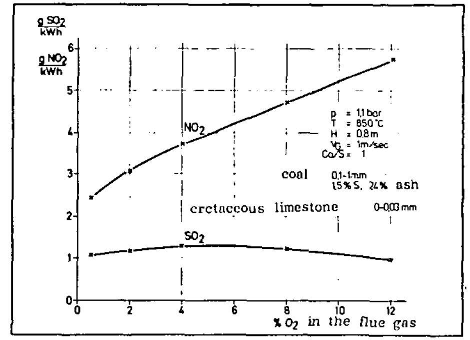

Figure 7: SO_2 and NO_x emissions as a function of excess air

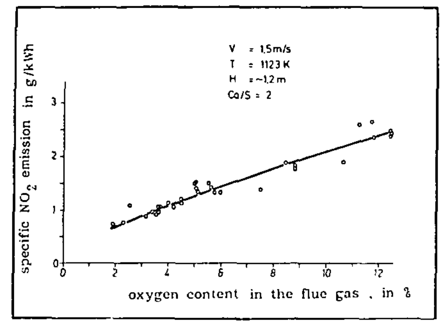

Figure 8: Specific NO_2 emission from the semi-technical fluidized bed plant

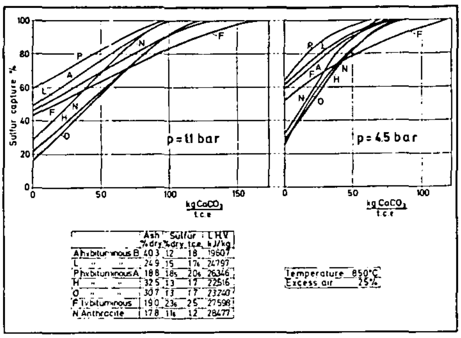

Figure 9: Sulphur capture as a function of the amount of limestone added per t.c.e.

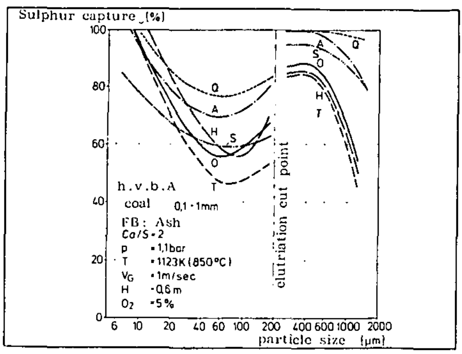

Figure 10: Sulphur capture as a function of limestone particle size