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REGENERATIVE THERMAL OXIDIZERS BASED ON THE PEBBLE-HEATER TECHNOLOGY

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ABSTRACT

Regenerative thermal oxidizers are used for cleaning the exhaust gas of different industrial furnaces. They are more and more demanded as the environmental regulations become more strict. On the other hand, there is a steady effort to develop systems with lower investment and especially operational costs.

The new Pebble-Heater technology with annular beds and radial fluid flows has been used as a basis for developing a system of a regenerative thermal oxidizer which can meet the market requirements. Theoretical studies and numerical simulations have shown that an extremely effective heat transfer at $P\acute{e}clet$ numbers around Pe=30 in granular beds of natural materials/minerals (like quartz-gravels, volcanic stones or fire-clay) is possible. Temperature gradients of more than 20 K/cm have been realized, so that the required bed thickness may be as low as 35 cm. That also results in a lower pressure drop. Those studies have shown that under optimized conditions the natural materials/minerals are much more effective than artificial forms (ceramic rings, saddles, honeycombs...) developed especially for heat regenerators. The price ratio between the two is more than 1:100.

The realized facilities have justified the expectations. A heat recovery degree of more than 98%(!) has been achieved, with a total pressure drop of about 18 mbar. The next best system on the market has at least a 100% higher energy (gas/oil) consumption and a 50% higher fan power consumption. Some other characteristics of the new system have also shown important improvements for the process. The bigger volume of the hot chamber (reaction zone at 800°C) results in a longer residence time (1,5 - 1,7 seconds), so that the destruction of different pollutants (aldehyds, benzols, phenols...) and carbon-monoxide is more effective. The measured emission values are far beyond the limits set by the regulation authorities. The realized facilities have capacities in the range of 15.000 - 45.000 m³_{STP}/h. The next development stages are a scale-down (1.000 - 5.000 m³_{STP}/h) and a scale-up (over 100.000 m³_{STP}/h) of the existing technology. It is intended to substitute some very expensive catalytic facilities with the new system. With the extremely high heat recovery degree, the reaction zones at 800°C - 1000°C do not cause high operational costs any more. Lowering the reaction temperature (the main advantage of the catalytic facilities) is no longer a decisive parameter for the facility design.

KEYWORDS

Thermal oxidizers, off-gas cleaning, VOC destruction, heat recovery, regenerators, emission control, pebble-heater

INTRODUCTION

Exhaust gases from various industrial furnaces often contain a high concentration of carbon monoxide and volatile organic compounds (VOC). This is usually the case at different drying furnaces (e.g. drying of wooden chipboards) or at furnaces with zones for preheating and drying of input material with combustion gases in counter-current flow (e.g. tunnel kilns for ceramic or bricks). The first approach was to install an additional chamber for afterburning the fumes and transforming pollutants into CO₂ and H₂O. That solution is not very expensive in investments, but requires large amounts of additional energy (natural gas or liquid fuel). That is only a reasonable solution in cases of very high VOC concentrations and with good opportunities for the recovery of waste energy. The regulations in Germany, and more recently in Europe, too, are becoming more and more strict. In many cases the emissions are limited to 50 mg/m³_{STP} of CO, 50 mg/m³_{STP} of total carbon and just a total of 20 mg/m³_{STP} of benzoles, aldehyds and phenols. On one hand, the traditional approach makes it difficult to stay within these limits, on the other hand the gases with steadily lower concentration of pollutants have to be treated nowadays. In such cases, the required energy consumption is too high and the whole treatment too expensive.

A more contemporary solution is to use a catalytic thermal oxidizer with incorporated regenerator or recuperator. The oxidation takes place at a lower temperature (450°C - 550°C). Together with the energy recovery (preheating the inlet raw gas with outlet clean gas), that results in much lower energy consumption. However, the investment costs are too high, mostly due to high specific costs of catalysts. In many cases the catalysts have to be periodically exchanged, due to deactivation with sulphur, chlorine, silicon or/and dust.

There is a constant need for an off-gas cleaning facility which can fulfill the given emission restrictions, but with lower investment and especially operation costs. Those were the prerequisites and motivations to develop a new regenerative thermal oxidizer based on the Pebble-Heater technology.

PEBBLE-HEATER (PH) – WHAT'S THAT?

"Pebble-Heater" is a common name for regenerators filled with bulk material mostly of a spherical shape (pebbles). The state-of-the-art design has a vertical column of pebbles through which gas flows axially. The problems connected with such designs (such as channeling, wall heat losses, pressure drop, scaling problems and inhomogeneous temperature field) are also well known in technical practice.

At ATZ-EVUS a new concept of the Pebble-Heater has been developed. The main difference lies in the flow direction: gas flows radially through the pebble-bed, which is fixed between two coaxial cylindrical grids. The inner grid, the so-called hot-grid, is made of porous ceramic bricks. The outer grid, referred to as cold grid, is made of a gas permeable steel construction (e.g. perforated steel plate). All other extraordinary characteristics result from that at first sight small difference. Higher flow velocity and/or smaller pebble diameters may be used, as there is no danger of fluidization. That provides a very high specific surface (or surface to volume ratio) and consequently an excellent heat transfer. That results in a high thermal efficiency (units with more than 98% are in operation) and a temperature gradient in the range of 1500 – 2000 K/m. The pebble-bed does not have to be thick in radial direction, so that the pressure drop is also low. In the end, that leads to a very compact unit at low investment costs. That

new technology has been developed primarily to substitute the technology of hot wind stoves (Cowpers) for supplying blast stoves with hot blast (*Brotzmann & Stevanovic*, 1998).

DESCRIPTION OF NEW FACILITY

The new facility of a regenerative thermal oxidizer differs from the state of the art (the so-called 3 chamber system) already from the outside view: instead of three square or cylindrical regenerators placed in a row, they are built on top of each other in a common cylindrical shell. The facility again contains three regenerators (two main regenerators and a third auxiliary one in between) with a common central combustion chamber (or reaction zone). Such a design allows for a low pressure drop, a compact form and thus a small occupation area for the installation.

Another important point is the wall heat loss. Inside the combustion chamber the temperature is up to 900 °C. To minimize the wall losses, the previous facilities (3 chamber systems) had to be equipped with a high quality fibre insulator. Despite this insulation the radiation losses could not be entirely avoided. Due to the design of the new system in form of a hollow cylinder with a centrally located combustion chamber (high temperature zone), the pebble-bed around the combustion chamber acts as peripheral insulation. At the outer shell the same temperature prevails as in the raw gas, so that a normal external insulation is sufficient.

The operation of the new facility may be described in four phases (see *Figure 1*). During phase 1 raw gas (crude gas) enters the upper regenerator (1). By flowing through the pebble-bed the raw gas is heated almost up to the reaction temperature (between 750 °C – 900 °C, usually 800 °C). In the broadly designed combustion chamber (4) the residence time is long enough and the temperature field is very homogeneous, so that all pollutants are sufficiently thermally oxidized. The clean gas flows through the lower regenerator (2), transfers its heat to the pebble-bed and leaves the facility. With a fan (6) it is transported through the chimney (7) into the atmosphere. In spite of the extremely high thermal efficiency (i.e. regeneration rate) of over 98% it may become necessary to switch on the auxiliary burner (5) when the concentration of VOCs in the raw gas is too low to compensate for the off-gas losses of some 15-30 K.

As the amount of heat stored in the upper regenerator (1) is decreasing and the one in the lower regenerator (2) is increasing, the flow direction of the two gas streams has to be changed periodically, e.g. every 5 minutes. Therefore, in the next phase the raw gas flows through the lower regenerator and takes heat from the pebble-bed. In order to avoid that during the switching a certain amount of raw gas may get into the clean gas, the upper regenerator is first purged with recirculated clean gas in phase 2. During this purging (lasting some 10 seconds) the clean gas together with the purging stream is led through the small intermediate regenerator (3). As this phase is essentially shorter than the phases of a normal operation (phase 1 and 3), the amount of heat stored in the intermediate regenerator is respectively much smaller. Subsequently, in phase 3 the clean gas flows through the upper regenerator (1), which is therefore heated up. After a repeated short purging in phase 4, phase 1 starts again. The small intermediate regenerator (patented by $Fa\beta binder$, 1998), which is periodically heated up during the purging phases, is cooled by counter-purging with a small amount of cooling air during the phases of normal operation (1 and 3).

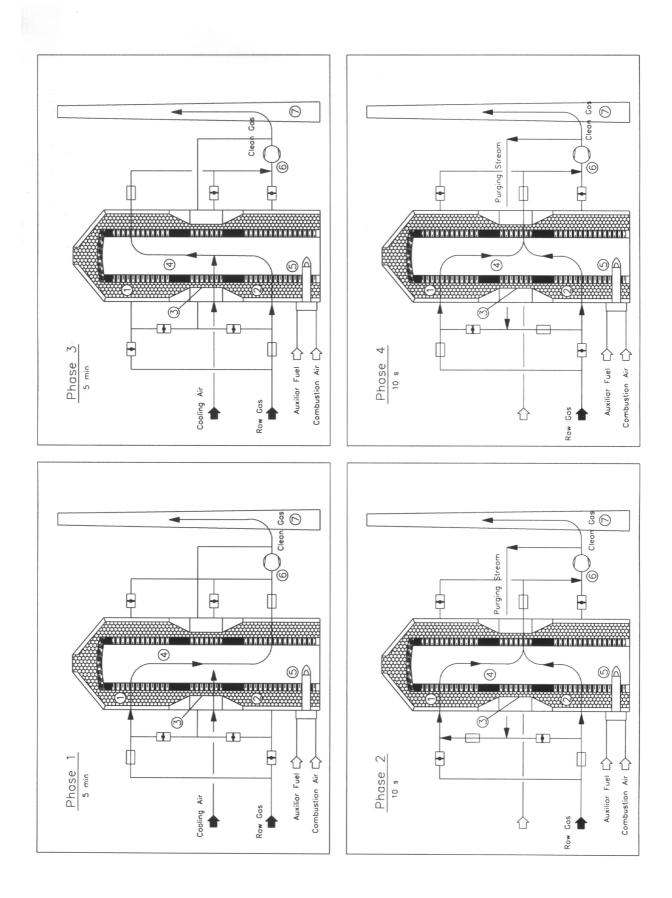


Figure 1: Operation of regenerative thermal oxidizer based on PH-Technology

MATHEMATICAL MODELLING

The mathematical model and respective numerical code have been developed for the simulation of a pebble-heater operation. With some small changes they are used nowadays for design optimization of newly developed thermal oxidizers.

The physical model of the pebble-heater consists of a combustion chamber in the center and a hollow cylinder filled with pebbles around it. The heater is axial-symmetric and the upper and bottom walls are adiabatic, so the heat is transferred just in radial direction. Due to a very high specific surface available for the heat transfer (usually between 500 - 1000 m²/m³, depending on the bulk material), the temperature difference between gas and solid phase is very low, i.e. negligible compared to the whole temperature change. In such cases the so-called "homogeneous" model for the pebble-bed may be used (*Vortmeyer & Schäfer, 1974*). Respectively, just one energy balance equation may be used. The original equation of *Vortmeyer & Schäfer* was derived for a cylindrical pebble-bed with axial flow. It had to be rewritten for the radial geometry in the form:

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\Lambda_{r}\frac{\partial T}{\partial r}\right) = \frac{\partial}{\partial r}\left(m_{o}c_{Pf}T\right) + \left[\left(1 - \psi\right)\rho_{s}c_{s} + \psi\rho_{f}c_{Pf}\right]\frac{\partial T}{\partial t}$$
(1)

with the following notation:

r - radial coordinate

t - time

 Λ_r - "effective" heat conductivity

T - temperature

 m_o - mass flux of gas

 ψ - void fraction (i.e. bed porosity)

 c_s , c_{Pf} - specific heat of solid and gas phase

 ρ_s , ρ_f - density of solid and gas phase.

The "effective" heat conductivity, as well as all other physical properties, is a function of the temperature and thus of the radial coordinate (temperature strongly depends on radial position). The mass flux of gas is also a function of the radial position, due to a change in the flow cross section. The "effective" heat conductivity in radial direction (Λ_r) encounters the heat conductivity of fluid and solid, the convective heat transfer between fluid and solid, as well as the effect of radiation between solid particles (pebbles). In the literature there are several correlations for those terms. We use the correlations given in *Bauer*, 1988 and *Till*, 1981, which are valid up to a *Péclet* number of Pe = 30. For some very specific applications there is a need for a correlation in the region of Pe > 30. As it is not available in the literature, an experimental facility for its evaluation will be erected in the near future.

For solving the partial differential equation of second order (1), it is necessary to know one initial and two boundary conditions. Some known temperature distribution over the radius has to be given as initial condition. Heat fluxes on the hot and cold end (i.e. hot and cold grid) are the required boundary conditions.

The partial differential equation (1) can not be solved analytically, first of all because the "effective" heat conductivity and gas flux are not constant. That type of equation (basically *Fourier*-equation) has been effectively solved (*Stevanovic & Studovic*, 1981) using the Crank-Nicolson numerical method. Its main advantage is impliciticy, i.e. it enables pretty high time

steps without disturbing the calculation stability. A numerical code based on that method has been developed for simulating the pebble-heater operation. A typical result of the code with the characteristic temperature profile through the pebble-bed is given in *Figure 2*.

Simultaneously with solving the energy equation (1), the numerical code calculates the pressure drop through the pebble-bed by integrating the equation:

$$\frac{dp}{dr} = \frac{1}{\psi^2} \mu \xi \frac{\rho w_o^2}{2 D_e} \tag{2}$$

with newly introduced parameters:

p - pressure

 $\mu\xi$ - friction and path factor

 w_o - gas velocity

 D_e - equivalent pebble diameter.

For the friction and path factor $\mu\xi$ the famous Ergun equation (*Ergun*, 1952) may be used. We found that the correlation of *Kast*, 1984 gives better results. However, the best results were achieved with our own correlations based on our own measurements. That was especially the case when the shape of the pebbles deviates from ideal balls.

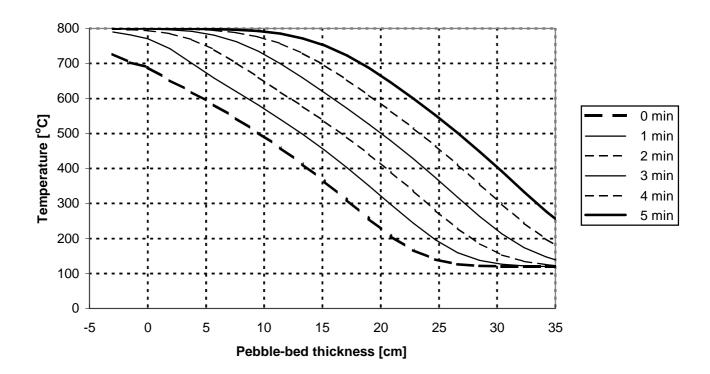


Figure 2: Typical temperature distribution inside the pebble-heater

DESIGN OPTIMIZATION

Just with some minor changes, the same code is nowadays used for the optimization of newly developed thermal oxidizers. First of all, the calculations have shown that an excellent heat transfer, together with a low pressure drop may be achieved with some natural bulk materials / minerals like quartz-gravels, crude fire-clay or volcanic stones (Eifel-Lava). The results are as good, or even better, as with artificial ceramic materials like honeycombs, saddles, rings, etc. The price ratio between the two is in the range of 1:100. That is an important factor for the reduction of investment costs.

The code is also used to minimize the operational costs. Two important parameters are the efficiency of the heat recovery and the pressure drop. The efficiency of the heat recovery usually is defined in practice (with simplifying assumption that the specific heat (c_p) of gas is always constant) as:

$$\eta = \frac{T_{cc} - T_{clean}}{T_{cc} - T_{row}} \tag{3}$$

with the following notation:

 η - heat recovery efficiency

 T_{cc} - temperature of combustion chamber (i.e. reaction zone)

 T_{clean} - temperature of cleansed gas T_{raw} - temperature of raw gas.

Higher efficiency means lower temperature difference $\Delta T = T_{clean}$ - T_{raw} and thus lower heat losses and energy consumption. It is well known that intensifying the heat transfer by choosing smaller pebble diameters (higher specific surface) or higher bed thickness will decrease that ΔT and the fuel consumption, but will increase the pressure drop and the power consumption (for a circulation fan). The code makes it possible to choose an optimized set of parameters like: bed thickness, bed height, hot grid diameter and available pebble diameter with the aim to reach the point with the lowest operational costs. That point is not constant, but depends on local energy prices and pollutant concentrations. The best designs optimized that way have an heat recovery efficiency of 98% and a pressure drop of 18 mbar through the whole facility. Those values have been verified in practice. They are much better than the values of similar facilities, which use more expensive ceramic elements specially developed for heat storage applications.

REDUCTION OF EMISSIONS

Eight facilities of this new kind are already in operation, with a capacity range from 15.000 m³_{STP}/h to 45.000 m³_{STP}/h. Depending on the pollutant concentration, some of these facilities are always in autothermal operation (i.e. without additional energy comsumption), some are never in autothermal operation, the others constantly change between those two operation modes.

The bigger volume of the hot combustion chamber results in a longer residence time (1,5 - 1,7 seconds). Together with the very homogeneous temperature field in the hot chamber (absence of cold walls), this results in a more effective destruction of different pollutants. With a combustion chamber temperature between 750°C and 800°C and independent of the raw gas

concentration all facilities easily meet the limits imposed by law. *Table 1* shows an extract from the official measurements at a facility which is mostly not in autothermal operation. On the other hand, the following graph (*Figure 3*) presents the pollutant concentrations in the raw and clean gas at a facility which is running steadily in autothermal operation.

Table 1: Emission measurements in a brick factory (after a thermal oxidizer for 30.000 m³_{STP}/h)

	Measurements			Emission Limitation by 'TA Luft'	
Substance	Mass Concentration c _n (mg/m ³)		Flow Rate (g/h)	c_n	fulfilled
	Range	Mean Value	Mean Value	(mg/m^3)	(yes/no)
Anorg. Fluorine	1,06 - 1,37	1,27	21,4	5	yes
Sulfur Oxides *)	9,3 - 12,1	10,2	173	500	yes
Nitrogen Oxides *)	25,3 - 30,4	27,8	1020	500	yes
Dust *)	0,3 - 0,5	0,4	14,3	5	yes
Total Carbon	3,5 - 6,7	4,8	80,7	50	yes
Benzole	0,20 - 0,28	0,23	3,9	20	
Aldehyde C ₁ -C ₃	1,86 - 2,91	2,22	37,5	in Sum	yes
Phenoles	0,01 - 0,02	0,01	0,26		

^{*)} related to 18 vol-% Oxygene in the off-gas

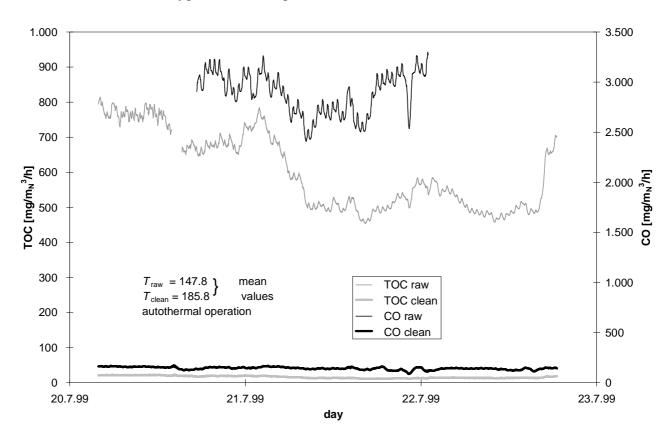


Figure 3: Regenerative thermal oxidizer for 35000 m_N^3/h - Gas Analysis from 20./23.07.1999

RESULTING ADVANTAGES

Compared to the state of the art, the newly developed regenerative thermal oxidizers have the following advantages:

Operation costs

Due to a very high thermal efficiency (over 98% realised), the energy consumption at low VOC concentrations is several times lower than in other systems. Comparing it with the best efficiency of 96% that is achievable with today's state of the art, the new facility has an energy consumption which is at least twice as low. At higher VOC concentrations our facility can easier reach an autothermal operation (i.e. without additional energy consumption). Last but not least, the power consumption for the fan is drastically reduced due to the lower pressure drop: the best regenerative system requires about 50% more power, a catalytic facility even 4 times more! In the case of a 30.000 m³_{STP}/h facility this leads to saving a minimum of 30.000 DM/year for natural gas and between 18.000 DM/year and 108.000 DM/year for power (for common local price conditions in Germany).

Investment costs

In the new facility natural materials like quartz gravel, crude fireclay or volcanic stones (Eifel-Lava) are used as heat storage mass. With their price ranging between 20 and 200 DM/t, they are very attractive compared to the high quality ceramic structures, like honeycombs or saddles, which cost around 3.000 DM/t and even more after being coated with a catalyst. That results in a reduction of the investment costs between 10% and 40%, depending on the capacity and application conditions.

Operation and maintenance

The newly developed facility is very flexible in operation: a change in VOC concentration doesn't provoke any additional problems. If necessary, the reaction temperature of usually 800 °C can be easily increased to 850 °C or 900 °C, resulting in even lower emissions. The facility needs almost no maintenance. A dust loaded raw gas is also acceptable, the facility acts in a certain way as a dust filter. At higher dust loads it is necessary to recirculate the heat storage mass periodically (e.g. once a year). That way the formed "filter-cake" is destroyed and an increase in pressure drop avoided.

Space requirement

Due to the placement of three beds on top of each other in a tower-like design, the floor space requirement is much smaller than for other facilities. For capacities of up to 60.000 m³_{STP}/h, the outer shell is about 4m in diameter (of course, some additional space is required for a fan). That provides special advantages in case of reconstruction and re-equipment of an existing production plant.

FURTHER DEVELOPMENT

The next development stages are a scale-down $(1.000 - 5.000 \text{ m}^3\text{STP})$ and a scale-up (over $100.000 \text{ m}^3\text{STP}$) of the existing technology. It is intended to substitute some of the very expensive catalytic facilities with the new system. With the extremely high heat recovery efficiency, the reaction zones at 800°C - 1000°C do not cause high operational costs any more. Thus, the lowering of the reaction temperature (the main advantage of the catalytic facilities) is not a decisive design parameter any more!

The newly developed facility has some characteristics which can be very useful for the destruction (not just removal) of dioxins and furans. Besides a high temperature in the reaction zone, a sufficiently long residence time in the reaction zone and a homogeneous temperature field, fast cooling (quenching) of the cleansed gas through the pebble-bed is especially important. The residence time in the critical temperature range from 250°C to 400°C is only around 50 ms! Thus, the so-called "de-nuovo-synthesis" (i.e. recombination) of dioxins and furans, which is responsible for the emissions most of the time, could be efficiently surpressed. It is intended to prove those advantages in practice in the near future.

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