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Effects of deposition and fouling on thermal behaviour of glass furnace regenerators

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Samenvatting

Een mathematisch model is ontwikkeld om de afname in het thermisch rendement van glas-ovenregeneratoren te voorspellen als gevolg van vervuiling door rookgascondensaten. Het model bestaat uit vier deelmodellen voor de:
1. beschrijving van het thermisch gedrag van de regeneratorpakking;
2. beschrijving van het chemisch gedrag en condensatie van afgassen in de regenerator;
3. globale beschrijving van de warmte-over-dracht van de vlammen in de glasoven;
4. bepaling van de verdamping van zwavel-, chloor- en natriumverbindingen.

De afname van de warmte-overdracht in de rege-nerator en dus de afname in het thermisch rende-ment van de oven is voorspeld:
- voor verschillende regeneratorstapelingen;
- als functie van de productiesnelheid;
- als functie van de glassmelttemperatuur;
- als functie van het schervenpercentage.

Afhankelijk van de verschillende procescondi-ties en pakkingen is volgens berekeningen maar ook volgens de praktijk een toename in het ener-gieverbruik van enkele procenten per jaar te ver-wachten [1] veroorzaakt door regeneratorvervui-ling. De zogenaamde cruciform en chimney block pakkingen zijn minder gevoelig voor ver-vuiling dan basketweave of pigeon-hole stape-lingen. Als de glastemperaturen stijgen zal zo-wel het stofgehalte in de rookgassen als de ver-vuiling van regeneratoren snel toenemen. Hogere schervenpercentages geven volgens het model minder snelle afnamen van het regenera-torrendement.

Summary

A model has been developed to predict the de-crease of the thermal performance of glass furna-ce regenerators due to fouling by flue gas con-densates. The model consists of four parts: ·
1. a description of the thermal performance (heat transfer) of regenerator checkers;
2. a description of chemical reactions in and deposition from flue gases in the regenera-tor;
3. a description of the heat transfer in the fur-nace combustion chamber;
4. determination of volatilisation of sulfur, chloride and sodium components from the melt.

The aging and the reduction of the thermal effi-ciency due to fouling has been predicted:
- for different checker work constructions;
- as a function of the pull rate;
- as a function of glass melt temperatures;
- as a function of the applied cullet fraction in the batch.

Depending on the different conditions, the pre-dicted increase in energy consumption is a few percents per year [1], mainly due to fouling. Cru-ciform and chimney block checkers seem to be less sensitive for this fouling than pigeonhole and basketweave packings. As glass melt tempe-ratures increase, dust emissions and fouling rates are going up. The model is in quite good agree-ment with practical observations in industrial furnaces.

1. Introduction

Furnace lifetimes have been considerably increas-ed over the last fifty years. Improved refractory materials and optimized process control contribu-ted to a decrease in corrosion rates. However, du-ring the 6 up to 10 years furnace lifetimes, the specific energy consumption generally increases from 5 till more than 15 %. This is not only caused by increasing air leakages, attack of refractory ma-terials and insulation but mainly by the fouling of regenerators by condensation products from the flue gases.

For soda lime glass furnaces, these condensates contain for 80-95 % sodium sulfates. Sodium com-ponents vaporize from the molten glass mainly by reaction with water vapor in the furnace atmos-phere, leading to the formation of volatile sodium hydroxides.

Sulfur dioxide formed by the sulfur in the fuel or by decomposition of sulfates in the melt, reacts with sodium hydroxide at temperatures below 1100-1200°C. This leads to condensation of sodium sulfates in the regenerator packings.

The sodium sulfate condensation takes place at the surface of the colder regenerator bricks. This cau-ses deposition of liquid sodium sulfate above 884°C and fouling by crystalline material below this temperature. In the lower and cooler regenera-tor sections, sodium sulfate dust will be formed, this dust partly sticks at the refractory surfaces.

The deposition products have a lower thermal conductivity than the refractories: the heat transfer from flue gasses to the bricks will be reduced by these insulating deposition layers. After the rever-sal in the combustion cycle, the heat release by the preheated packings to the air is also limited by these salt layers.

Apart from the reduction of the heat conductivity by the deposition process, sodium sulfate may cause severe damages by corrosion or mechanical attack during sublimation processes.

The deposition rates depend very much on the concentrations of the sodium compounds and sul-fur dioxide in the flue gases. The volatilisation from the batch blanket and the molten glass, deter-mines these concentration levels. The flue gas tem-peratures and concentrations of volatilisation pro-ducts at the entrance of the regenerator chamber are very important for the fouling and thermal

behaviour of regenerators during the furnace life-time. The flue gas composition and temperatures are governed by the processes in the furnace: combustion, radiative heat transfer and volatilisation from the batch blanket and the melt.

In the next paragraphs four sub models for thermal behaviour of regenerators, deposition, heat transfer in the furnace and volatilisation will be briefly presented. Examples of calculation results for several process conditions and different regenerator checkerworks will be given later.

2. The regenerator heat transfer model

The modelling of the thermal performance of new regenerators has been developed by Barklage-Hilgefort [2]. His method has been based on a numerical difference method in which the elementary heat transfer processes in the regenerator checker works are mathematically simulated.

Figure 1 shows a regenerator. The calculation process acts as follows: The regenerator packing is divided in a certain number of layers, starting from the flue gas entrance in the direction of these flue gases. The total heat transfering surface area of the checkers and the heat capacity depends on the structure and dimensions of the packing and the refractory materials which have been used. The checker work may consists of different sections with different packing arrangements.

First a certain not necessarily realistic temperature distribution for the checker work is assumed. The temperature decrease of the flue gas flowing through the regeneator will be calculated per layer and per short time period Δt. The increase of the temperature of the refractories will be calculated also after this Δt. This procedure will be repeated for the next Δt period and the temperature decrease or increase during this time step is added. After

one half of the regenerator cycle period, in which the regenerator is heated by the flue gases, these calculations will be continued, now for cold air entering the regenerator at the former flue gas exit. The air will be preheated and the refractory materials will be partly cooled by this process. After this air preheat half cycle, the calculation continues again for the situation of the regenerator be heated up by flue gases. The calculation process is carried out until the exit temperatures at a certain time during the half cycles of the flue gases and air remain constant. The most important steps in this calculation procedure are the heat transfer from flue gas to the refractory materials but most of all the heat transfer to the air [3].

At a certain moment and a certain position (layer) the heat flux is given by:

$$Q_s = \alpha \cdot (T_{fi}(t) - T_{rsi}(t)) \tag{1}$$

with: α = the heat transfer coefficient (including radiative, conductive and convective heat transfer)

$T_{fi}(t)$ = flue gas or air temperature in layer i at time t
$T_{rsi}(t)$ = refractory surface temperature layer i at time t.

According to Hausen [4], $T_{rsi}(t)$ can be approximated as a function of time and the temperature of the centre of the refractory brick and Q_s can be calculated as:

$$Q_s = \alpha \cdot (T_{fi}(t) - T_{rmi}(t)) / (1 - \gamma(t)\alpha.) \tag{2}$$

with $T_{rmi}(t)$ = time dependent temperature in the centre of the brick

The function $\gamma(t)$ depends on the dimensions of the refractory materials and the heat capacity and conductivity of this material. This function can be calculated separately for every time and per layer using Hausen's approximations.

By this procedure the heat transfer can be calculated very accurately and fast. The accuracy can be improved by using not only the temperature of the gas and refractory at the beginning of timestep Δt and the beginning of layer i, but by using the average values in the layer and during this timestep. The convective heat transfer coefficients are derived using simple Nusselt number relations.

$$Nu = A + B \cdot Re \tag{3}$$

with Re = Reynolds number for the gas flow
A and B are coefficients depending on kind of checker work and emperically derived "A" numbers vary between 7 and 18, "B" numbers vary between 0.006 and 0.012.

From these Nusselt numbers the convective heat transfer can be determined. The heat transfer coefficient is corrected for radiative contributions in the very hot regions, depending on water vapor and carbon dioxide concentrations in the flue gases.

3. The combustion chamber model

The regenerator heat transfer model needs some input parameters which depend on the combustion process in the furnace and the heat transfer to the superstructure and the molten glass.

The flue gas composition and time depending flue gas temperature are used by the regenerator model. The flue gas temperature depends on the air preheat temperature, determined by the regenerator model and the radiative heat transfer from the flames to the superstructure and to the surface of the molten glass. Figure 2. presents the most important processes.

Therefore a relatively simple combustion chamber model has been developed. The heat transfer in the furnace combustion chamber by convection is only a few percent from the radiative heat transfer and is neglected in this submodel.
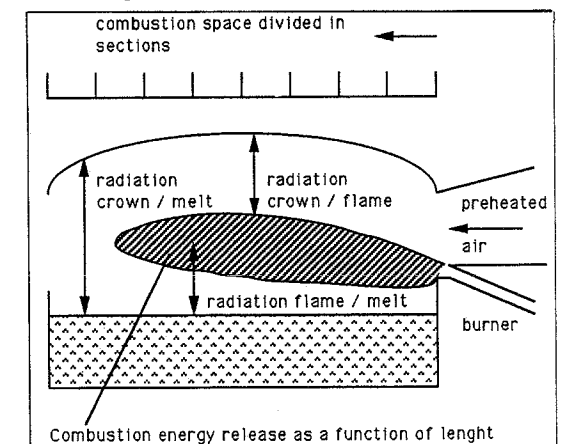
Figure 1. >
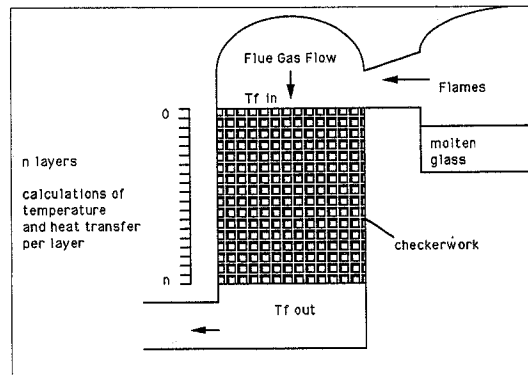*Scheme of a regenerator divided in layers for the calculation procedure*

Figure 2. >>
Combustion and radiation in combustion chamber of glass furnace

Schematically the calculation process exists of:
a. Division of the combustion chamber along the combustion path in ten or twenty segments. Initially, a first approach for the temperature profiles of the superstructure and the flame have to be assumed;
b. A certain empirical function is assumed for the burning process, which gives the amount of combustion energy generated in each segment;
c. Per segment a thermal balance is calculated: the heat input by the gas flow entering the segment + the combustion energy are used for the radiation to the superstructure and the molten glass surface and for the flue gas heat contents leaving the segment.

The radiative emission considers the radiation from flame to glass and reverse, from flame to superstructure and reverse and from superstructure to glass and reverse.

The calculation has to be carried out iteratively: temperatures of the superstructure and the flame (layer of gas) are adjusted until these values remain constant and until the heat balances per segment are correct.

The molten glass surface temperature will be assumed to have a certain known value.

The emissivity coefficient is calculated as a function of gas layer or flame thickness, flame temperature and flame composition [5]. Absorption coefficients for glass and refractories are dependent on material properties.

The radiation is assumed as to be grey. In reality the radiation depends very much on the wavelength. Carbon dioxide and water vapors radiate at certain spectral bands.

In addition to the energy transfer to the superstructure, the molten glass and batch blanket, some extra energy losses have to be considered, like leakages of the furnace, radiation through these leakages and the forced cooling of the metal line by air. In the model constant values for these losses have to be assumed.

The combustion chamber model and the regenerator heat transfer model need each other: the air preheat temperature and air flow rate calculated by the regenerator heat transfer model are necessary for the combustion chamber model. On the other hand the temperatures, the composition and the volumetric flows of the gases leaving the furnace are needed for the regenerator model.

4. The volatilisation model

The fouling of a regenerator depends on the condensation of volatilisation products originating from the melt or batch blanket. For soda lime glasses, sodium components vaporize partly from the batch blanket but mainly from the molten glass surface.

These volatile sodium components react with sulfur oxides during the cooling process, leading to sodium sulfate formation below about $1250^{\circ}C$. This sodium sulfate gas will condensate below approximately $1100^{\circ}C$: this results in dust formation or salt deposition.

The deposition of sodium sulfate is the process which determines the fouling of regenerators and therefore the reduction of the thermal efficiency of older regenerators: the aging process.

Important for the deposition process are mainly the concentrations of sodium compounds in the flue gases but also the sulfur oxide concentrations and to some extent the chloride concentrations [6,7].

4.1. The sodium volatilisation

Sodium components are entrained by the combustion gases by:
— carry over of light soda particles;
— sodium chloride volatilisation from the batch blanket, sodium chloride is an impurity in synthetic soda;
— sodium oxide reactions at the melt surface with the water vapor, leading to formation of volatile sodium hydroxides;
— sodium sulfate volatilisation from the molten glass into gas bubbles, during the primary fining process.

The model takes into account:
a. the carry over dependent on the density of the soda used, carry over from light soda is assumed to be twice the carry over from dense soda;
b. the fraction of cullet, soda carry over is given as mg carry over per kg of glass molten from primary raw materials.

The sodium chloride volatilisation is assumed to be proportional to the soda concentration in the batch and the concentration of the sodium chloride impurities in the synthetic soda.

Most important for the sodium volatilisation appears to be the sodium hydroxide formation.
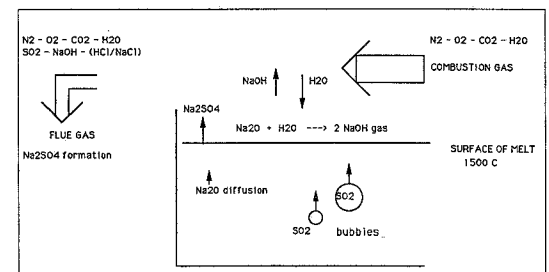
Conradt's model [8] has been used to describe this volatilisation process.

Sodium sulfate volatilisation is of minor importance, calculations using Conradt's model for vaporisation at the molten glass surface and estimations for sodium sulfate diffusion into bubbles show that sodium sulfate vaporisation contributes for only 1 % or less to the sodium entrainment in the combustion gases.

4.2 Sulfur oxides in flue gases

The main sources for sulfur are the fuel, sulfate additions in the batch, sulfur impurities in the raw materials and sulfur in the cullet.

In the model all the sulfur from the fuel is assumed to be converted into sulfur dioxide gas.

The total sulfur retention in the glass depends very much on the final glass colour [9].

In the model it has been assumed that flint or float glass contains 0.05 till 0.07 weight-% sulfur, green or emerald green glass has a concentration of about 0.04 weight-% sulfur and in light amber glass about 0.02 % has been dissolved. In dark amber glass the concentration of sulfur can reach values above 0.07 weight-%.

The difference between the total sulfur input by fuel, sulfates and sulfur in the cullet minus the sulfur output by the glass has been assumed to be released as SO_2, which will be emitted by the flue gases.

4.3. Chlorides in flue gases

The main chloride sources are synthetic soda and cullet. The chloride impurities in the soda appear to be much more volatile than in the glass. In this model only the chloride vaporization from the soda in the batch blanket has been considered. The

chloride-volatilisation per kg of glass has been assumed to be proportional to the soda content in the batch and the chloride concentration in the soda. The volatilisation or formation of gaseous sodium, sulfur and chloride compounds lead to concentrations in the flue gases entering the regenerator chambers.

5. Deposition of sodium sulfate in the regenerator checkerworks

The deposition by condensation at the relatively cool surfaces in the checkers has been described by reference [7].

During the cooling of the flue gases from about 1400°C down to 500°C in the regenerator, chemical reactions between flue gas components take place. The chemistry depends very much on the composition of the flue gases: for furnaces producing sulfate refined soda lime glasses, the formation of sodium sulfate from sodium chloride, sodium hydroxides and sulfur oxides causes condensation of this salt.

Condensation reactions occur at the surfaces of the checker bricks: deposition rates are determined by the diffusion of sodium and sulfur compounds from the flue gases through a diffusion boundary layer towards the brick surfaces.

The calculation process consists of:

a. calculation of the thermodynamic equilibrium composition of the flue gases at the entrance of the regenerator;
b. calculation of equilibrium flue gas composition in the first layer (top layer of regenerator);
c. calculation of equilibrium flue gas composition at positions directly in contact with the surface of the regenerator bricks assuming a certain total sodium, sulfur and chloride concentration at these locations;
d. calculation of the diffusion of all sodium, sulfur and chloride compounds through the flue gas boundary layer at the brick surfaces, using diffusivities, concentration differences and Sherwood numbers for mass transfer;
e. checking the restrictions that:
    for no condensation: the net diffusivity rates of all sodium, all sulfur and all chloride components have to be zero;
    for sodium sulfate deposition: the total net diffusion of all sodium components have to be twice the net diffusion of all sulfur components, because by sodium sulfate deposition for two atoms of sodium, one atom of sulfur are used, the net diffusion of all chlorides has to be zero.

The total concentration of sodium, sulfur and chloride in the flue gases at the refractory boundary will be adjusted and the calculation continues from point d.

f. After this iteration process, the deposition of sodium sulfate in this regenerator layer is calculated from the sodium and sulfur mass transfer rates.
e. The total amount of sodium and sulfur separation from the flue gas by deposition is calculated and the remaining concentrations for sulfur, sodium and chloride compounds in the flue gases is determined.
g. The same calculation procedure will be used for the next layer in the regenerator.

At the end, the deposition and flue gas composition has been calculated as a function of the position in the regenerator.

Figure 4 illustrates this deposition/condensation process.

Dust formation in the flue gas takes place in the cooler sections of the checkers, this dust also diffu-

ses partly to the refractory surfaces, causing deposition of fluffy salt layers.

This process has been described by reference [10].

6. Combination of the four models

For a new glass furnace the calculation procedure starts with the input of the dimensions of the regenerator checkerworks and the packings, the thermal properties of the different refractory materials,
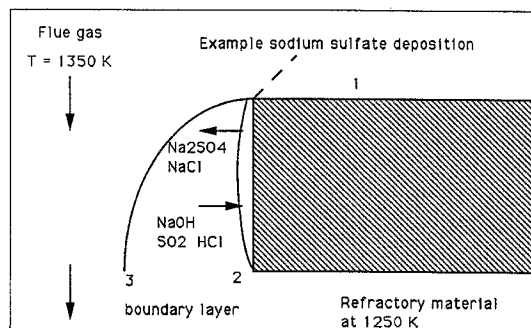
glass furnace data, energy consumption data, air excess and some other process data. A first assumption of the temperature of the flue gases entering the regenerator and an assumption of the temperatures of all the checkerwork layers will be made. Then the heat transfer from the flue gases to the packings as a function of time and layer position will be calculated for the period of this half cycle of the combustion process.

After this step, the preheating of the air in the second half cycle is calculated.

Using the values for the air preheat temperature and an initial assumption for the superstructure temperatures of the furnace and assuming a certain combustion course, the thermal performance of the combustion chamber will be calculated. This leads to values for temperatures in the furnace for flue gas temperatures and flue gas flows entering the regenerator. The regenerator calculation starts again, this procedure will be continued until the exit flue gas and air preheat temperatures become constant values. The thermal performance of the new furnace is now determined. From the batch composition, glass colour, flue gas velocities and temperatures determined by the combustion chamber model, the concentrations of sodium, sulfur and chlorides are calculated using the here mentioned volatilisation modelling. The deposition rates in the regenerator are determined by the
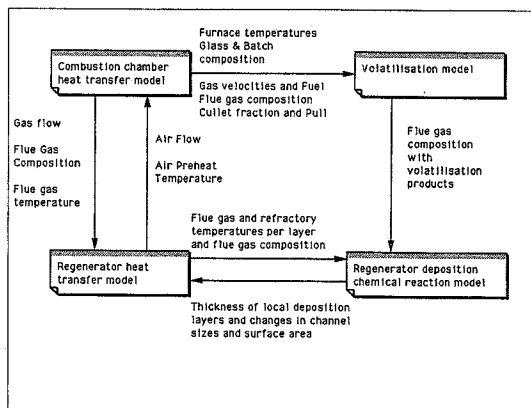
deposition model. Because of formation of the deposition layers the flue gas channels sizes reduce, the surface areas of the checkerworks changes and the heat conductivity of the packings decreases. Regenerator heat transfer calculations will be carried out after a period in which the the deposition process took place, using adapted data taking the

deposition per regenerator layer into account. The heat conductivity changes and the channel width will decrease. New air preheat temperatures will be calculated and the heat transfer in the furnace will be determined by the combustion chamber model. Figure 5 gives a diagram for the coupling of the four sub models.

The thermal behaviour and fouling can be determined as a function of time during the whole furnace lifetime. Timesteps of 1 or 2 months are generally used for the calculation of fouling and the change in thermal performance.

7. Model calculations for different process conditions

Results from model calculations will be presented based on an end port fired regenerative container glass furnace producing soda lime glass.

Preliminary calculations show that the heat conductivity of the sodium sulfate is essential for the aging process. From TNO laboratory experiments with sodium sulfate deposits gathered from the cooler sections of a regenerator, the heat conductivity for sodium sulfate dust has been determined on 0.2 W/m.K. This value can be applied for deposition layers below the sodium sulfate melting point of 884 °C, above this temperature a value of 1 W/m.K has been assumed. The density below the melting point is approximately 1650 kg/m^3, above the melting point: 2680 kg/m^3.

The heat capacity is of minor importance.

Figure 6 shows the influence of the sodium oxide concentration in the melt at three temperatures of the molten glass surface, on the emission of dust

by the furnace, calculated from the volatilisation and combustion chamber model.

An increase of the sodium concentration in the glass slightly increases the dust emissions.

High melting temperatures result in very high volatilisation and dust emission rates. This leads also to a very strong aging effect for the regenerators.
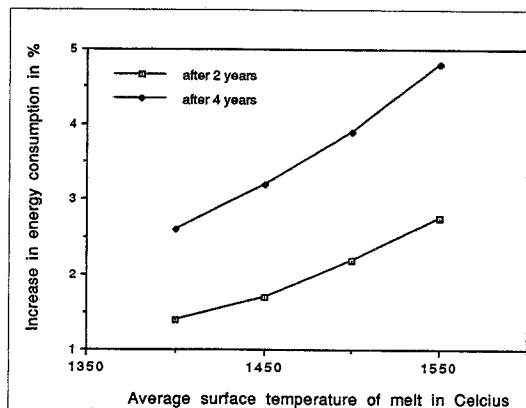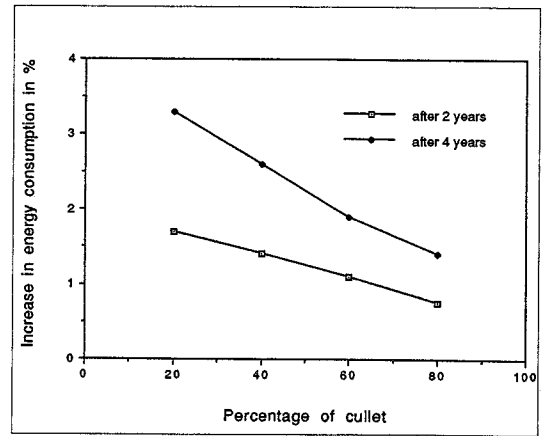
Figure 7 shows the effect of the glassmelt surface temperature on the increase of the specific energy consumption after 2 and after 4 years of operation.

Furnaces which use high fractions of cullet will have less batch carry over and less sodium chloride volatilisation from the soda. The aging of furnaces, using low amounts of cullet is much more intensively than for high cullet melters, as figure 8 shows.

Calculations including electrical boosting show that the aging decreases in the case that the electrical energy supply increases. Figure 9 shows the influence of electrical boosting and the pull rate on the change in thermal behaviour of a glass furnace producing flint glass in a tank with a surface area of 102 m^2. The aging is accelerated by higher pull rates as can be seen from these graphs. At high pull rates, flue gas velocities are very high, leading to high Sherwood numbers. Volatilisation increases and so the deposition rates and the fouling of the checkerworks will increase. Often the specific energy consumption reduces, going from low pull rates up to a certain value, further increases in glass production rates again lead to higher specific energy demands. For melters which use intensively electrical boosting, the aging of regenerators is

moderate, because of the relative low fossil fuel consumption, which gives only moderate flue gas velocities in the furnace and in the regenerators.

8. Results of calculations for different checkerworks

Calculations have been carried out for a regenerative furnace for different kinds of packings.

The flue gas velocities have been changed for a few cases. Flue gas velocities between 0.35 and 0.6 m/s (referred to 273 K and 1013 mbar) are used in the calculations.

The aging, given as the increase of specific energy consumption after 1 year has been derived for diagonal staggered pigeonhole packing, a combination of basketweave (top layers) and pigeonhole (lower layers) packing and chimney block checkers.

For a heat conductivity of the deposition layers of 0.45 W/m.K, which is slightly higher than experimentally derived for sodium sulfate powder, 14 weight % Na_2O in the glass and a molten glass surface temperature of $1480°C$, the effect of aging on the energy consumption is given for these situations by figure 10.

Figure 10. >
Aging and change in specific energy consumption for different regenerator packings and flue gas velocities

The staggered pigeonhole packing and the chimney blocks are less sensitive for fouling than the basketweave checkers. High flue gas velocities, for example for high glass pull rates, lead to an acceleration of the aging of the regenerators. A 50 % higher flue gas velocity causes aging effects which can be 60-80 % higher.

As the channel width in the checkerworks decreases, the specific surface area (m^2 heat exchanging surface/m^3 checker) increases. The deposition will be spread over a larger surface area for small channel sizes and although the total deposition may increase, the deposition layer thickness remains relatively small for these situations. This means that the decrease in thermal efficiency of regenerators with small channels is less pronounced than for checkerworks with wide channel openings.

Figure 11. >
Change in energy consumption after 2 and 4 years depending on channel sizes in checkers made by chimney blocks
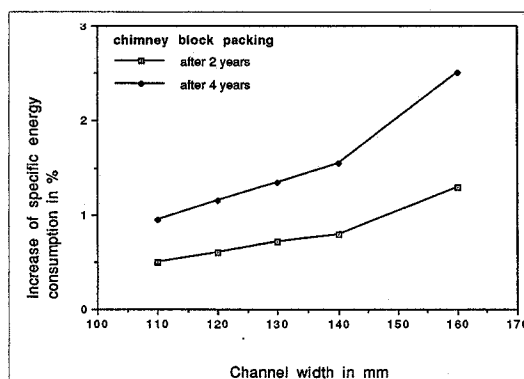
Figure 11 presents the calculated increase of the specific energy consumption after 2 and after 4 years for a chimney block packing as a function of the channel width.

9. Conclusions

The effect of process conditions, construction of regenerator checkerworks and refractory materials used for the packings on the change in thermal performance of glass furnaces can be derived quite accurately by using a coupling of four submodels, describing the thermal behaviour of regenerators, the combustion and radiative heat transfer in the glass furnace, the volatilisation processes and the

chemical reactions plus deposition in the regenerators.

Not only the quality of the new regenerator can be determined, but also the thermal behaviour after several years. Practical observations show that the increase of the specific energy consumption for industrial glass furnace is about 1 till 3 % per year. Model calculations for several conditions show increases of 0.25 till 1.5 % per year only by the effect of fouling by sodium sulfate deposition. Part of the decrease of the thermal efficiency of glass furnaces seems to be caused by corrosion or attack of the superstructure and the aging of the insulation.

Chimney block packings and diagonal staggered pigeonhole checkers appear to be less sensitive for aging than basketweave arrangements. Wide channels could lead to a fast increase in energy consumption for the furnace but will also give reduced risks of channel blockages.

The model can be used to optimize regenerator design and the choice for regenerator packings, which could lead to average energy savings of 2-5 % during the furnace lifetime.
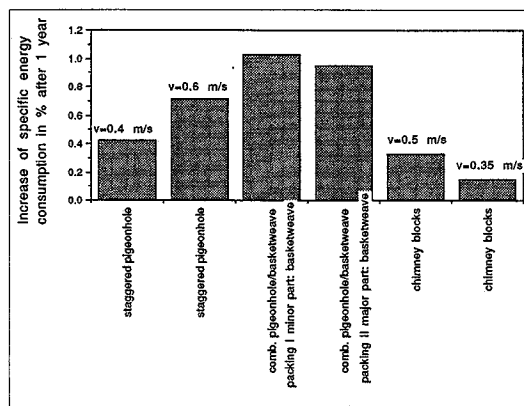
10. Acknowledgement

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