EVOLUTION OF REFRACTORY IN THE GLASS INDUSTRY

Stefan Postrach, Ignacio Ramirez

RHI Magnesita
Kranichberggasse 6

1120 Wien, AT

ABSTRACT

Since the nineteen-fifties, the performance of glass furnaces has improved significantly. We have seen a significant improvement in furnace life. There has been an increase in output while energy demand for the process has been reduced. Factors like improved furnace design, construction, and refractory lining concepts, including the quality of refractories, have contributed heavily to this development. A very important step was of course the invention of fused cast products in the 1920s. However, it took decades to implement fused cast products in different areas of the glass furnace; for example, to replace traditionally used alumina-based bonded products in glass contact areas. In addition, several incremental improvements contributed to the evolution process, but hardly any new product line has been invented. Silica, high-alumina, zircon-based and basic products still determine the lining concepts in which case huge improvements were achieved regarding quality, quality assurance and by optimizing the combination of different materials. An important precondition for such improvements is a clear understanding of the processes that influence refractory corrosion.

This paper will describe important refractory developments and will lead through the evolution of refractory for glass furnaces. In addition to the most important developments of fused cast products, examples for certain furnace assemblies will be given, e.g., melter bottom, melter crown including the development of the insulation concepts, regenerator checker pack and casing. These examples show nicely that the development was driven by evolution: once a problem was solved, another aspect -which was not consequently in the minds before became the problem. Present day trends will also be mentioned, as these trends may give a glance to the future.

As mentioned above, a key aspect that led to the improvements was by clearly understanding the complex conditions in a glass furnace and the influence on refractories. These important lessons that we have learned in the past 70 plus years can help us solve new challenges that come ahead.

INTRODUCTION

The glass production process experienced huge improvements over the last decades, proven by KPIs like furnace lifetime and specific furnace load [1], see Figure 1, but also observable on the quality of glass products. During this evolution the operation conditions changed permanently and heavily. The most important aspect was of course the temperature increase in the glass melting process [2] (Figure 2). Higher furnace temperatures increased corrosion effects and even new and unexpected mechanism became evident.

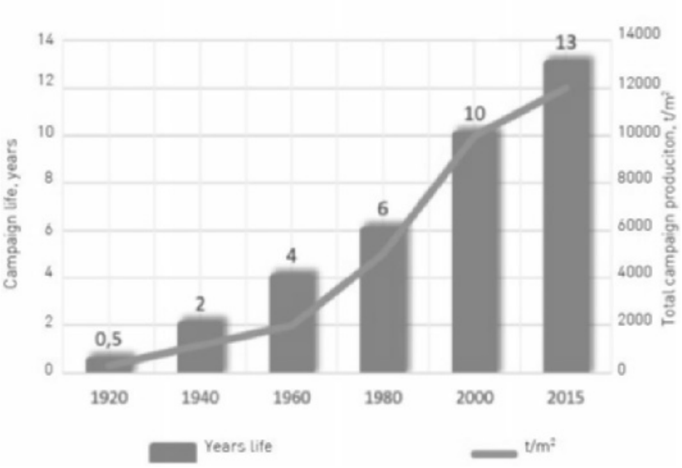


Figure 1. Historic development of furnace lifetime and specific melter load

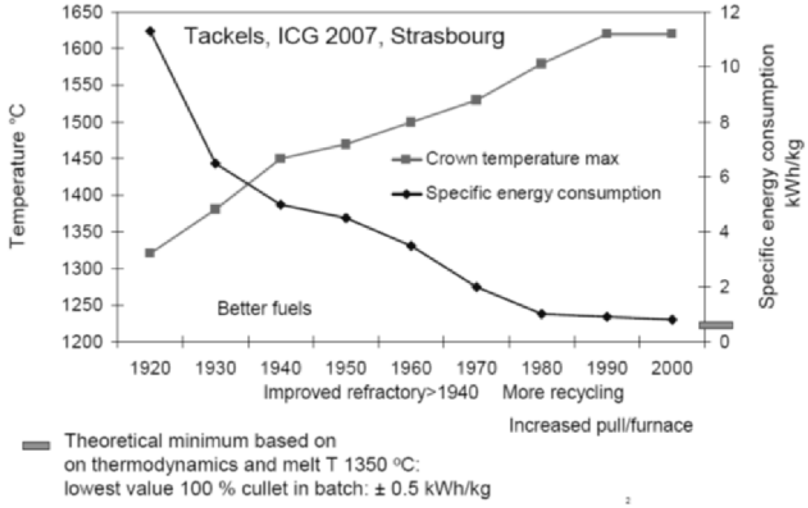


Figure 2. Historic development of crown temperatures in glass furnaces

This paper describes how the lining concepts had been changed over the decades and how the refractories contributed to the improvements in glass melting technology. However, it should not be ignored that other factors supported the improvements as well:

- Furnace and refractory design
- Installation accuracy
- Heat-up processes
- Improved raw material quality including cullet
- And improved equipment's, e.g., the combustion technology and level of automation.

Latest push to prolong the furnace lifetime further resulted out of the development of various hot repair techniques in the last ten years. But now let's focus on the question how refractories could contribute and even influence the development of furnace design. In total there are four general aspects that are of course connected to each other:

1. Understanding the refractory behavior: like in every other application of refractories it is of upmost importance to understand the operation conditions and the influence on the

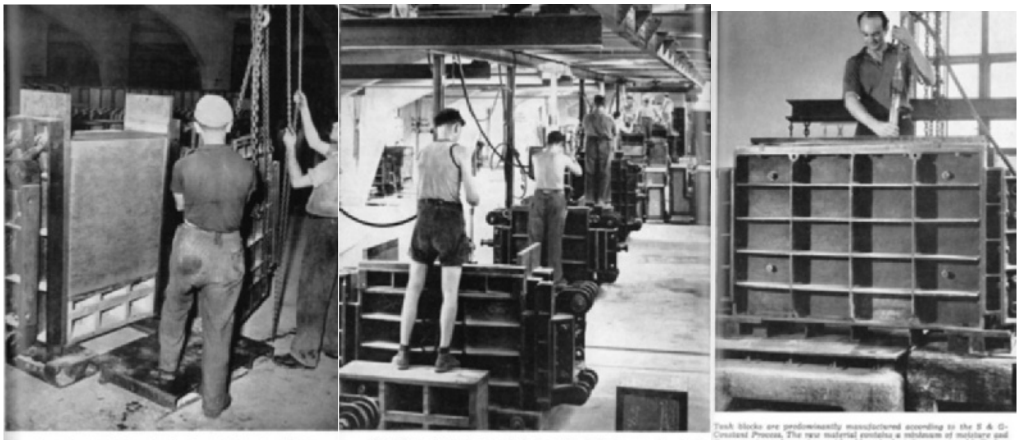
corrosion mechanism. Academic work helped, as well as a close communication and exchange between glass producer and refractory supplier! In this paper some examples of academic work will be given.

2. New products: it is well known that the number of components to produce refractories is limited to mainly six elemental refractory oxides and their derivatives: SiO_2, Al_2O_3, MgO, Cr_2O_3, ZrO_2 and CaO. Carbon, commonly used in refractory products for the steel industry, is hardly found in the glass industry. A lot of the products within this group of oxides are quite old and the potential of concrete new products is limited. A significant contribution on the furnace lifetime and glass quality was the implementation and improvement of so-called fused cast products. Today a glass melting furnace without those products is hard to imagine. Further one should not forget that also other new products moved borders, for example calcium aluminate for tin bath bottom block application (in contrast to fireclay blocks those do not form nepheline) and lime free silica bricks (which increase the corrosions resistance significantly).
3. Adaption of lining concepts to changing requirements: over the last decades it was possible to derive conclusions and therefore to adapt concepts to the changing operation conditions. A furnace part that experienced many changes is for sure the regenerator. As described later, you will recognize that no other glass furnace part was facing so many changes in operating conditions as the regenerator. Further, improved concepts help to save energy, like the melter bottom concept and melter crown improvements.
4. Finally, many refractory producers improved their quality systems which is a very important aspect as most of the raw materials used exhibit strong variating characteristics. The latter point for sure inspired the replacement of natural raw material by synthetic ones, for example, fused raw materials used in different products today.

The focus of the paper will be mainly on points two and three and on the evolution of the refractories for soda-lime glass application.

REFRACTORY EVOLUTION OF THE GLASS CONTACT AREA

The glass contact area is one of the most critical parts of a glass furnace as it is holding the entire glass melt and it has a direct impact on the glass quality. In 1954 the glass contact area of soda-lime furnaces was mainly built out of fireclay blocks with an Alumina content of 25-40% [3]. The lifetime of those furnaces was 1to 3 years. Those tank blocks had been produced manually as can be seen in Figure 3 a-c [4]. The development of the fused cast products goes back to the 1920´s, in 1925 Corning Glass Works filed a patent for the first fused cast product. The standard composition was based on 70% Al_2O_3 and 28% SiO_2 with voids and the first fused cast AZS blocks had been installed mid of 1950´s. These fused cast products were mainly installed in critical areas in combination with fireclay blocks for flint glass furnaces. At these early stages it was also common to have 2 or more layers, one on top of the other.



The problem of handling the burned and unburned blocks puts a ceiling to the measurements of DIDIER tank material. Our photograph shows the opening of a heavy mould in order to strip the dry-pressed block.

The picture shows the manufacturing of tank blocks according to the S & G-Constant Process at Niederlahnstein Works. The technical equipment demonstrates that tank blocks up to the largest practicable sizes

Tank blocks are predominantly manufactured according to the S & G-Constant Process. The raw material contains a minimum of moisture and the percentage of grog is considerably raised at the expense of ball clay, resulting into a batch which can only be worked by means of pneumatic rampers or high pressure forming. Thus bricks of excellent accuracy in dimensions and high mechanical strength are obtained (Tolerance ± 0,15 %).

Figure 3 a-c. Impressions from tank block production in 1960

In 1960´s the fused cast products experienced two important developments [5]:

- The oxidation process to improve corrosion resistance and glass quality: during the melting process it appeared that due to the reaction between the AZS melt, the graphite electrodes and the surrounding atmosphere, carbides and nitrides formed within the melt. Those components within a fused cast block have a negative influence on properties like corrosions resistance, blister formation and exudation. A solution for this problem was the oxidation of the melt before pouring into the molds.
- The AZS product with 41% ZrO_2 had been developed; AZS41. Compared to AZS32 this product exhibits a 30% higher corrosion resistance against soda-lime-silicate glass melt, therefore, still of common use today.

This moment can therefore be seen as the starting point to improve the melter lifetime. However, this development was not fast as still in the 1980´s two-layer sidewall concepts (incl. fireclay) were applied as described by Trier [6] and Tooley [7] (1984), even though the problems of upward drilling were already known (described in 1959 [8]), see Figure 4 a-c.

During the 1990´s sidewall blocks up to 2000 mm height were in use and the common concept was to install AZS41 for sidewall blocks and AZS32 for the bottom paving. The average furnace lifetime was around 8 years.

After those fused cast products became more and more the standard for the glass contact area, the next weak points popped-up, for instance, weir wall, throat, and doghouse corner blocks. As a result, Busby [9] reported a growing interest and application of Cr_2O_3 (26-30%) containing fused cast AZS products (ACZS). However fused cast ACZS did not prevail, but rather iso-chrome became more popular for the critical areas like throat (in container melters), weir wall (if installed) and doghouse area. F. Gebhardt reported that in 1990 iso-chrome products were installed successfully [10].

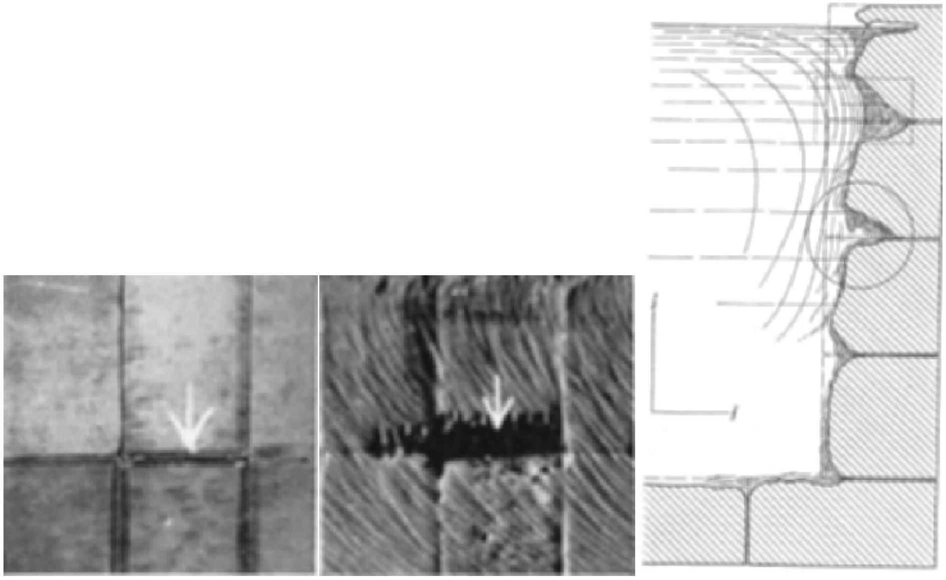


Figure 4 a-c. Jebsen-Marwedel's explanation of upward-drilling

The glass line of the melter remained a weak point due to interfacial corrosion mechanism, Marangoni convection. Overcoating became more and more a standard solution, with different approaches regarding material selection and timing of installation.

It is also worth to mention that a new fused cast product was developed during the 1990´s: High Zirconia Fused Cast (HZFC) with a ZrO_2 content of around 95%. This product offers huge advantages for glass contact with special glass but shows no significant advantages in soda-lime-silicate glass compared to fused cast AZS.

REFRACTORY EVOLUTION OF THE MELTER CROWN

It´s been at least 70 years that silica is in use for melter crown construction. It gathers many valuable properties for the application like good refractoriness, good creep under compression, lightweight, low glass defect potential and inexpensive. Through all these years the silica concept has had the opportunity to improve in different categories.

As for other parts of the furnace, improvement comes from understanding the corrosion process. The rat hole formation process had been explained in 1959 by Jebsen-Marwedel [8] (Figure 5), however the same problems happen from time to time still today.

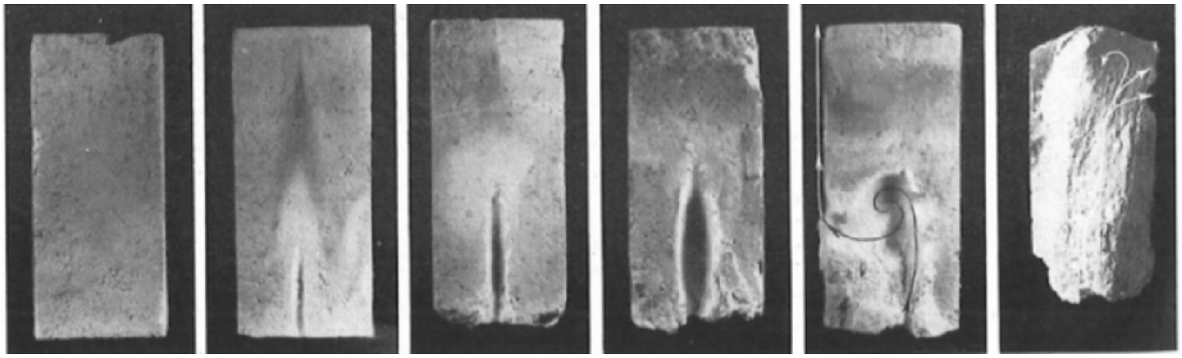


Figure 5. Description of rat hole formation process

Only to gain some perspective about the improvements, in 1954 Günther [3] described that Silica was at its limit under crown temperatures of 1500°C. Today crown temperatures are at least 100°C higher thanks to the following developments:

A better understanding of the silica crown corrosion mechanism and crystal modification properties has allowed the development of the crown´s insulation concepts. Avoiding insulation due to safety reasons is no longer a standard and we have learned that by insulating the crown the condensation temperature is shifted towards the melter crown surface. Therefore, by increasing the insulation the alkali corrosion risk is reduced since its harder for alkali to find the required temperatures to condensate. Nowadays, four and even five insulating brick layer concepts are state-of-the-art. By increasing the insulation and decreasing the crown´s inner temperature gradient the cristobalite´s thickness also increases. Cristobalite is mechanically more stable and more alkali resistant than tridymite. Of course, an energy efficiency improvement is also obtained by adding insulation, see Figure 6.

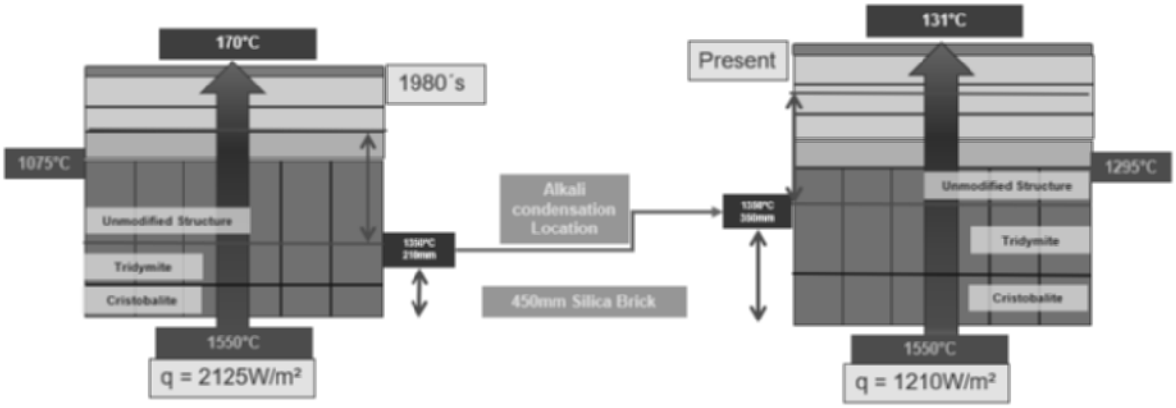


Figure 6. Silica insulation concepts

Residual quartz content has turned out to be a critical parameter for the crown´s aftergrowth and was under study by the 1980´s. In 1984 Trier [6] reported expansion curves according to different problematic residual contents; 7 and 19%. Thanks to further research, today we know that residual quartz should be kept below 0.5% and is controlled with the firing process but also the quartz supply grain size is equally important. Impurities have played a fundamental role mainly affecting the hot properties of the refractory. The impurity level has gained importance up to the point that several classification standards have developed for this reason. Shulver [11] defined standard and super-duty Silica according to their alumina and titania content. ASTM C416 [12] defined type A and B silica bricks according to a flux factor calculation which considers the alkali, alumina, and titania content.

As a final comment on the silica´s quality we should mention that the dimensional accuracy also improved due to the understanding of the relation between opened gaps and rat holing in the crown. Therefore, the brick´s dimensional tolerances have tightened and today the tolerances of the taper should be not more than +/-0.5 mm.

Further, the following aspects contributed to the increased lifetime of silica crowns:

- The importance of proper installation has increased through the years, focusing on thinner mortar joints.
- Although it varies between engineering concepts, the crown´s design features have also contributed to the lifetime increase. The drip course, expansion joints and the buffer layers are among the most common design features with particular purpose.
- Related to design but also to refractory development; proper joint sealing has shown its importance as the corrosion mechanism is better understood.
- The heat-up procedures have also evolved to take better care of the crown.

For the melter crown a few innovations have raised besides the traditional quartzite-based refractory. During the late 1980´s we also saw the introduction of honeycomb hot face crowns (Figure 7) which increase the surface area and emissivity coefficient, resulting in an efficiency improvement (Figure 8). In the 2000's concern about the lime content in Silica bricks motivated further developments due to the negative influence on the corrosion resistance [13]. The first lime-free silica bricks were developed and installed since 2005. Removing one of the brick´s weaknesses allowed higher application temperatures and improving resistance against corrosive agents. Figure 9 shows the installation of lime-free-Silica (Stella® GNL) in stressed areas of a regenerative heated float melter.

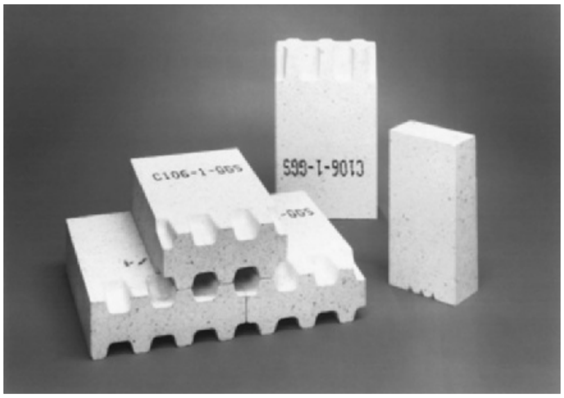


Figure 7. Silica bricks with honeycomb structure

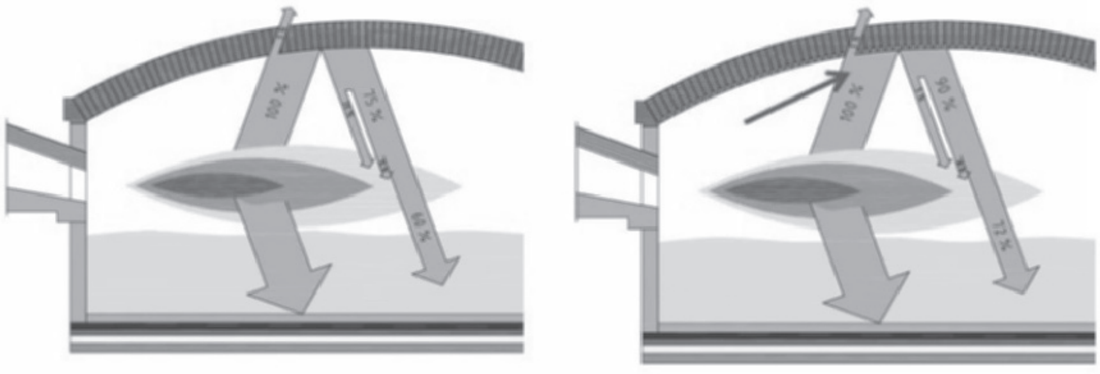


Figure 8. Improved efficiency with honeycomb structure in melter crowns

Figure 9. Installation of lime-free-Silica (Stella® GNL) in stressed areas of a regenerative heated float melter

REFRACTORY EVOLUTION OF THE MELTER BOTTOM

The evolution of the melter bottom concept is a clear example of how the continuous improvement process introduced a solution for a requirement and in consequence the next challenge appeared. With the objective of energy saving, glass homogenization and increased melting temperatures, insulation increased over the years showing an equal increase on the floor wear rate. In 1974, Tooley [7] described the transition from simple, 300 mm thick clay flux melter bottoms, towards the present multi-layered concepts. Currently, 750 to 850 mm are common thicknesses. Trier [6] (1984) shows several tank bottom construction concepts which give a fine idea of the bottom´s insulation gradual increase, basically no insulation layers initially with gradual layer additions on further designs. He also describes a rammed monolithic layer for special situations, showing that around the 1980´s the monolithic and safety layers weren't standard yet. Figure 10 shows the different concepts.

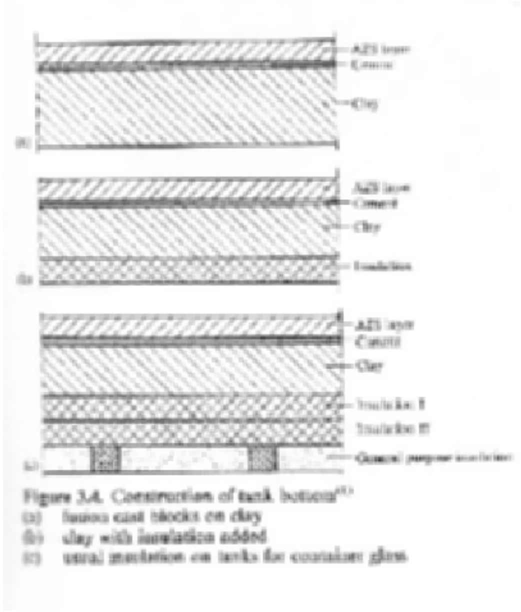


Figure 10. Different bottom concepts. No insulation (a), one layer (b), three layer (c)

In the same decade, Busby [9] (1986) concluded that the added insulation increased the floor´s temperature which led to aggravating the floors wear. He reported that although the paving slabs showed good performance, metal and glass mobility through the joints proved to be the weak path on the furnace´s bottom. With such arguments he justified the implementation of a monolithic layer describing the following requirements: corrosion resistance, no bubble generation, and a joint-free barrier to avoid infiltration.

Downward and upward drilling became the topic of study for refractory selection, development, and furnace design. As described on the glass contact topic, the upper paving, 75 to 150 mm thick, has shifted mainly to the fused cast AZS solution although bonded AZS had also been regarded. Below the pavers different combinations are possible. The present designs usually have in common at least one monolithic and one safety layer. These layers have taken two main paths depending on the furnace requirement. A Zircon concept has remained popular to prioritize the metal corrosion resistance with its encapsulation capabilities. For furnaces with no metal contamination concerns, glass corrosion resistance is the main priority, and it is addressed with zircon-mullite bonded AZS grades. Figure 11 shows a typical concept.

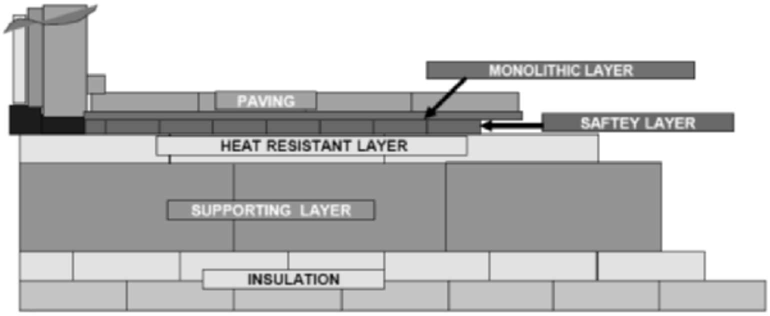


Figure 11. Typical bottom concept

A tendency for larger pavers and sublayer blocks with tighter dimensional tolerances became desirable due to the joint thickness reduction and faster installation.

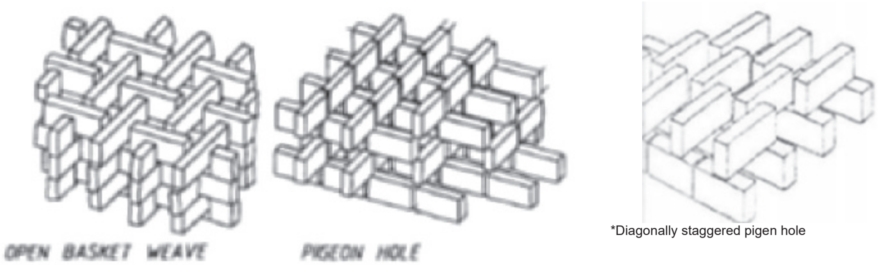
REFRACTORY EVOLUTION OF THE REGENERATOR

As already mentioned above the regenerator passed through a lot of changes and still today the conditions are varying heavily.

By mid of 1950´s the regenerator casing was made of fireclay bricks and the checkers were mainly based on fireclay with an Al_2O_3 content of 43% [14]. Due to the high alkali corrosion the discussion about alternatives for the heavily attacked blocks started and low iron MgO had been introduced successfully14). This was the starting point to switch to MgO-based products, especially for the checkers but also for the walls. By the beginning of 1960´s more oil was used instead of gas, consequently V_2O_5 corroded the checkers and the condensation of additional sulphates became a problem. Since then, two height zones were distinguished, above and below 1200°C, leading to different solutions for each zone. Mid of the 1980´s Tooley described a typical checker concept as follows: 1/3 $MgOCr_2O_3$ (MgCr) at the lower end and MgO on the upper 2/3 [7]. Still in 2002 MgCr [15] was widely used not only for the checker work but also in the upper casing area, due to the expected benefits in regards of creep-resistance over pure MgO bricks.

By the end of 1980´s the consequent replacement of MgCr by MgO-$ZrSiO_4$ products started in Europe. This solved environmental disposal issues. Independent of this, still in 1999 the regenerator was described as a weak point [10]. This was due to changing operation conditions: especially the top-layers were corroded by finer batch and higher cullet-share, further the waste gas inlet temperatures increased from 1450 to 1550°C [10].

Now let's have a look at the checker shape types. This helped a lot to improve the lifetime, performance, and energy efficiency. Until 1980´s, straight brick checkerpacks were state of the art (see Figure 12). In 1973, cruciform based on fused cast material were developed. This was the first thin-walled checker but the actual breakthrough of thin-walled checkers was in 1980´s with the development and implementation of chimney pots. This concept offers huge advantages regarding the stability of the checker work and already in 1986, the lifetime of the chimney block setting exceeded the lifetime of the container glass furnace, at the time about 5 years long, and went into a second campaign.


OPEN BASKET WEAVE

PIGEON HOLE

*Diagonally staggered pigen hole

Figure 12. Different checker systems

SUMMARY AND LEARNINGS

- The glass production, glass furnaces and the installed refractory materials experienced huge developments in the last 100 years.

- New refractory products and lining concepts had been developed and refractories are not seen as the limiting factor anymore as 20 years lifetime can be reached to produce soda-lime silicate glasses and high performing furnaces are state-of-the art.
- The challenge for refractory producers today is to support the development of new technologies to reduce the CO_2 footprint of this fantastic material, glass.
- To speed-up solutions connected to refractories, a close contact between glass furnace operators, design companies and refractory producers is necessary to understand the requirements in detail, as we have learned that changes may influence the linings performance.
- For sure hybrid furnaces, new electrical and combustion concepts will influence the refractory lining concepts, probably much more than the switch towards oxy-fuel firing almost 30 years ago.
- Refractory suppliers are also working on CO_2 reductions of their own products. One important aspect is to make the CO_2 footprint visible on the technical data sheets for the customer´s information.

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