

Automation to react to fuel gas quality and achieve optimum process parameters in glass melting

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The glass industry consumes high quantities of fuel gas from national natural gas distribution grids for melting glass batches. The quality of this fuel gas can change quickly within a day and also shifts with seasonal cycles, according to the blended composition of methane lean and methane rich sources.

Highly accurate temperature control during a glass melt is absolutely critical for production quality. Burner energy efficiency and waste gas emissions pollution control are also highly important issues which can be influenced by changes in natural gas composition. To meet this application, an automated method of feed-forward process control using a fast response micro GC-TCD arrangement can be used. The critical question of the most suitable carrier gas for this application is discussed along with calibration gas mixture requirements.



1 Introduction

The glass industry consumes high quantities of fuel gas from national natural gas distribution grids for melting glass batches. The quality of this fuel gas can change quickly within a day and also shifts with seasonal cycles, according to the blended composition of regionally generated biogas (methane lean), propane vaporisation to enrich the biogas, piped north sea natural gas (methane rich) or Russian natural gas (medium methane rich). In some countries, such as NL and Germany, the biogas contribution to the grid has grown significantly in recent years and the mix of natural gas sources continues to diversify.



Highly accurate temperature control during a glass melt is absolutely critical for production quality, and failures to control within a tight range can result in

batch wastage. It is therefore highly beneficial to make feed forward adjustments to the burner operation to mitigate for changing fuel gas quality. Burner efficiency, waste gas emissions pollution control and furnace refractory lining life are also highly important issues which can be influenced by changes in natural gas composition.

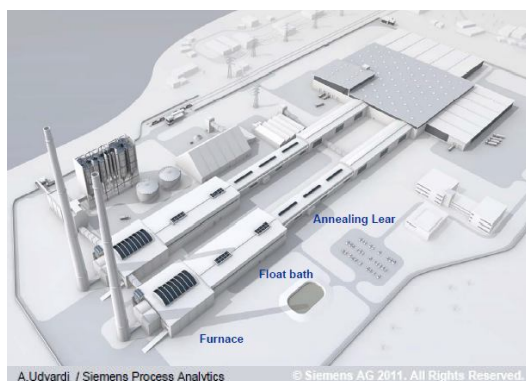
To meet this need, an automated method of feed-forward process control using a fast response micro GC-TCD arrangement can be used. This system analyses the fuel gas quality and computes its probable thermal characteristics using methods similar to those in ISO6976 [2] to allow the required adjustments in burner operation to be made. The analytical matrix and the overall system control loop parameters can vary from site to site, and over time within the same site and therefore the critical question of the most suitable carrier gas for this application must be addressed. The most suitable calibration gas mixtures must also be considered.

2 Glass Production and Melting

2.1 Overview of Glass production process

Although glassmaking dates back to ancient times, in many ways it reflects the same basic processes in today's industry. In the 21st century however, mass production of float glass to comply with modern building standards and changing legislation has become an art that the ancients could not have imagined.

Alongside glass for packaging, float glass used for glazing in buildings and housing accounts for one of the biggest single usages of glass in the modern world. Float glass production has been relatively unchanged for more than 60 years, but the final product has changed dramatically from a single equilibrium thickness of 6.8 mm, to today's range from sub-millimetre to 25 mm, exhibiting almost optical perfection.

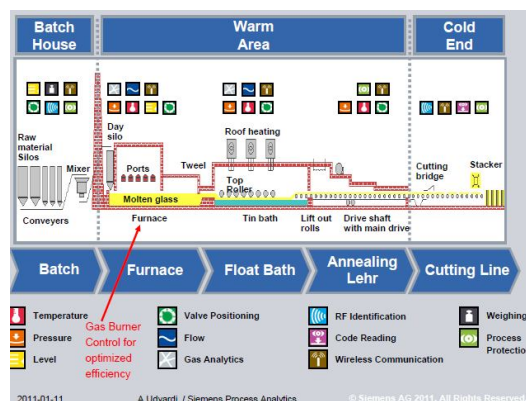


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A commercial glass manufacturing line comprises a series of steps that begin when a mixture of fine-grained ingredients, typically sand, soda ash, limestone, dolomite and some other minor elements, are combined and melted to form molten glass in furnace at temperatures reaching 1,500°C. The temperature profile during the melt, ie how fast the glass is melted and to which maximum temperature the glass melt is exposed can have a dramatic effect on the glass. A few degrees of temperature profile change can influence hardness, toughness, brittleness and colour to the extent that a whole melt batch might be scrapped. And this waste represents a loss of energy efficiency, loss of productive time for the glass process line and increased environmental impact.

The subsequent steps of refining and homogenising occur in separate stages, driven by extremely high temperatures. In the case of float glass, this continuous melting process can take as long as 50 hours to deliver a quality of glass free from inclusions and bubbles to the float bath. Here the liquid glass floats on a bath of molten tin as it hardens and where its thickness and width are adjusted.

The next step is cooling. In the case of float glass, the sheets are lifted out of the liquid tin onto conveyer rolls at a temperature of about 1,100°C and then into the annealing kiln or "lehr". The glass comes out of the lehr at about 350°C and is cooled towards room temperature by open air fans.



[1]

2.2 Glass melting burners and temperature control

To operate the burner efficiently, the plant should have various process control measures in place. A burner needs to operate as close as possible to a stoichiometric mixture of one to one, in terms of the amount of oxygen molecules and fuel coming in. If there is more oxygen than is required, there will also generally be more nitrogen than is required. And this will increase the amount of oxides of nitrogen (NO_x) produced, which will have downstream consequences in terms of flue gas treatment or emissions. Conversely, if there is too little oxygen and a surplus of fuel, the fuel will not be completely burnt, leading to wastage. Since methane is the main constituent of natural gas, the emission of this fuel into the atmosphere will also have global warming implications.



COROX® convective glass melting in operation

Linde's oxyfuel melting portfolio includes its COROX® convective glass melting technology. Consisting of a unique roof burner, it delivers a more targeted and efficient application of energy, thereby ensuring enhanced heat transfer.

2.3 Fuel selection for glass melting

In a few cases, glass production furnaces are fired by a direct electrical source, but more generally, the enormous amount of heat required

derives from a burner that consumes natural gas and oxygen. The oxygen is pumped into the chamber by fans and combined with jet streams of natural gas, which produce flames that cause the ingredients to react and melt in a matter of minutes.

2.3 Oxidant selection for glass melting

According to recently published IED BREF notes for Glass melting [5], the best available technologies to reduce NO_x emissions from glass melting furnaces can be one, or a combination of a variety of techniques from combustion modification and oxyfuel melting. In altering combustion, options include a reduction of the air/fuel ratio, reduced temperature, staging the combustion process, employing flue gas recirculation, adjusting fuel selection and the installation of low NO_x burners.

Employing greater levels of oxygen substantially increases the thermal efficiency of a furnace, as radiant heat transfer of furnace gases produced by oxyfuel combustion is significantly more efficient than those of airfuel. As is well known, combustion in the heating process is about fuel, oxygen and ignition. Air contains only 21 percent oxygen, with the remaining 79 percent being practically all nitrogen. In the combustion process nitrogen can essentially be regarded as ballast - with this ballast acting as a negative influence as it neither takes part in, nor helps, combustion. However it is heated up in the process anyway. Heating air rather than oxygen substantially increases the production of NO_x.

3 Natural Gas Composition

3.1 Natural Gas Sources and calorific value

The composition and calorific value (similar to the Wobbe index) of piped Natural Gas can change quickly - within minutes. It can also change significantly throughout the course of a day and also has longer term macro shifts, with seasonal cycles – see figure below.

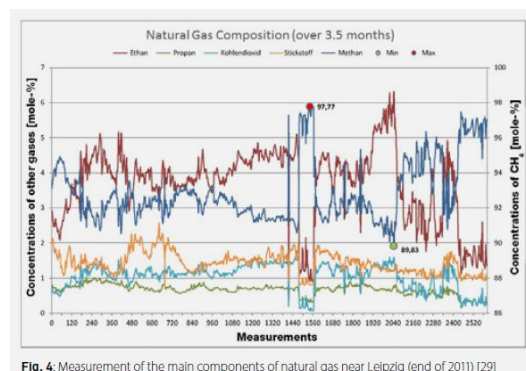


Fig. 4: Measurement of the main components of natural gas near Leipzig (end of 2011) [29]

[29] Giese, A.: "Auswirkungen von Gasbeschaffenheitsschwankungen auf industrielle Prozesse", 87. Glastechnische Tagung, Bremen, Germany, 2013.

Changing natural gas qualities:
impact on industrial gas-fired
applications

by Jörg Leichter, Anne Giese

[4]

One main reason for this change is that various sources are mixed to create natural gas. In various countries and regions there are standards within which the natural gas composition & calorific value must remain, but these are not standardised across the world and not even standardised within Europe and the EU, where natural gas pipelines cross-cross the continent in a diversified energy supply network. See figure below.

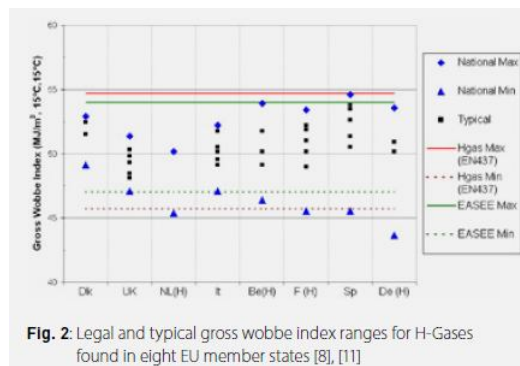


Fig. 2: Legal and typical gross wobble index ranges for H-Gases found in eight EU member states [8], [11]

[8] Cagnon, F.: "National situations regarding gas quality" MARCOGAZ, UTIL-GQ-02-19, 2002.

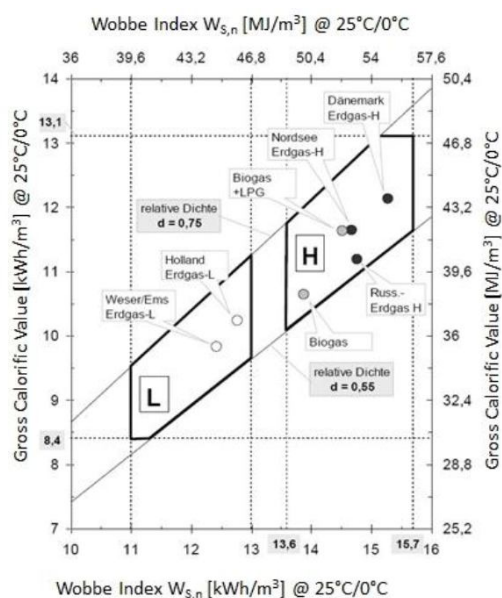
[11] Abbott, D.: "The impact of variations in gas composition on gas turbine operation and performance", Energy Delta Institute Quarterly, vol. 4, no. 1, 2012.

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Using Germany as an example, the blended composition of natural gas arriving at a user, such as a glass melting furnace, may be a mix of regionally generated biogas (methane lean, low calorific value), intermittent slugs of high calorific value vaporised propane to enrich the biogas, piped north sea natural gas (methane rich, high calorific value) or Russian natural gas (medium methane content, and therefore medium calorific value). See figure below.



Permissible gas quality ranges in Germany

[11] "Technische Regel - Arbeitsblatt DVGW G260 (A), 'Gasbeschaffenheit', Bonn, 2013.

Impact of Changing Natural Gas Qualities on Industrial Combustion Processes

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[3]

Across continental Europe, the use of Russian natural gas has been on the increase in recent decades and in some countries, such as NL and Germany, the biogas contribution to the grid has grown significantly in recent years. So, the mix of natural gas sources continues to diversify and the need for automated smart process control reactions to these variations is therefore of increasing importance to many industrial consumers of natural gas.

Since these composition changes can take place in a very short time frame of several minutes, the furnace must be able to react to these changes within a similar time frame to ensure stable and optimum operation.

3.2 Estimation of natural gas calorific value from measured chemical composition

The measurement of natural gas calorific value can be done using flame techniques and calorimetry. This is the classical approach, but it is slow in comparison to the time frame that the natural gas composition and calorific value changes. For a fast process control loop which is able to direct process responses in the same time frame that the natural gas composition is changing, a faster method of measurement is required. Analysis of the fuel gas chemical composition, and computation of the

implied calorific value using methods similar to those in ISO6976,[2] has in recent years of practical application been shown to be a good proxy for flame methods of calorific value measurement. It does, of course, rely on a fast and accurate method of chemical species measurement and a rapid response Micro GC-TCD is appropriate, since it can enable a process loop response time in the order of 1 to 3 minutes.

Fulfills requirements of ISO 6974-5

Concentration - Range (Mol. %)	Repeatability according to ISO 6974-5 (2001); Mol fraction (%) absolute
$50 < x_i < 100$	0.03
$1 < x_i < 50$	0.011
$0.1 < x_i < 1$	0.006
$x_i < 0.1$	

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[1]

4 Burner and fuel process control system

4.1 Oxygen injection feed back loop

The best way to ensure optimal operation of the burner is to measure and control the amount of oxygen in the burner flue gas using a feed back control loop. This ensures there is a small residual amount of oxygen emerging in the escaping flue gases. Although this is the most economically and environmentally efficient way to run the process, plant operators should guard against having a large excess of oxygen which could impact production costs.

To achieve the right balance, oxygen should be measured in the furnace, or in the regenerator heat exchangers where the flue gases leave the furnace and pre-heat the gases coming into the furnace. This measurement is fed into a feedback process control loop and the measurement is typically achieved using instrumentation such as a Zirconia oxygen analyser, which is reliable and robust in this very hot operating environment. The instrument's sensor requires periodic calibration either with ambient air, or with a specialty gases calibration mixture of percentage level oxygen in nitrogen, close to the measurement point, to ensure accuracy of measurement.

4.2 Natural gas composition feed forward loop

While a feedback control loop is essential to measure oxygen levels in the melting furnace and make adjustments to the oxygen or natural gas being fed in, the more sophisticated process control strategy is to use a feed forward control loop in combination with the feedback control loop. The feed forward loop measures the chemical composition of the natural gas (as a proxy for calorific value) coming into the furnace and enables

automated predictive and proactive feed forward adjustment of natural gas flow rate and associated stoichiometric oxygen flow rate.

Components of Burner Gas

Components	Measuring ranges (mol%)
Methane	50 - 100
Nitrogen	0 - 25
Carbon dioxide	0 - 20
Ethane	0 - 20
Propane	0 - 15
i-Butane	0 - 10
n-Butane	0 - 10
Neo-Pentane	0 - 1
i-Pentane	0 - 1
n-Pentane	0 - 1
Hexane ⁺	0 - 3
Hexane	0 - 1
Heptane ⁺	0 - 3
Octane	0 - 1
Nonane ⁺	0 - 1
Oxygen	0 - 4
Helium	A constant concentration can be added to the component list.
H ₂ S	< 500 ppm Not measured

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This process control loop ensures that the thermal input to the furnace remains under control so that the temperature profile of the glass batch melt is controlled according to the optimum process requirements.

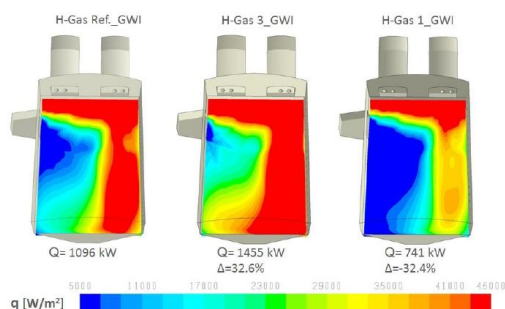


Figure 4: Heat flux distribution into the glass melt for the first scenario (constant air ratios) (Source: GWI)

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[3]

4.3 Fast-response micro GC-TCD instrumentation

These feed forward control loops usually incorporate gas chromatography instrumentation with a thermal conductivity detector (GC-TCD) to measure the quality of the natural gas coming into the furnace and allow for feed forward adjustments in either oxygen or the natural gas flow rates, based on the calorific or heating value of the natural gases coming in. This is a critical factor, since natural gas is fundamentally a mixture of gases whose composition changes over time, impacting on the total calorific value and furnace temperature profile.



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[1]

Use of a Micro GC, in contrast to a larger general purpose conventional laboratory GC, is convenient for two reasons. Firstly, the compact design allows the instrumentation to be used in-situ, close to the process to minimise sample line length and therefore achieve rapid response times. Secondly, the compact purpose-built design will have a short GC column optimised for the natural gas composition measurement application which will again enable rapid response times for this process control loop.

5 Specialty gases required for GC-TCD operation

5.1 Carrier gases

5.0 grade Helium (99.999%) is the most typical carrier gas for gas chromatography in general and is also the standard choice for this application. The benefits are ease and safety of product handling, good speed of separation (essential for this feed forward process control loop application) and broad

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range of applicability. The disadvantage of using Helium in this application is that the natural gas sometimes contains Helium... and the use of Helium as the carrier gas prevents the measurement of Helium as a component of the natural gas.



5.0 grade Hydrogen is also suitable, with an even faster column velocity and separation speed performance than Helium. However, some users prefer not to handle flammable cylinder gases, due to safety reasons. The separation resolution is also not as good as Helium for some species and matrices. However, use of hydrogen may be suitable in Helium-rich natural gas streams.



5.0 grade Nitrogen and Argon are also potential GC carrier gases. They are inert and easy to handle and are generally abundantly available at suitable 5.0 grade chromatography purities on many industrial sites. However, their speed of separation is not ideal for this rapid response application and they give poor sensitivity to the TCD.

Instrument manufacturers recommendation should be considered.

5.2 Zero gases

As with most Gas Chromatography applications, the carrier gas doubles up as the zero gas. Additional zero gas selection is not usually required.

5.3 Calibration gas mixtures

The most suitable calibration gas mixture is a multi-component mixture of hydrocarbons with a similar composition to the natural gas stream. Clearly, one of the reasons for this application being important is that the composition of the natural gas stream changes... so selection of a suitable calibration gas mixture that is representative of the general fuel gas composition is recommended. In some cases, it might be suitable to use a suite of 2 or 3 calibration gas cylinder with different "synthetic natural gas" mixture compositions to give a breadth of calibration points across the spectrum of operation.

Since this is a process control application, not a legislatively controlled emissions monitoring application and not a natural gas custody transfer application, regular certification of the synthetic natural gas mixture is highly suitable. ISO17025 or ISO Guide 34 Accredited certification for these synthetic natural gas mixtures is, of course, technically feasible and commercially available, but these accredited products are more complex to manufacture and therefore add cost, which is not generally required for this process control application.



5.4 Specialty gas equipment

BASELINE 5.0 grade specialty equipment is a good choice for this process control application. REDLINE 6.0 grade equipment would also be suitable, but may be over-specified.



6 Other Specialty Gases used in Glass production

6.1 Sulfur Dioxide

Towards the end of the float glass production process, molten glass is 'fed' through metal rollers in order to smooth and 'iron out' any defects on the surface of the glass. In this extremely reactive environment, measures must be taken to prevent the glass from reacting with the rollers and other materials handling equipment in order to mitigate damage to the final glass product and to extend the life of costly capital equipment such as the rollers. This is achieved by injecting an atmosphere of sulphur dioxide (SO₂) around the equipment, so that the rollers and the sheets of glass are never actually in contact with each other. Instead, a very thin film of SO₂ gas reacts with the surface of the glass to produce desirable chemicals that prevent the molten glass reacting with the roller metal, which would damage the rollers and compromise the quality of the glass.



The dosage of SO₂ is critically important – too little or too much SO₂ can detrimentally impact the glass quality or the rollers. The gas supply system used to deliver the SO₂ is as important as the quality of the gas itself. The plant operator responsible for SO₂ delivery needs to ensure correct dosing under variable conditions, including, for example, an increase in production or a change to ambient conditions – particularly changes in humidity are an important factor because more SO₂ will be required in a wetter humid atmosphere.

6.2 Ammonia for pollution mitigation

Float glass production facilities are often obliged to monitor and control their emissions profile in order to manage the associated gaseous environmental pollutants such as carbon dioxide, oxides of nitrogen (NO_x) and SO₂. Burning air and fuel produces large quantities of oxides of nitrogen. At the high temperatures, nitrogen reacts with oxygen from air to produce NO_x, while some chemicals typically present in natural gas, such as sulfur compounds, also react with oxygen from air to produce SO₂.

Therefore typical stages in flue gas clean-up from the burner are DeNO_x and desulfurisation via a SO₂ scrubber, occasionally with carbon dioxide knock down, before gas is emitted to the atmosphere. In some technologies such as SCR and SNCR, as emissions gases flow through the DeNO_x unit, ammonia is added to the flue gas to reduce the NO_x back to their molecular nitrogen, while in the SO₂ stripper, various chemicals absorb the SO₂, changing it from a gaseous form into a liquid form, where it can be handled and treated more efficiently.



Ammonia can be added to the DeNO_x unit because it contains a large amount of hydrogen that is able to reduce the NO_x to react with the oxygen in nitric oxide and convert nitric oxide back to nitrogen, which is regarded as an environmentally acceptable gas to emit. Ammonia is supplied in bulk deliveries to major float glass facilities, or in drums and cylinders to smaller scale, R&D and pilot glass production facilities.

6.3 Process control and CEMS gas mixtures

Essential to monitoring the different clean-up operations are process control gas mixtures containing, in this application, oxides of nitrogen or SO_2 , and frequent calibration is needed with very accurate and precise specialty gases mixtures. These mixtures calibrate the continuous emissions monitoring (CEM) instrumentation in the process train, measuring the flue gas as it comes through all the process steps and eventually goes up the smoke stack. In general, nondispersive infrared sensor (NDIR) sensors or Fourier transform infrared gas analysers (FTIR) are used for these measurements, both of which require a broad range of calibration gas mixtures, typically mixtures of nitric oxide in nitrogen, or mixtures of SO_2 in nitrogen at relatively low concentrations, sometimes in parts per million.

The role of specialty gases will be critical to ensuring compliance with the new EU Industrial Emissions Directive 2010/75/EU as they are essential for calibration of the emissions measuring instruments – and are precisely prepared for each glass manufacturer to meet their emission monitoring needs and enable traceability to national standards using an approved scheme of accreditation. In many cases where CEM systems are installed, the composition and quality of these calibration mixtures will be independently regulated and controlled by external auditors and in these cases as the preference, or requirement, for accredited schemes of measurement, such as ISO17025 is strong.

6.4 Glass coating and etching gases

The role of industrial gases continues to the final stages of float glass manufacture. Krypton can be used in a technology known as “sputtering deposition” to coat the surface of the glass with a thin film of metal. This surface treatment maximises its energy efficiency and reduces the requirement for electrical heating in a building. This application is often strictly specified for buildings in the USA and in Europe to ensure these structures comply with energy efficiency regulations.

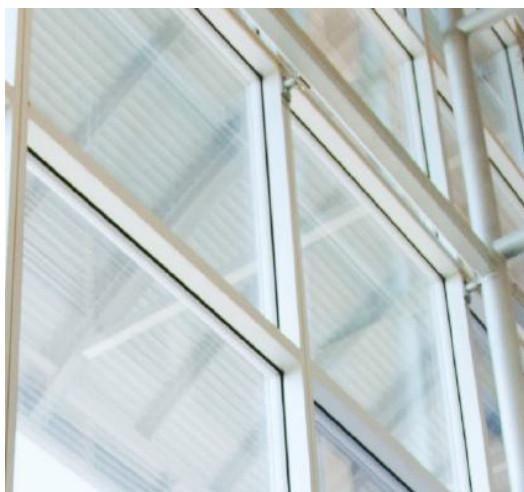


In a similar application, hydrogen fluoride is used to etch the surface of the finished glass. This highly aggressive gas produces a non-reflective surface on float glass and enhances other surface treatments. The etching process is particularly important for window glass used in buildings located in hot climates to minimise building refrigeration costs and deflect UV rays, which can be harmful to human skin.

Additionally, silane (SiH_4), has proved itself to be a highly effective glass coating in reducing the ability of ultra violet (UV) light to penetrate glass – particularly that which is destined for use in window manufacture. Silane reacts with the glass surface in a chemical reaction and helps to mitigate the adverse effects of UV rays on human health, including both short-term issues such as sunburn and potentially longer-term issues including skin cancer. These are points of concern to health authorities, particularly in countries where intense sunlight and heat are unrelenting, such as in the Middle East.

6.5 Double glazing insulation gases

The inert gas argon or the much rarer krypton can be injected between sheets of float glass when double or triple glazing is produced. This achieves superior sound and heat insulation properties that deliver significant energy efficiency benefits, while reducing condensation, dampness and noise levels inside the building.

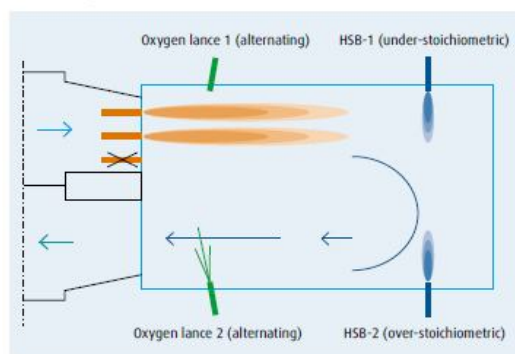


6.6 Oxygen as a burner oxidant gas

Use of oxygen injection to enrich the oxygen concentration of natural air in so-called LowNOx burners can improve process intensity and can also reduce NOx emissions considerably. The process involves injecting additional oxygen through high-pressure lances to create a more intense, directional flue gas recirculation effect within the furnace. As a result, the main air/gas burner system produces a diluted, staged combustion process. The fuel dilution leads to a more homogenous flame and a reduced flame temperature. As the flame temperature has a direct impact on NOx levels, this lowers emissions significantly. A lower flame temperature also reduces the concentration of hydrocarbon radicals in the furnace, thereby limiting NOx formation. In addition, an improved heat transfer rate shortens the window during which NOx can form.

Figure 4: Top view of COROX® LowNOx installation

■ Air ■ Flue gas



6.7 Maintenance and welding gases

The numerous materials handling devices with moving parts in a glass factory require frequent

maintenance and repair. Since they are predominantly of steel construction, welding gases and gas mixtures are often required for maintenance operations.

7 Application in other industry sectors

7.1 Thermal processes

Glass manufacturing is a high-temperature, energy-intensive undertaking associated with the combustion of natural gas and oxygen. This characteristic is also found in other industries, such as iron and steel processing, refining and cement manufacture. Whilst these industries often use liquid or solid fuels for combustion, where they do use natural gas, the principles of the combustion control application described here are transferrable.



7.2 Natural gas quality measurement

The principles of natural gas quality measurement described in this application have transferability to other areas of natural gas supply grid operation and control. Furthermore, the process control loop for burner optimisation is transferrable to many industries using combustion of natural gas for high thermal load applications, such as oil refining, power generation and chemicals production.

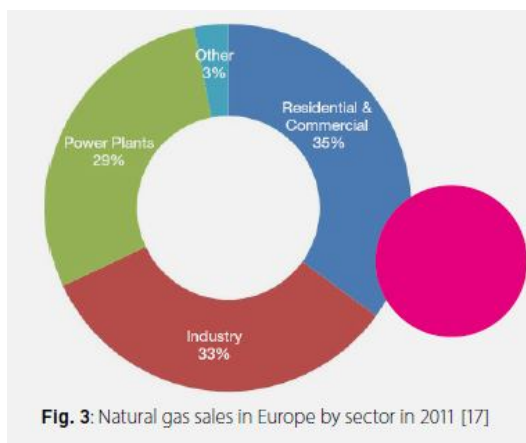


Fig. 3: Natural gas sales in Europe by sector in 2011 [17]

[17] "EUROGAS Statistical Report 2012", Eurogas, 2012.

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8 Conclusion

Economic and environmental factors are driving the need for automated process optimisation in many industries. And, the diversity of energy supply is resulting in rapid shifts in natural gas composition. In order to achieve the optimum process parameters with the transient quality of natural gas, a feed forward process control loop using a micro GC-TCD operating with high purity specialty gases and high accuracy specialty gas calibration mixtures is an appropriate solution.



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