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DECARBONISATION OPTIONS FOR THE DUTCH CONTAINER AND TABLEWARE GLASS INDUSTRY

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Manufacturing Industry Decarbonisation Data Exchange Network

Decarbonisation options for the Dutch container and tableware glass industry

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This report was reviewed by Ardagh Glass Dongen B.V., O-I Netherlands B.V., Libbey Holland, PBL and ECN part of TNO remain responsible for the content. The decarbonisation options and parameters are explicitly not verified by the companies.

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FINDINGS

Summary

This report outlines the current situation of the Dutch container glass and tableware glass production and the options and preconditions for its decarbonisation.

Currently, manufacturing of container glass in the Netherlands takes place at four locations: Dongen (Ardagh Group), Moerdijk (Ardagh Group), Leerdam (OI Netherlands) and Maastricht (OI Netherlands). Tableware glass is produced in Leerdam (Libbey Holland). An overview of their production capacities and the registered emissions (NEa, 2018) is given in Table S1.

Producer	Location	Main activity	Number of Furnaces	CO ₂ emissions 2017 [kt CO ₂ eq/yr] (NEa, 2018)	Production capacities (packed) [kt/yr]
Ardagh	Dongen	Glass-containers for food and beverage	3	92.8	300
Glass	Moerdijk	Glass-containers for food and beverage	2	60.2	200
0-I	Leerdam	Glass-containers for beverage	3	75.0	280
Europe	Maastricht	Glass-containers for food and beverage	3	96.9	250
Libbey Glass	Leerdam	Tableware for daily use	2	27.5	40-45

Table S1 Overview of ETS registered glass producers of container glass andtableware glass in NL (2017).

Glass production takes place through a series of processes, consisting of batch preparation, melting & fining, refining & conditioning, forming, annealing, surface treatment, and inspection. The furnace, where melting and fining of the glass takes place, accounts for the majority of energy consumption. In container glass production, this part of the process typically consumes about 75–80% of the energy. Tableware glass production is somewhat more energy intensive, where forming and moulding consume more energy, such that the relative contribution of the furnace to the energy consumption is approximately 65–70%. Combustion emissions from the furnace account for the majority of the emissions, but from the furnace also process emissions take place, which accounts for approximately 15%.

The main findings are:

- All-electric melting, as well as biofuel and hydrogen combustion are promising alternatives to traditional natural gas consuming melting activities, provided that they are developed further, and their impact on the process and the respective risks have been thoroughly investigated. These options depend on availability and sustainability of large volumes of those energy carriers as well as the presence of infrastructure and pricing conditions.
- The potential of waste heat recovery options can be further exploited by exploring their optimal use within traditional melting and coupling with innovative melting methods.
- A sound legal and regulatory framework for technology deployment is necessary for enabling substantial improvements and innovations by the container glass industry.

FULL RESULTS Introduction

This report outlines the current situation of the Dutch container glass and tableware glass production and the options and preconditions for its decarbonisation. It is part of the MIDDEN project: the Manufacturing Industry Decarbonisation Data Exchange Network. MIDDEN aims to support industry, policy makers, analysts and energy sector in their common efforts to achieve deep decarbonisation.

It addresses energy requirement and CO_2 emissions estimates, and includes a review of decarbonisation options. The focus is on the operations of two producers of container glass within the context of the Dutch industry, namely Ardagh Group and O-I, and one tableware glass producer; Libbey.

Scope

The aim of the MIDDEN project is to compile a database of material and energy uses of industrial processes at a plant level for the Dutch manufacturing industry. The scope of this research will focus specifically on the manufacturing of container glass and of tableware glass in the Netherlands¹.

Production locations include:

• Dongen (Ardagh Group), Moerdijk (Ardagh Group), Leerdam (OI Netherlands, Libbey) and Maastricht (OI Netherlands).

Processes include:

• Batch preparation, melting & fining, refining & conditioning, forming, annealing, surface treatment, and inspection.

Products include:

• Container glass for food and beverage of varied colours (i.e. flint (colourless), dark/light amber, and emerald green) and tableware glass.

The main decarbonisation options are:

• Full electrification of the industrial process, oxy/fuel furnaces coupled with waste heat recovery options (and/or syngas production), biomethane furnaces, on-site steam/electricity generation and post-/oxyfuel-combustion CCS.

Reading guide

Section 1 gives a general introduction to the glass industry in the Netherlands. In section 2 we describe the current situation of glass production processes in the Netherlands and in section 3 we describe the relevant products of these processes. Options for decarbonisation are systematically quantified and evaluated in section 4. The feasibility and requirements of those decarbonisation options are discussed in section 5.

 $^{^{\}rm 1}$ Separate MIDDEN reports are published on the glass wool and fibre glass industries in the Netherlands, see www.middenweb.nl

1 Glass production in the Netherlands

1.1 The importance of the European glass industry

The glass manufacturing industry is an energy- and capital-intensive industry which has one of the highest production volumes per capita worldwide (Eurostat, 2011). It is a rather heterogeneous sector which is responsible for the production a multitude of packaging formats for applications such as beverage, food, pharmaceuticals and personal care, and also for the production of tableware glass, fiber glass, glass wool, and some other niche applications. Nearly 62% of the European glass packaging market is held by the beer, cider and food markets, where the largest user of glass packaging is beer which accounted for 22.7% of the total glass packs used by European countries in 2018². The demand in the food sector is experiencing growth, mainly due to an increase in demand from seasonings, dressings and sauces.

Overall, European countries have experienced growth of around 2.1% CAGR³ in the period 2014–18, reaching a total of 1.043 trillion packs (i.e. rigid plastics, metal, glass etc.) in 2018 (Figure 1; PCI, 2015). Driven by a diminished demand in the beer, cider and soft-drinks industries, glass experienced a slight decrease by about 0.27% in a European level in the period 2014–2018. Globally the packaging market is expected to grow with a CAGR of 5% between 2019–2023, of which a large driver is an increase in the alcoholic beverage market in Asia Pacific (Market Research Future, 2019).



Figure 1 European packaging market by material in the period 2010–2018².

Glass-making operations in EU27 are often interlinked to location-specific economic conditions and energy polices, as well as fluctuations of fuel and electricity prices, availability of resources and supplier network in the value chain (Schmitz et al. 2011; Ecofys, 2009). About 80-90% of the process heat in European glassmaking industries are produced by means of natural gas which is the main cause of the carbon dioxide (CO₂) emissions of the industry. (Fiehl et al. 2017). The ambitious EU27 objectives associated with the significant reduction of CO₂ emissions is gradually increasing the pressure on the European glass industry for achieving deep decarbonisation by 2050.

² Canadean (2015); cited in: https://www.all4pack.com/Archives/Packaging-market-challenges-2016.

³ Compound annual growth rate (CAGR)

1.2 The Dutch glass manufacturing industry

The Dutch glass industry has been at the forefront of using the latest technologies, performing at world level in terms of quality, production and energy efficiency, and environmental performance (VNG, 2012). Typical examples are the introduction of the first oxy/fuel furnaces in Europe by O-I in 1994, the first batch and cullet preheater by Ardagh Group in 1997, and the world's first fully thermochemical-based glass melting furnace installed at Libbey (technology owned by Praxair/Linde) in 2017 (van Valburg, 2017). The Dutch glassmaking operations are highly diverse, consisting of five glassmaking sectors with 8 production sites and 19 melting furnaces. Container glass exceeds 75% of total glass production at a national level and the rest comprise glass wool, stone wool, glass fibres, tableware, and special glass production. Six companies participate in the Emissions Trading System (ETS) as part of internationally operating groups, while the flat glass production was discontinued in 2015.

The developments in the container glass industry are driven by advancements in the food and beverage industry together with the demand for glass as packaging material. Such developments are associated with improvements in manufacturing process efficiency, with an emphasis on curtailing energy use and carbon emissions (FEVE, 2016). Over the period 1992–2010, the sector achieved an energy efficiency improvement of 22%, mitigated CO₂ emissions by 10% and reduced SO₂-, dust- and NO_x-related emissions by 65–75% (VNG, 2012; FEVE, 2016). This trend of reduced energy consumption of container glass furnaces was primarily driven by factor such as furnace size, pull rates (amount of glass melted per unit of time), selected raw materials (carbonated or calcined), recycled glass content (cullet), efficiency of residual heat recovery systems (if any), level of furnace insulation and sealing, and share of electric boosting. However, the average energy use has begun to plateau (Figure 2), showing that the practical limits for energy efficiency have been reached and breakthrough technologies are needed to achieve further energy reduction towards the theoretical minimum and alternatives are needed to achieve major further CO₂ emission reduction.





Nearly 550,000 tonnes of primary raw materials are used annually in the Dutch glass production sector: silica sand (55%), limestone (20%), soda ash (17%) and other materials

(8%) (JRC Report, 2013). Recycled glass in the form of cullet (i.e. broken pieces of glass) is used in the majority of glassmaking sectors to substitute virgin raw material, among which the container and fibre glass are the products with the highest recycled content (Schmitz et al. 2011; Section 3.2). Natural gas is the dominant energy source, which is particularly used for melting, refining and annealing (see Section 2). Non-melting processes typically account for about 20–25% of the total energy demand, where a substantial share of this amount refers to electricity consumption.

Producer	Location	Main activity	Number of Furnaces	CO ₂ emissions 2017 [kt CO ₂ eq/yr]	Production capacities (packed) [kt/yr]
Ardagh	Dongen	Glass-containers for food and beverage	3	92.8	300
Glass	Moerdijk	Glass-containers for food and beverage	2	60.2	200
0-I	Leerdam	Glass-containers for beverage	3	75.0	280
Europe	Maastricht	Glass-containers for food and beverage	3	96.9	250
Libbey Glass	Leerdam	Tableware for daily use	2	27.5	40-45

Table 1 Overview of ETS registered glass producers of container glass and tableware glass in NL (2017).



[1] CO2 emissions retrieved from NEA database; Reference year 2017.

Figure 3 Overview of container glass and tableware glass production locations in the Netherlands.

1.3 Ardagh Group

Headquartered in Dublin, Ardagh Group S.A. is a glass producer which has consistently increased its scope, scale, and geographic presence in the past few decades (Ardagh Group, 2017a). The company expanded on a global level after a series of acquisitions by Yeoman International in 1998, playing a major role in the consolidation of the global metal and glass packaging industries. The company is now active in 22 countries worldwide with over a hundred facilities for beverage and can manufacturing, 33 of which manufacture 5 million tonnes of glass per year. The company employs approx. 23,500 people, half of which are employed within glass packaging activities. The company had approximately EUR 7.6 billion in full year revenue in 2017 (increased by 20% compared to 2016), where glass packaging accounted for 42% of total glass revenues in 2017, some of which include Heineken, AB InBev, and PepsiCo.

Ardagh Glass Dongen B.V. and Ardagh Glass Moerdijk B.V are two of the principal operating subsidiaries in the Netherlands. They employ approx. 600 employees in its two production facilities, which are located in key industrial areas with a favourable access to transportation network (Ardagh Group, 2017b). Established in 1961, the Dongen plant and its nine production lines account for the annual production of 1.3 billion bottles and jars. The plant includes 3 classic regenerative furnaces, two of which produce flint (colourless glass) and one that produces emerald green glass. The furnaces operate with natural gas and electric boosting, and incorporate batch and cullet preheating. The average recycling amount of glass is 76%, out of which 10% is domestic.

Located in an industrial area, Moerdijk plant was built in 1998 and contains 2 oxy/fuel furnaces with bubbling technology. The plant produces exclusively for Heineken, having a long-term supply agreement which covers the investment. In 2009, a EUR 45 million project leaded to the construction of its second furnace, which is dedicated for the production of two different sized bottles (Heineken, 2018). The investment has reduced the CO_2 footprint of the plant by 1–2% and delivered annual cost savings that exceed the initial investment. The average recycling amount of glass is 83%, steam is produced from exhaust gases for driving a cooling air fan and electric boosting will be applied shortly in both furnaces for increasing capacity by $10\%^4$.

The company's glass production facilities are regulated under the EU ETS, and hedges a portion of its carbon purchases using European Union Allowance future contracts (Ardagh Group, 2017a). Ardagh seeks to optimise the use of recycled glass in the production process, aiming for prolonging furnace life and for decreasing energy costs and carbon emissions (Ardagh Group, 2018). In the Dongen plant, the company recently installed equipment to measure CO in the exhaust gases to increase energy efficiency by heating up less air (i.e. 200ppm CO). Ardagh has been working on the development of more environmentally friendly packaging design, as well as the infinite exploitation of permanent recyclable materials.

1.4 O-I Netherlands B.V.

Owens-Illinois Inc. (known as "O-I" since 2005) is a company headquartered in Perrysburg, Ohio, US which specialises in container glass products. The global footprint of O-I spans 23 countries, employing 26,500 people at 78 plants, out of which 8,000 are employed in 11 European countries. In 2018, the company reached a market value of EUR 3.1 billion with EUR 5.97 billion revenues and EUR 8.5 billion worth of assets⁵. The company is improving its competitive advantage through investments in R&D, launching new product solutions on an

⁴ Sven-Roger Kahl, Ardagh Glass Dongen B.V., Personal communication.

⁵ Retrieved from Fortune: http://fortune.com/fortune500/owens-illinois/

annual basis. O-I distinguishes itself by the ability to maintain flexible production lines, as well as the use of lean six sigma processes for projects evaluation, priority setting and problem solving. The company is also the preferred partner for many leading food and beverage brands, including Diageo, SAB Miller, Nestle Waters and Coca-Cola.

In the Netherlands, the closure of an installation in Schiedam in August 2017 moved a part of its production to the peripheral O-I plants. The company now operates 2 facilities for container glass production with 650 employees combined, initially owned by Vereenigde Glasfabrieken and incorporated in 1995 into the French Danone Group before acquired by O-I (Wikipedia, 2019). The first bottle in Leerdam was produced in 1765. The current Leerdam's beer bottle plant has an oxy/fuel and two end-port regenerator furnaces. Regular production is 800–900 tonnes per day in 7 production lines⁶, comprising 200 different products of green, light amber and dark amber glass with a recycled content of about 75%. O-I completed a EUR 21 million investment in 2014, which included a furnace rebuild, machine upgrades and quality equipment to strengthen the plant's production and quality capabilities, targeting at a 15% decrease in CO_2 emissions (Derijck, 2013). The Maastricht plant produces approx. 1 billion jars and bottles per year, with a gradual increase in the beer bottle production (currently 30%). It operates since 1834 and the current facility includes 3 end-port regenerator furnaces for the production of flint and amber glass. Nearly 400 different products are manufactured in 9 production lines, having a recycled content of 60-65%.

The company announced its long-term sustainability goals in 2009, regarding the reduction of energy consumption and CO_2 equivalent emissions per container. According to a 2007 baseline, the average manufactured container consisted of 34% post-consumer glass and required 2.14 MJ to produce, resulting in 0.179 kg of direct and indirect CO_2 emissions. The company now aims to increase post-consumer recycled content in their products to 50% by 2025 while achieving a 25% reduction in emitted greenhouse gas by 2030 (O-I, 2019). O-I is currently using more post-consumer glass than any other glass-container maker (approx. 4.5 million tonnes), and targets at a 60% global average of containers that consist of recycled materials.

1.5 Libbey Glass

Originally founded in East Cambridge, Massachusetts in 1818 and based in Toledo, Ohio, Libbey Inc. (formerly "Libbey Glass Company") is one of the largest glassware and tableware manufacturers worldwide. The company operates six manufacturing facilities and added glassware, dinnerware and flatware to its line-up in 1997 through the acquisition of World Tableware Company. The company produces over 1.2 billion pieces consisting of 1,000 stock glassware items, while employing 6,600 people worldwide and supplying tabletop products to retail, foodservice and business-to-business customers in over 100 countries (Libbey, 2018). Libbey's retail customers include Crate & Barrel, Target and WalMart. By collaborating with technology providers such as Praxair, Libbey puts particular strain on the development of technologies for increased glass efficiency. The company is committed to reduce emissions, save energy and lessen its reliance on natural resources, aiming to align with the European roadmap towards a carbon free economy by 2050 and position its Leerdam location as sustainable and premium production tableware site (de Diego et al. 2017; 2018).

Libbey introduced partnership with Royal Leerdam in 2002, which implemented changes in its glassware furnace in May 2017 for shifting from recuperative to oxy fuel combustion. Starting from November 2017, a project partially funded by European Union with a LIFE

⁶ Joost Laven, O-I Netherlands Schiedam B.V., Personal communication.

grant (LIFE 15 CCM/NL/000121) of EUR 2.3 million aimed at the advanced heat recovery in a second oxy/fuel fired glass furnace with a capacity of over 100 tonne per day (tpd) (Praxair, 2017). The concept which is broadly known as OPTIMELT[™] Thermochemical Regenerator (Optimelt TCR) takes advantage of the high temperature flue gas using a regenerative system and reforms natural gas into hot syngas (Section 4.3). It is reported that fuel consumption reaches 440–480 m³/hr for glass pull 65–85 tpd (de Diego et al. 2018). A vacuum pressure swing adsorption (VPSA) system by Praxair is used for the on-site oxygen production of about 56 tonnes per day at Libbey's facility (de Diego et al. 2016). Royal Leerdam glassware now offers a broad range of glass tableware products for the retail market, the foodservice market, next to a Libbey-branded glassware range.

The Leerdam plant emitted around 25–30 kt of CO_2 in 2017. The company set the goal of reduced energy consumption and CO_2 emissions of 45–60% and annual NO_x emissions by 25–35% compared to its past oxy/fuel installation and in line with Low- NO_x burners for glass (Herbst et al. 2017; Laux et al. 2016). The first reductions in air emissions were reported in 2018 after upgrading to Optimelt TCR. Given the high potential in energy savings of the Optimelt technology compared to other existing options for using waste heat, the company targets at the exploitation of residual heat in percentages greater than 20%. This would enable the application of additional energy efficiency measures such as oxygen, natural preheating or batch/cullet preheating with subsequent benefits of reduced CO_2 emissions. Lastly, the goal of increasing the recycled glass content final products to over 50% would allow further reductions in process emissions but requires higher cullet quality from the market.

2 Manufacturing processes of glass

Glass manufacturing involves the conversion of raw materials into finished products that fulfil certain requirements (Chang, 1995). This conversion is accomplished using a great variety of activities that apply energy (i.e. mechanical, thermal, electrical, or chemical) to produce controlled changes in the configuration properties of materials (NAP, 1995). However, industry emissions are intrinsically linked to the production process due to those activities (CAT, 2017). Therefore, CO₂ and energy efficiencies need to be assessed in light of the melting techniques which are applied, as well as the existing infrastructure, fuel input, and glass characteristics.



2.1 Container glass production

Figure 4 Process activities of glass-container production (Emhart Glass, 2012).

The production of container glass can be roughly subdivided into seven process steps: batch preparation, melting & fining, refining & conditioning, forming & moulding, annealing, surface treatment and inspection which are further described in the following Sections. Most of the energy is consumed in the melting of the heterogeneous mix up to the conditioning step. This part is assumed to be responsible for about 88% of the total energy consumption of the plant.

The methodology that we apply to estimate the melting heat starts by determining the energy requirement for the fusion of input materials in quantities which are retrieved from publicly available reports. That way, the heat released in the combustion space can be obtained by subtracting the melting heat, the enthalpy exchange of input materials and the heat losses through combustion space and glass tank refractory from the enthalpies of fuel and flue gases (Koshmanesh et al. 2007). Heat losses can be very furnace specific, but in our estimate any unknown values are initially filled with industry averages. Depending on energy recovery options and/or electric boosting applied in the process as well as the oxidant used for the combustion of fuel, the respective temperature windows of flame, furnace and flue gases are extracted and an estimate for the energy use is obtained. A reduction of the energy required, because of energy recovery options or the application of electricity for part of the energy requirement, results in a reduction of the amount of fuel demanded, which in turn requires less heating of air/oxygen for combustion. To achieve an estimate of energy consumption of the glass melting process, this calculation is done in an iterative way, illustrated in Figure 5. Values are expressed per tonne of molten glass when related to the

furnace, and expressed per tonne of packed glass when referring to the glassmaking process as whole⁷.



Figure 5 Method followed for estimating specific energy consumption of furnaces.

These more detailed calculations for specific plant types and glass types were performed and are presented in Appendices B and C. The generic process flow chart in Figure 6 is deemed representative for the average Dutch container glass manufacturing process.

⁷ Division of tonnes of molten glass by the inverse of internal cullet rate (i.e. pack-to-melt ratio; Section 3.2).



Figure 6 Flowchart of the manufacturing process of container glass⁸.

⁸ Regenerative furnace; Natural gas to total fuel input ratio 92%, indirect emissions factor 0.5kg/kWh. Formulas applied from Madivate (1998); normalised for 1 tonne of packed glass.

2.1.1 Batch preparation

The glass melting process starts from a granular mixture of natural and/or synthetic raw materials (i.e. batch). Soda-lime-silica glass is usually made by melting batches of silica sand (SiO_2) , soda ash (Na_2CO_3) , limestone $(CaCO_3)$ and dolomite $(CaMg(CO_3)_2)$. Alumina carriers (Al_2O_3) are added as stabilisers to enhance the chemical durability, while fining agents such as sodium sulphate are added to promote the removal of bubbles from the melt (Hujova et al. 2017). Small quantities of other additives are included to give desired characteristics to individual glasses (Ecofys, 2009). Recycled glass is used in variable quantities and can be found in proportions as high as 60–90% for external and 5–25% for internal cullet which originates from production losses. The materials are fed into the weighing area, carefully sorted and mixed according to a precise formula to form to a homogenous composition.

Variations in batch compositions are used among glass-making companies which ultimately define the colour (i.e. flint, dark/light amber, and emerald green; see Appendix C) and other desired characteristics of the produced container glass (see Chapter 3). Depending on the demand for glass and location-specific type of production, the plant may switch to other products for about 5–9 times per week⁹. The operation requires electricity as primary energy source and accounts for approximately 1.0–1.5% of total energy demand (IETD, 2018), but this may depend on the glass type.

Parameter	Amount	Unit per tonne of packed glass	Comments
Input			
Electricity	0.05	GJ	Assumed to be 1% of total energy use
Silica sand	190-195	kg	Table C1; Average of three glass types
Soda ash	50-58	kg	Table C1; Average of three glass types
Limestone	25-35	kg	Table C1; Average of three glass types
Dolomite	20-35	kg	Table C1; Average of three glass types
Sodium sulphate	1-2	kg	Table C1; Average of three glass types
External cullet	733	kg	Assumed 66% external cullet
Domestic cullet	111	kg	Assumed 10% internal cullet
Output			
Indirect CO ₂	8	kg	Emission factor 0.5 kg CO ₂ per kWh (WEC Report, 2018)
Batch mix	1152	kg	Heterogeneous glass mix

Table 2 Batch preparation energy & material requirements.

2.1.2 Melting & fining

For transforming the batch and cullet mixture into molten glass, furnaces employ combustion heating (e.g. air/oxy-fuel burners), direct electrical heating or their combinations (e.g. electric boosting; see Appendix A). The melting accounts for about 75–80% of total energy

⁹ Joost Laven, O-I Netherlands Schiedam B.V., Personal communication.

use (i.e. 4.6 GJ/t packed glass), typically 50% of which is dispersed in the exhaust gases and in structural losses depending on the degree of insulation, sealing and heat recovery options (Tapasa et al. 2012; see Section 4.3). The majority of total heat is absorbed by the raw materials mixture during the first 0.75–1 hour, while the molten glass resides in the furnace between 8–32 hours depending on the melting technique, furnace capacity and glass quality standards. Process emissions occur due to carbonate decomposition in the process input and account for 10–15% of total direct emissions (Ecofys, 2009). Glass production is a continuous process and the furnace operates for many years at a time, as long as its lifetime allows for. Downtime for furnace maintenance typically lasts 35–40 days.

Melting of raw materials, homogenisation and fining takes place in the melting end of the tank. After the glass went through a narrow channel called throat, the refining, final homogenization and conditioning of the glass takes place in a refiner and forehearth. The cold batch is heated by electrical heating and mainly by fuel-burning systems until batch-to-melt conversion takes place between 850–1200°C. The resulting glass melt contains bubbles and is further heated to temperatures of maximum 1550°C depending on glass specifications (i.e. colour, quality specifications, etc.). The mixture is continuously charged into the furnace to compensate for the glass withdrawn and keep the glass level in the furnace constant (IETD, 2018).

Depending on location-specific strategies by container glass companies, dolomite may be used in the batch mixture. The decomposition of calcium carbonate occurs in a two-stage process (Olszak-Humienik et al. 2015):

$$CaMg(CO_3)_2(s) + Heat \Rightarrow CaCO_3(s) + MgO(s) + CO_2(g), \Delta H_f=-1780 kJ/(kg CaCO_3)$$
 (1)

$$CaCO_{3}(s) + Heat \Rightarrow CaO(s) + CO_{2}(g), \Delta H_{f} = -3303 \text{ kJ/(kg CaO)}$$
(2)

Sodium carbonate serves as a flux for silica, enabling a decrease of the melting point of the mixture hence decreasing the residence time of the melt in the furnace (Hubert, 2015). Sodium carbonate requires lot of heat to completely decompose:

$$Na_2CO_3(s) + Heat \Rightarrow Na_2O(s) + CO_2(g), \Delta H_f=-3576 kJ/(kg Na_2O)$$
 (3)

Decomposition of carbonates occurs between 600–900°C or even lower when dolomite is used while the addition of cullet can accelerate the melting of the mixture (Deng et al. 2018). The mix starts exhibiting liquid characteristics in temperatures around 800–850°C. Gases are released from fining agents across all temperature trajectories of the glass melt. Chemical homogenization is achieved in the bulk of the glass melt in the glass melting tank and further downstream¹⁰.

¹⁰ Oscar Verheijen, Celsian Glass and Solar B.V., Personal communication.



Temperature [°C]



2.1.3 Refining & conditioning

The other separated section of the tank is the refiner (i.e. refining end) in which the melt starts to cool and to condition toward its uniform working viscosity. An initially bubble-rich melt is produced by CO₂ from carbonate-containing batch ingredients, together with water and air trapped in the interstices of the raw material batch (De Jong et al. 2011). After the fining process completes, the remaining bubbles require the refining process, taking place under controlled temperatures, starting from 1550°C and dropping down to 1400°C where the solubility of the gases in the melt increases with decreasing temperature (Hubert, 2015; Butler et al. 2011). The refining time typically lasts 60–90 minutes and can influenced by a number of factors such as viscosity, batch composition, sulphate used and melting time (Watts, 2017).

The bubble-free molten glass is then distributed to a forehearth, a brick lined canal which delivers glass to the forming machine. The forehearth can be gas-fired or electrically heated, and its performance is determined by the range of pull rates (i.e. amount of molten glass produced), range of temperatures where the production maintains an acceptable degree of homogeneity, and its ability to maintain temperature stability (Beerkens, 2012). The outcome is a thermally and chemically homogeneous melt made available at the defined temperature level (Conradt, 2008). Energy required in both activities is 6–7% of the total consumption (JRC Report, 2013).

2.1.4 Forming & moulding

The conditioned glass exits the melting tank and is transported to forming machines at a constant rate, where a plunger pushes it down through a narrow tube (Emhart Glass, 2012). As the stream of glass emerges from the tube, it is cut off using shears and turned into gobs of molten glass (i.e. even shapes which can be moulded). This takes place in 8–20 machines called Individual Sections (IS) which can be independently maintained. The forming machines give shape to the container glass through automated processes known as pressblow (mainly for jars) and blow-blow (mainly for bottles). By pressing the sheared gob into a blank mould using compressed air, the gob gets its intermediate shape (i.e. parison) which is then moved from the blank side to the blow side for final blowing and shaping (Figure 8). The energy used in forming containers is about 5–6% of the total energy use, and depends largely on the electricity for compressed air and the weight of the container.



Blank side

Blow side

Figure 8 Blow-blowing process for container glass forming (Qorpak, 2019).

2.1.5 Annealing

Once the blown containers are formed, they are loaded through a conveyor belt into a lehr for thermal treatment (i.e. annealing). Their temperature is rapidly brought back up to approx. 580°C, then reduced gradually and uniformly until no further strain can be induced (IETD, 2018). The product is cooled afterwards by fan air to room temperature. Without this activity, which accounts for approx. 4–5% of the total energy consumption, the glass would be low-resistant to cracks under small thermal or mechanical shocks (Hubert, 2015). The duration of the activity is important, both for the successful relieving of the glass from its internal stresses and for the reduction of the costs associated with the energy requirement. Depending on the product type (i.e. size and wall thickness), a 15-minute residence is required close to the annealing heating point, followed by a 30–60 minute residence close to a higher, strain point. Emerging from the lehr, the container glass is fully cooled and conveyed to further steps.

2.1.6 Surface treatment

The coating of glass surfaces gives to the glass new physical and optical properties. Glass containers are coated with organic compounds to give the surfaces a degree of lubricity, thus preventing abrasion in handling (IETD, 2018). The two main types are stearate- and polyethylene-based coating, which add strength to the container, facilitates the application of labels and enables glass manufacturers to make lightweight products (Worrell et al. 2008). In contrast, the Internal Fluorination Treatment (IFT) is the process that makes soda-lime glass to treated soda-lime glass and is applied to prevent blooming (Qorpak, 2019). The energy required for post treatment is about 0.3–0.8% of the total energy use (Beerkens, 2012).

2.1.7 Inspection

Before the shipping of the final product, further activities include the Inspection, which uses fully automated devices to eliminate defective containers, and the Packaging of glass for transportation to the client's factory (Meuleman, 2017). It includes measurements in terms of height, diameter, verticality, choke and thickness of containers. Critical defects are discarded as dangerous, while main-faults defects make the container unusable. Secondary defects represent a lowering of the quality of the container but do not affect the functionality of the container. As a result, the inspection contributes to the plant's domestic cullet, sending rejected glass for re-melting.

Parameter	Amount	Unit per tonne of packed glass	Comments
Input			
Natural gas	0.54	GJ	Needed for annealing
Electricity	0.48	GJ	Needed for forming, annealing, surface treatment & inspection
Molten glass	1.1	t	Glass melt exiting the furnace
Output			
Direct CO ₂	30	kg	Emitted from annealing lehr
Indirect CO ₂	61	kg	Emission factor 0.46 kg CO ₂ per kWh (Schoots et al., 2017)
Glass product	1000	kg	Packed glass

Table 3 Post-melting energy & material requirements.

2.1.8 Overall container glass-making process

The total energy consumption of a container glass plant is estimated at 5.8–6.5 GJ per tonne of packed glass and depends on the glass colour and amount of furnaces and articles produced¹¹. Energy is coming from both gas and electrical boosting and electricity is normally used in rates 8–15% of the total melting energy (Figure 6; see Appendix A). Figures 9 and 10 illustrate the energy consumption and related direct and indirect emission for the various subprocesses.



Figure 9 Energy input per subprocess for the production of container glass.

¹¹ Joost Laven, O-I Netherlands Schiedam B.V., Personal communication.



Figure 10 Total direct and indirect CO₂ emitted per industrial glass-making subprocess.

Furnace designs are bound to the plant's space, while the investment decision on a furnace type is mostly affected by the quality of the product, stability and lifetime. The frequency of furnace maintenance as well as the future of a furnace which approaches its end of life (approx. 15 years) depends on the company strategy. An indication for the latter would be a steep increase of energy consumption or the considerable wear of refractories which increases the risk of unexpected leaks.

The otherwise lost residual heat is taken as heating medium from the waste gas channel of the melting end. Waste heat recovery options are interlinked to the furnace type (i.e. air/fuel, oxy/fuel) as well as the temperature and purity of the flue gases exiting the furnace or the regenerator (see Appendix A; Section 4.3). The recovered heat from flue gas is about 60–68% depending on the heat recovery system, which helps to maintain the primary energy input for melting at the levels of 4.5–4.8 GJ per tonne of glass. A heat recovery system of high efficiency results in residual gases of low heat content which prevents a further use of the exiting gases. Options that do not use the residual heat directly back to the melting process may be more preferable for process control reasons (see Section 4.6), but may also require the cleaning of flue gases and also stay above 200°C to avoid sulphuric acid (e.g. preheating natural gas for annealing).

Flue gas Treatment

Due to organic contamination in high cullet-containing batches as well as the sulphur originating from fining agents, the exhaust gases are often cleaned in an electrostatic filter when exiting the furnace and after entering a regenerator. Desulphurisation and normal operation of the filter takes place in temperatures between 250–380°C. Pollutant-abatement systems may be installed after the regenerator for cleaning this stream of process and combustion emissions (e.g. deSOx and deNOx).

The furnace can be sealed using a baghouse filter, an air pollution control device and dust collector which is attached to the furnace. As a consequence, if a regenerative furnace is connected to a baghouse filter it is mandatory to cool the flue gasses, because filter bags only withstand 220°C (TNO, 2010). Waste gas cooling takes place in a quench chamber that is operated either with air or water, or a combination of both (Sorg, 2011). If the flue gases are

cooled down a lot, the cleaning system of flue gases may not work properly (HREII, 2018). For sulphur removal, scrubbing takes place at a temperature of around 350°C and it is filtered at around 200°C. Below this temperature range, undesired sulphuric acid is formed.

Tableware glass production

Royal Leerdam is originally specialised in stemware and uses a production technology which is partly differentiated from container glass making. The furnace currently in use to produce tableware glass has been constructed in 2018 and is of the innovative Optimelt type. The Optimelt Thermochemical Regenerator (Optimelt TCR) takes advantage of the high temperature flue gas using a regenerative system and uses 20% of the flue gas together with natural gas to make a hot syngas in the right mixture (Section 4.3). It uses oxygen rather than normal air for combustion, and a vacuum pressure swing adsorption (VPSA) system by Praxair is used for the on-site oxygen production of about 56 tonnes per day at Libbey's facility (de Diego et al. 2016). The remaining flue gas is of insufficient volume to use for additional preheating purposes, given the oxy/fuel technique and size of the furnace.

Compared to container glass production, the energy consumption of the forming and moulding fase of tableware glass production takes significantly more energy. Whereas for container glass production the furnace consumes 80% of the total energy/t packed glass, for tableware glass this is only 65–70%, with the additional 15% going to forming and moulding. Typically, cullet rates that can be used are lower because of the quality required for tableware glass. Only internal cullet is currently used in the process, as there is no tableware cullet available in the cullet market.

Special thermal processes are followed downstream the melting with the prospects to produce durable glassware with prolonged service life. Duratuff[©] Treatment and Heat Treat are the activities which increase the glass resistance to thermal and mechanical shocks by strengthening the upper portion of pressed tumblers and blown ware, respectively. By treating just the upper third of the glass at a lower temperature puts a safer amount of stress on the glass and positively affects the way that glass breaks (i.e. smaller jagged pieces). Other activities include SheerRim/DTE® that features a crack-off rim which is fire polished to smooth the edge and improve durability¹². In addition, one-piece stemware is made from one continuous piece of glass which is stretched in a second machine process, while two-piece glassware has the bowl made in one machine and the stem and foot in a second one.

The production of container and tableware glass are among the processes which allow product light-weighting as an effective means to reduce environmental impacts (IFC, 2007). This can be done by increasing the product count that can be manufactured from a given weight of molten glass (see Section 3.1).

¹² Retrieved from Libbey website: <u>https://b2b.libbey.com/solutions/after-process/</u>

3 Glass products and application

This chapter consists of an overview of the main materials used in the glass-making process, as well as physical/chemical/economic/environmental properties of the respective compounds. Emphasis is also given on the recycling of glass products and the benefits associated with their use. Aside from container glass, grades of commercial glass products include:

- **E-glass** (CaO.Al₂O₃.B₂O₃.SiO₂) is the glass type which is known for its low electrical conductivity and mechanical strength. Its properties makes it a good fit for the production of optical products such as fibre glass for electronics. Variations of this type may replace B₂O₃ with MgO or TiO₂.
- **A-glass** (Na₂O.CaO.B₂O₃.SiO₂) is a low-cost type of glass which is mostly known for its fiberisability. This makes it applicable for insulation (i.e. glass wool) and reinforcement purposes (i.e. fibre glass).
- **Borosilicate Glass** (Na₂O.B₂O₃.SiO₂) is known as for its laboratory use (e.g. Pyrex, Duran), as well as for the production of cooking utensils and headlights. Its resistance to thermal shocks is among the most important properties.
- **Quartz** (SiO₂) has a high-temperature and thermal-shock resistance, making it useful for telecommunications (i.e. optical glass fibres), as well as for laboratory and chemical equipment. Although pure silica can be made into high quality glass (i.e. vitreous silica), this would require the batch to be heated in temperatures far above 1700°C.

3.1 Container glass in the Netherlands

Depending on the batch mixing formulas, variations in the chemical compositions give desired characteristics and properties to the glass product (Qorpak, 2019). Soda Lime Glass (Na₂O.CaO.SiO₂) is commonly used for the production of container, flat and tableware glass, as well as lenses and lighting. Annealed and tempered soda-lime are two of its grades, which show differences in terms of mechanical, thermal and electrical properties¹³. Flint and emerald green consist of oxidised glasses, while light/dark amber of are known as "reduced" glasses containing high level of carbon (GPI, 2019). The low-cost feedstock allows the mass production of this type.

An increase of sodium oxide (Na_2O) content may decrease the viscosity of the glass product, which is a measure of its resistance to flow and its tendency to prevent crystallisation. Calcium oxide (CaO) improves the chemical resistance of glass, while an increased content of aluminium oxide (Al_2O_3) makes a favourable influence on the tensile strength (Figure 11).

¹³ For analytical comparisons, visit <u>https://www.makeitfrom.com/material-properties/Annealed-Soda-Lime-Glass</u>



Figure 11 Relation between oxides and effect on glass properties (Hubert, 2015).

Applications of Container glass

Glass products contribute in a major way to society's sustainability goals for the reduction of energy use, renewable energy production and efficient use of resources (WBG, 2004). At the same time, the production meets the strictest requirements in food and other critical contacts, responding to growing marketplace and the high levels of customer demand for more sustainable packaging (PB & DNV GL, 2015). Container glass manufacturers provide with a comprehensive line of stock products for the following categories:

- Beer, wine, spirits, specialty, non-alcoholic beverages (i.e. bottles) 75%;
- Food (i.e. jars, bowls, plates, cups), pitchers, vases 20%;
- Pharmaceuticals, chemicals, cosmetics (i.e. flacons), and personal care 5%.

Production of Container glass

Being dependent on sales of food and beverages, the demand for glass containers does not fluctuate greatly with business cycles compared to flat glass demand (Butler et al. 2011). Yearly production of glass containers in Europe is projected to reach 17.74 million metric tonnes (t) by 2020 with a compound annual growth rate of $3.32\%^{14}$. In the Netherlands, the organic growth of container glass industry is estimated at 1.0-1.5% per year¹⁵. Despite the closing down of the float glass facility by AGC, the total production capacity of 1.26 million tonnes reduced by only 0.5% in 2015 compared to 2012 (van Valburg, 2017). Apart from the anticipated process enhancements (see Section 1.3.2; 1.3.3), no expansion investment plans have been publicly announced by the Dutch glassmakers.

Market value

The light-weighting industry trend, as well as the growth of end use industries have been some of the main drivers of the glass market (Hatzilau et al. 2016). At the same time, gas and electricity prices and the competition from alternative materials play a role in shaping price levels in the glass industry. Depending on market preferences, costs and packaging developments, the position of glass relative to its competitors varies widely between regions and products. Such competitors can be plastics (e.g. PET-polyethylene), metals (e.g. steel and aluminium) and laminated cartons (JRC Report, 2013).

Glass furnaces that use significant electric boosting and contain a large amount of molten glass could be able to accommodate some flexibility needed to profit from rewards, grants

¹⁴ Retrieved from the Ceramic Industry: <u>https://www.ceramicindustry.com/articles/95243-glass-container-market-expected-to-see-steady-growth-in-europe-through-2020</u>

¹⁵ Sven-Roger Kahl, Ardagh Glass Dongen B.V., Personal communication.

and lower electrical tariffs (Eurotherm, 2019). Lower peak power demand can lead to lower tariffs, therefore the overall cost of electrical energy can be decreased by ensuring that agreed peak power levels are not exceeded (Eurotherm, 2019). Glass container manufacturers have a scope to increase their margins by joining forces with glass recycling companies to develop efficient glass collecting practices¹⁶. According to CE Delft (2016), the opportunity costs of the freely obtained ETS allowances were forwarded in glass-container product prices to a lesser extent than other glass products (i.e. 20–50%)¹⁷.

3.2 Recycling of glass

Role of recycled glass in the glass industry

The container glass sector draws the vast majority of recycled glass, which comes from postconsumer glass or glass processing plants (Drummond, 2011). The recycled glass content of the total heterogeneous glass mix varies between 50–80% for external recycled cullet and 10% for internally recycled glass and depends on the requested glass quality and operating constraints (e.g. product changes per year)¹⁸. By partly replacing a batch of mineral raw materials with recycled glass, the facility reduces:

- the use of heat for the endothermic chemical reactions between batch components during the glass formation by the same rate (De Jong et al. 2011);
- the fuel consumption by around 2.5% for every 10% addition of cullet, for typically used cullet ranges, purely calculated as melting energy saved (Madivate, 1998; Drummond, 2011; Butler et al. 2011). For tableware the energy savings are somewhat lower (1.5–2% per 10% cullet addition).
- the demand for raw materials, among which the most significant are carbonates such as soda ash, limestone and dolomite that decompose in the furnace releasing CO₂. In principle, the addition of 1 tonne of recycled glass leads to a reduction of 1.2 tonnes of primary raw materials, along with consequent cuts in process emissions (JRC Report, 2013).

Recycling rate of glass in the Netherlands

The Netherlands is amongst the best performing countries in Europe in the field of recycling waste, setting higher collection targets for packaging glass than its European counterparts (KIDV, 2016). At the same time, the glass recycling landscape is evolving and there is a lot of public attention for further increasing the return of container glass from household residual waste to the glass industry. In 2013, the amount of packaging material introduced to the Dutch market per capita was 167 kg and the total percentage for collected packaging waste was 94% (Eurostat, 2016, cited in KIDV, 2016). In 2015, 83% of the consumed 492.000 glass tonnes were collected, resulting in a gradual increase of 3% compared to the previous year¹⁹.

National recycling rate (%) =
$$\frac{\text{National tonnage of cullet collected (tonnes) x 100}}{\text{National glass consumption}^{20} (in tonnes)}$$
 (1)

Headquartered in Eindhoven, Maltha Glass Recycling is responsible for the recycling of glass containers in the Netherlands. The main challenge for environmentally and cost-effective recycling of container glass arises from the dispersed nature of its sources, and the

¹⁶ Retrieved from Businesswire: <u>https://www.businesswire.com/news/home/20180514005833/en/Glass-</u> Packaging-Market-Europe---Compatibility-Glass

¹⁷ For hollow glasses, the range of average expected cost pass-through is 30–80% (CE Delft, 2016).

¹⁸ Joost Laven, O-I Netherlands Schiedam B.V., Personal communication.

¹⁹ Data on glass container collection for recycling (FEVE, 2015).

²⁰ The national consumption of glass containers is equal to the national production of all glass containers (incl. returnable glass containers) plus the net imports and exports (unfilled and filled).

consequent need for infrastructure regarding its colour separation, identification and removal of glass-ceramics²¹, collection, and transportation to processors to produce furnace-ready feedstock (Butler et al. 2011; VNG, 2012).

Glass waste and disposal

The cullet ratio depends on glass colour, level of impurities and market availability (Glusing, 2003). In theory, container glass can be made from 100% cullet and can be endlessly recycled without losing its intrinsic properties (FEVE, 2016). However, there are practical limitations to this approach. Firstly, the marginal environmental and financial burdens of collecting too much post-consumer waste may exceed the marginal benefits (Butler et al. 2011). Secondly, at least 1% of batch composition is typically used for controlling the colour and fining of the glass. By melting a cullet-only batch, the final product would contain lots of gaseous inclusions called "blisters", which can be released by heating the glass at high temperature for a long time²⁵. Bubble removal takes place by adding fining agents to initiate the fining process (approx. 0.2–0.55% of total amount of glass) and to neutralize contamination from recycled cullet.



Figure 12 Life cycle of primary/secondary material for container glass²².

The industrial process does not create residual waste glass. Everything is reused as domestic cullet, with the exception of some very contaminated glass which were disregarded during quality check (approx. 25 t rejected every 6 years). In the Netherlands, no recycled cullet is land filled as waste. Roughly 70–75% can be used in the CG industry, the rest is dismissed for quality reasons and used in other applications. The reason behind this is the likelihood of problems associated with foaming, bubbles and refining when using low-quality cullet. This happens due to organic contamination caused by plastic labels, misplaced items which contaminate the entire load, or crushed cullet of extremely small particle size²³.

²¹ Oscar Verheijen, Celsian Glass and Solar B.V., Personal communication.

²² VDI (2015), cited in Anglarill, A.G. (2018). Life cycle assessment of a conventional float glass production and comparison with regenerative alternatives.

²³ Sven-Roger Kahl, Ardagh Glass Dongen B.V., Personal communication.

4 Options for decarbonisation

Developments associated with increased recycled cullet, improved furnace design and combustion control, improved insulation, and more effective regenerators have led to significant energy efficiency and emission improvements in the glass-container industry (Hatzilau et al. 2016). A transition to innovative methods is needed to unlock new possibilities for reducing combustion-related CO₂ emissions, making a step forward in the aftermath of improving energy efficiency (HRE, 2018; Meuleman, 2017). After industry consultation and literature review, the main decarbonisation options discussed in this report are air/fuel, oxy/fuel and electric furnaces. These options may go hand in hand with options such as alternative energy carriers, waste heat recovery, batch reformulation and innovative processes (see also Appendix A). We categorise the various decarbonisation options according to a framework laid out in Figure 13, in which seven categories are distinguished.



Figure 13 Categories of emission-reduction options in the full production chain.

An overview of potential options and measures is shown in Table 4, where the technologies and their characteristics are summarised. Their level of development is indicated by the "Technology readiness level" (TRL), which ranges from 1 for basic technology research to 9 for successful end-use operation. The costs of CO₂-recuding technologies are related to annual production capacities (or emissions in the case of CCS), and ranges may be provided for Capex when data are partly over- or underestimated. Note that the costs only include onsite costs and exclude any costs associated with changes outside of the fence (such as heavier electricity infrastructure, or heat transport infrastructure. The maximum emissions reductions refer to combustion emissions, process emissions are excluded from this table, and energy savings are expressed in tonnes of glass product.

Table 4 Basic and innovative technology options register for CO₂ and energy reductions in the Container glass industry.

Option	TRL	Market Entry	Capex (EUR million) ^{24, 25}	CO ₂ (C) - Energy (E) savings ²⁶	Data Source
Fuel Substitution: Substitution of the cu	irrently	used fossil	fuels		
Full electrification ²⁷	6-7	2030	0.5 /kt glass	100% (C) 15–25% (E)	Fraunhofer (2019a)
Green gas instead of natural gas	8	2020	-	100% (C) - (E)	PB & DNV GL (2015b Fraunhofer (2019a)
Hydrogen combustion	4	-		75–84% (C) - (E)	WEC Report (2018)
Process Design: Energy efficiency imp	roveme	nts and/or	substitution of production p	rocesses	
Electric furnace	7	2030	(3.5–5 per MW installed (typical size 10 MW furnace) (tableware 5–6 MW)	100% (C) 15–25% (E)	Reynolds (2019) PB & DNV GL (2015) Meuleman (2017)
Oxy-fuel furnace	9	Present	15	8–10% (C) - (E)	Sundaram (2016) Baukal (2013)
Batch/cullet preheating	9	Present	3-4	15% (C) 10-20% (E) ²⁸	Wallenberger (2010) PB & DNV GL (2015)
Electricity generation	8	2020	1.7-4.9	- (C) 5% (E)	Forni et al. (2014) Ricardo-AEA (2013) PB & DNV GL (2015)
Optimelt TCR Plus	7	2025	15+3.5-4.5 for 100 kt/yr furnace	15-25% (C) 15-25% (E) ²⁹	de Diego (2016)
Residual Energy us Ways of heat exploita	e: ition suc	ch as stear	n and/or electricity generatio	on	
Building heating	8	2020	1.0-2.1 ³⁰	1-2% (C) 1-2% (E)	PB & DNV GL (2015b IETD (2018)
District heating	8	2020	1.0-2.1	- (C) - (E)	Forni et al. (2014) Ricardo-AEA (2013)
Carbon Capture and Process of capturing a	d Utilis a and stor	ation and ring CO ₂ o	/or Storage: r alternative uses of the pote	entially emitted gas	ses

²⁴ Values expressed in rates of the year of respective sources.

²⁵ Only on-site costs taken into account. Out-of-fence costs are not in scope of this report.

²³ Only on-site costs taken into account. Out-of-fence costs are not in scope of this report.
²⁶ Values expressed as CO₂ from combustion and energy savings per tonnes of glass product compared to a state-of-art end-port regenerative furnace of 4GJ/t glass SEC, 220 kg/t glass direct CO₂, 76% cullet, 92% natural gas.
²⁷ For container glass production – for tableware glass forming&moulding so far has no electrification option.
²⁸ Fuel savings for cullet preheaters are 6% less compared to batch/cullet preheaters (Baukal, 2013).
²⁹ Approx. 20% compared to oxy/fuel; approx. 30% compared to air/fuel furnace.
³⁰ For 54% adoption rate (PB & DNV GL, 2015).

Carbon Capture ³¹	5-7 ³²	2030	50 EUR/kt packed glass	75% (C) - (E)	PB & DNV GL (2015t Fraunhofer (2019a)
				(-)	VNPI (2018)

4.1 Fuel substitution

The fuel predominantly used in the container glass industry is natural gas, which accounts for about 90% of the fuel mix. Fuel switching is challenging, as it may impose changes in operating permits and adjustment to installations such as new burners and control systems. The goal of fuel substitution is to rely on fuels with a lower carbon footprint but, at the same time, keep the same general manufacturing processes (Fraunhofer, 2019a). According to HRE (2018, Table 10), the diffusion level of the fuel switch option is estimated to be 88% by 2030 and approx. 99% in the horizon of 2050.

Full Electrification

The substitution of fossil fuel input with electricity typically refers to the use of electric furnaces (Section 4.3) or electric boosting in the melting process and the electrification of downstream activities (i.e. fining and annealing). This measure is interlinked to the glass quality specifications, as well as economic (i.e. electricity price, investment cost) and physical conditions (e.g. missing infrastructure, position of electrodes into the furnace) (Reynolds, 2018). All-electric solutions are applied in some niche glass manufacturing applications. However, significant furnace innovation is required for its adoption in high-scale container glass production, especially in the case of high cullet rates. The process prerequisites a steady supply of great amounts of electricity which requires additional reinforcement for the local grid (Fraunhofer, 2019a). The post-melting process of container glass is regarded to be suitable for electrification without substantial need for innovation. All-electric options can have emissions benefits provided the electricity comes from a renewable source (PB & DNV GL, 2015).

Green gas (biomethane) combustion

Green gas can potentially be used for glassmaking purposes, given the similar energy content of biomethane to natural gas. Green gas can be either upgraded biogas from anaerobic digestion or cleaned syngas from gasification of biomass, being 100% renewable (EBA, 2013). Using this fuel as a replacement source of heat would bring carbon emissions to net zero, as biomethane lifecycle absorbs the emitted CO₂ during production (Fraunhofer, 2019a). Material testing indicated that there were no negative consequences of biomethane firing for the material properties (Fiehl et al. 2017). However, the durability of the refractory of industrial furnaces as well as trace contaminations found in the biogas may pose additional challenges. Other concerns include the availability and sustainable production of biogas, as well as "the existence of a reliable and upgraded supply" (Fraunhofer, 2019a). Capital costs would not be incurred by switching to this fuel, provided that the system runs on gas burners (Fraunhofer, 2019a), so CAPEX estimates are identical to similar furnace types running on natural gas. It is assumed green gas is of the same quality as natural gas and can be directly substituted in equal amounts.

Hydrogen combustion

Hydrogen is often produced on an industrial scale using a steam methane reformer (SMR) or through a non-catalytic reforming process (see Section 4.3). Despite the evident benefits of this carbon-free fuel (WEC Report, 2018), hydrogen does not have a proven applicability in

 $^{^{31}}$ All costs are for CO₂ capture alone, including CO₂ purification and compression. Costs associated with transport and storage/utilisation are excluded (PB & DNV GL, 2015). Assumed that 50 EUR/t packed glass corresponds to 45 EUR/t melt. 75% of total emissions are avoided, which is 75% of 0.29 t CO2 (including post-melting process) in reference technology. Assumed 30% energy penalty, of which 60% natural gas and rest electricity. Cost per tonne CO₂ captured is then EUR 174.

³² For oxy-combustion capture TRL 6; for post-combustion TRL 7.

the container glass industry. Water vapour which is released from a complete hydrogen combustion creates foam and affects energy consumption, efficiency on the yield, and integrity of the furnace (PB & DNV GL, 2015). At the same time, hydrogen emits sub-optimally in the required wavelength compared to the heavy-fuel air flame of natural gas³³, which may change the efficiency.

Current research examines whether pure hydrogen or a blend with fossil energy achieve better radiative properties of the flame (e.g. by spraying biodiesel in a hydrogen flame), hence qualify for container glass making. This would allow a stepwise increase of hydrogen and make the process more renewable, but would also impact on the process (e.g. different burners and control schemes). The future availability and price of hydrogen also guides investment decisions, while its on-site production requires energy which can originate from fossil fuels or renewable sources. For a discussion of the various production processes of hydrogen, with their energy requirements and related emissions, can be found in Cioli et al. (2019).

4.2 Feedstock substitution

Batch reformulation

Decarbonising the existing batch can be achieved through the use of low-carbon materials for replacing currently used carbonates. For instance, process emissions can be potentially reduced by replacing limestone and soda ash with burnt lime and sodium-containing raw materials, respectively (VNG, 2012; Wallenberger, 2010). However, this alternative would transfer rather than combat the target of reducing CO_2 emissions to another sector. As to the drawbacks of this option, the quality of final product remains uncertain under current furnace designs, accompanied with an increase in material costs, transportation and chemical preparation (PB & DNV GL, 2015).

Alternatively, lower temperatures for mixing or melting may be achieved by adding small quantities of more innovative materials, hence reduce fuel consumption (PB & DNV GL, 2015). Also known as "batch separation" or "smart melting concepts", raw materials can be split into portions with different compositions, melting temperatures and reaction paths. That way, the desired reactions between the fluxes and quartz are promoted at correct timing, resulting in up to 50% shorter melting time and allowing more time for the refining process (Carty, 2013; PB & DNV GL, 2015). Shorter melting times are also achieved through the production of pre-mixed pellets with the correct proportions of ingredients. The technology was expected by 2020 for low-cullet containing batches at a relatively small additional cost (i.e. EUR 0.28 million), with the possibility to apply pelletised batch preheating with flue gas heat (VNG, 2012; Hatzilau et al. 2016, also discussed in Section 4.6), but no major developments have been made in recent years, pushing back the potential availability of this technology.

4.3 Process design

The industrial processes have already been substantially optimised, but process enhancements can continue to further reduce the energy consumption of high-temperature processes (Berntsson, 2017; NPI, 1998). Such improvements implicate a more complex technology that may result in additional maintenance and capital expenditure, the use of non-environmentally friendly chemicals, and limitations to equipment lifespan (Meuleman,

³³ Sven-Roger Kahl, Ardagh Glass Dongen B.V., Personal communication; Oscar Verheijen, Celsian Glass and Solar B.V., Personal communication.

2017). The focus is primarily on furnace energy reduction methods for improved energy intensity efficiency and subsequent reductions in carbon emissions.

Electric furnaces

Electric melting is a key decarbonisation technology, which is assumed to be available for commercial implementation post-2030 (PB & DNV GL, 2015). This type of melter is based on the Joule's Principle where the molten glass is used as the resistive element of the electrical circuit. All-electric melting generally implies a cold-top vertical melting, with the raw material being distributed evenly over the melting surface of the glass (Stormont, 2010). Most of the electrical power ends up in the melting process, and only relatively low energy losses come from transformers, busbar and control efficiency.



Figure 15 CFD model of 100m² design all-electric melting concept in container glass applications (Reynolds, 2019).

Electric furnaces are much more thermally efficient than gas-fired melters, using about 35% less energy compared to an as-is regenerative furnace (i.e. 3.2 GJ/t; excluding losses in electricity generation) (Meuleman, 2017). Commercially available electric furnaces have a relatively small capacity (approx. 150t). For some glass types the use of an all-electric furnace is difficult (e.g. conductivity issues in fibre glass). A 200tpd cold-top all-electric furnace can potentially achieve 2.75 GJ/t for 50% cullet (Reynolds, 2019). When melting flint container glass, a 250tpd all-electric furnace by Electroglass requires 710kWh of electricity per tonne of glass, equivalent to a thermal efficiency of 85% (Stormont, 2010). Polluting emissions are considerably less for an electric melter at the glass factory, where thermal NO_x or SO_x emissions are plunged and combustion related CO₂ are avoided.

As to the challenges of a standard full-scale container glass electric furnace, a cold-top solution affects the stability of the batch blanket³⁴. The mixture melts much faster for 80–90% cullet rates which prohibits the creation of a stable and controlled blanket that keeps the furnace sealed. Electric furnaces are currently not flexible when it comes to changing pull rates, which is important for meeting the fluctuating demand. It is also not easy to transform conventional furnaces into cold-top ones due to size, quality and lifetime. The large amount of power needed for this conversion often comes with additional investments to the electricity infrastructure. Electric melting would be only possible provided that sufficient renewable electricity is available and can also be delivered to a plant. Parameters for an electric furnace are summarised in Table 6.

Table 5 Parameters electric furnace for container glass

³⁴ Much heat is released from the open surface if the batch blankets get too hot, and there is not much heat capacity to keep the process running without isolating or insulating the top (e.g. through adding extra batch).

Inputs/emissions	Electric furnace	Unit
Electricity ³⁵	4.1	GJ/t molten glass
Cullet rate	50	%
Direct CO ₂ emissions	39	kg/t molten glass

Oxygen in Combustion

The use of oxygen instead of for the combustion of natural gas avoids having to heat the unnecessary nitrogen in air and as a consequence may decrease fuel usage. Additionally, the use of oxygen instead of air creates a reasonably pure CO_2 stream and reduces specific NO_x emissions while helping increase the flame temperatures (TNO, 2007; see Section 2.9). The additional cost for oxygen should be more than offset by the reduction in fuel cost in order to make it a cost-efficient option.

Development of lower-cost oxygen separation techniques allows the use of oxidiser with almost 100% content of oxygen, making the conversion to oxy/fuel economic (Baukal, 2013; Levine, 2001). Such techniques include vacuum pressure swing adsorption (VSPA) units, membrane separation, and cryogenic systems which produce grade of oxygen up to 99.5% (LexInnova, 2013; TNO, 2007; Cioli et al. 2019). The main concern for the deployment of this technology is the energy and costs required to produce oxygen, as well as the corrosion of refractory silica brick used to line the furnace roof. An optimal use of oxygen is required to offset this cost through fuel savings and emissions reduction.



Figure 16 (a) Premixing atmospheric air with oxygen; (b) Oxygen lancing; (c) Airoxy/fuel firing; (d) Oxy/fuel firing (Baukal, 2013).

a) Oxygen-enriched air

Oxygen enrichment uses a blower to deliver combustion air which mixes with oxygen and further mixes with the fuel inside the burner (Figure 16a). By supplying slightly more oxygen than the stoichiometric minimum, the unburned fuel is avoided and the efficiency penalty from excess air is minimised (see Appendix A). As a result, a mole fraction of oxygen in the oxidant stream between 21-30% is the range which compromises between energy savings and pollutant emissions such as NO_x (Worrell et al. 2008). Beyond that range, flame temperatures decrease as the dissociation reactions become appreciable (e.g. CO₂ to CO and O₂; H₂O to O₂ and OH) (Ming et al. 2003; Baukal, 2013). For this reason, oxygen enrichment and fuel injection are often linked in furnace operations for balancing the temperatures.

³⁵ Including electricity for batch mixing

b) Air-oxy/fuel firing

This type of combustion technologies entail injecting air and O_2 separately through a combined air/- and oxy/fuel burner (Figure 16c). The burner allows for higher concentrations of oxygen compared to oxygen enrichment, resulting in lower emission rates and greater melting capacities for responding to fluctuating demand. A conventional air/fuel burner can be retrofitted by inserting an oxy/fuel burner through it or by placing an O_2 lance to fire natural gas in the combustion space above the glass melt. This method has also been used to control the flame to produce a desired shape (Baukal, 2013).



Figure 17 (a) Oxidiser compositions for blends of air and pure O_2 ; (b) Natural gas savings vs. flue gas temperature (Baukal, 2013).

c) Oxy/Fuel Combustion

This technology involves increasing the percentage of oxygen in the furnace which makes the furnace more thermally efficient (PB & DNV GL, 2015; LexInnova, 2013). The major differences in the operation of oxy/fuel compared to regenerative furnaces are in the way that fuel is burned and the amount of electricity and oxygen consumed for the oxidant delivery (O'Connor, 2015). There are typically 4–6 staggered burners per sidewall and a limited number of exhaust ports. Oxy/fuel furnaces do not apply heat recovery systems but attempts have been made to use batch preheating for recovering part of the residual heat (Ming et al. 2003). It has been used as BAT in the glass industry to improve the efficiency of gas burning (Fraunhofer, 2019b).

Taking plant infrastructure, burner technology, furnace design, regenerators, pipework and air separation systems into account, the higher Capex for this technology compared to regenerative furnaces results in extended payback periods (PB & DNV GL, 2015). However, a complete oxy-fuel furnace in brand-new installations is approx. 35% cheaper compared to a cross-fired and 25–30% cheaper than an end-fired regenerative furnace (Messer, 2018). Oxy/fuel rebuilds (approx. EUR 0.6 million; Baukal, 2013) often require reduced construction time than air/fuel (Lievre et al. 2008). Despite requiring an energy intensive production of high-purity oxygen, oxy-fuel firing is still beneficial as it reduces the volume of waste gases by about two thirds, increases the product throughput rate by 0–10% (Ecofys, 2009; Lievre et al. 2008). Any pollutants such as nitrous oxides or sulphur dioxides are easier to remove in an oxy/fuel exhaust, since they are in much higher concentrations compared to air/fuel (LexInnova, 2013).

Heat integration techniques

Waste heat recovery considers the utilisation of the high-temperature exhaust gases in a cyclic process (Hatzilau et al. 2016). Exhaust gases have a typical heat content of about 25–

30% of the furnace energy input. Re-using this residual heat helps in decreasing specific energy consumption, reaching higher flame temperatures and mitigating direct CO₂ emissions during the glassmaking process (see Section 2.9). Waste heat recovery options (WHR) are suitable for air preheating, electricity/steam/hot water generation, thermochemical recuperation, natural gas preheating, batch/cullet preheating or infrastructure heat (Dolianitis et al. 2016; Fraunhofer, 2019a). For oxy/fuel technology, there is relatively less energy in the flue gases but heat can still be recovered for preheating oxygen or gas. The gases must remain above the sulphuric acid dew point (approx. 180°C). The optimal use of waste heat is location specific and the additional Capex must be balanced against incremental heat recovery.



Figure 18 Waste heat recovery in a glass melting plant (Khoshmanesh et al. 2007).

a) Batch/cullet and cullet preheaters

Batch preheating is a commercially available technology which improves furnace efficiency by evaporating moisture in the batch and lowering the overall furnace peak temperature (Springer et al. 2017). The heterogeneous mixture is conveyed to the top of the preheater (approx. 20–25m high), where it is preheated by direct contact with either flue gases or steam (Hatzilau et al. 2016). The flue gases exiting the regenerator are cooled down from 1500°C to 450°C before entering the batch preheater and leaving it at 250°C. The mix is ideally preheated at temperatures around 300°C, before cullet starts exhibiting liquid characteristics.

As innovations such as electric heating and furnace insulation are applied, there would be less waste heat available for preheating (Fraunhofer, 2019a). Specific energy consumption is reduced as both the temperature and mass of the flue gases decrease, which subsequently lower the direct CO_2 emissions by about the same amount (Hatzilau et al. 2016). For batch/cullet preheaters, preheating temperatures range between 275–325°C for air/fuel and 200–250°C for oxy/fuel firing (PB & DNV GL, 2015; Wallenberger, 2010; Sorg, 2011). The energy savings in the furnace using this technology range between 12–20%, the glass pull can be increased by 10% and the estimated lifetime is 20 years (HRE, 2018).

b) Electricity generation

On-site electricity generation for internal consumption can be applied in cases where there are no other uses for the recovered heat (HREII, 2018). This readily available option results

in less dependency on fluctuating energy costs and on external energy sources (i.e. cope with a power outage). This application can be considered as an energy efficiency measure, since investing companies reduce their electricity consumption without additional use of primary energy (HREII, 2013). The temperature and the amount of heat recoverable need to be high enough to drive a steam turbine which drives the electricity generation equipment (Forni et al. 2014). However, the aggressiveness of flue gases may impede proper operation of this technology.

The Organic Rankine Cycle (ORC) is a theoretically feasible solution for generating electricity from heat sources of container glass plants, having a conversion efficiency of 15–19% (Figure 19). The additional heat exchanger is the main component for heat recovery, located after flue gas treatment (e.g. de-SOx, de-NOx) to eliminate the need for cooling down the flue gases (HREII, 2018). It is suitable for temperatures 450–500°C of flue gases, as well as for low-power and discontinuous flows of hot gases with temperatures around 300°C (Hatzilau et al. 2016). It can only be used in combintion with electrostatic filters, not with bag filters. When combined with preheating systems, the quantity of recoverable energy is low for efficient power generation and supplementary firing may be needed to generate superheated steam to drive the turbines (IETD, 2018). In principle, if half the energy of exhaust gases exiting the furnace is recovered, it is possible to produce between 30–60kWh of electricity for every tonne of produced glass (HREII, 2018).



Figure 20 Organic Rankine Cycle (ORC) waste heat recovery³⁶.

c) Optimelt thermo-chemical regenerator (TCR)

Optimelt TCR refers to the advanced heat recovery for oxy/fuel fired glass furnaces. The recycled flue gases with CO₂ and water vapour are used for endothermic reforming of methane-based (including biomethane) fuels into a hot syngas (i.e. H₂ and CO; approx. 1200°C) in small regenerators (Hatzilau, et al. 2016). The technology entered the engineering phase in 2018 for glass-container furnaces of 200tpd, and research continues on examining the technical barriers of applying it to high-cullet batches (Section 4.1). The temperature required inside the reformer is between 800–900°C.

Having a 240tpd glass-container oxy/fuel baseline with 1MW electric boost and 30% cullet ratio, Praxair reported fuel savings of 21% for Optimelt TCR and an additional 5% for

³⁶ Retrieved from Turboden: <u>https://www.turboden.com/turboden-orc-technology/1062/the-orc-technology</u>

Optimelt Plus (Laux et al. 2016). Reductions in energy consumption and CO₂ emissions may reach 30% and 45–60% respectively, compared to air-regenerative furnace (de Diego et al. 2016). However, the high amount of CO requires the complete sealing of the regenerators to avoid leakage³⁷. The technology can be combined with heat recovery options such as integrated batch/cullet or cullet preheating, regenerative oxygen preheating, generation of reforming steam for TCR, and ORC turbo-generator (Laux et al. 2016).

Linde PLC/Praxair modelled the optimelt furnace and found the following potential energy consumption values when applying the Optimelt technique to container glass production: energy consumption of 3.2 GJ for a 240tpd Optimelt TCR furnace with 1MW electric boost and 30% cullet, with a further decrease by 5% when applying oxygen preheating (i.e. Optimelt Plus) and additional cuts of 4% when combining with a cullet preheater (de Diego et al. 2018; Laux et al. 2016; Herbst et al. 2017).



Figure 21 Side-fired 50tpd oxy/fuel glass furnace converted to end-port Optimelt (Laux et al. 2016).

Improved furnace construction

Both revolutionary furnace designs and modifications to existing furnace designs concern medium to long-term investments (VNG, 2012). Depending on the glass type, the deteriorating refractories, melt tank and combustion chamber contribute to an annual increase of losses by approximately 0.8–1.3% for air/fuel and oxy/fuel furnaces. A reconstructed crown and basin using improved refractories lead to higher operating temperatures, hence improved product quality and payback rates (PB & DNV GL, 2015a; Khoshmanesh et al. 2007). Making such improvements to a certain extent provides with better insulation to combat high corrosion rates and extend furnace lifetime while reducing heat losses by 50–60% (TASIO, 2016). On the other hand, a change from air/fuel to oxy/fuel results in a total rebuild of an existing furnace and usually comes with a total change of

³⁷ Joost Laven, O-I Netherlands Schiedam B.V., Personal communication.

footprint. Burners, gas and oxygen supply, refractory material will be new and in almost all cases the design will be significantly changed³⁸. In principle, innovative changes can only be done when a large furnace overhaul takes place.

4.4 Recycling and re-use

Increased cullet rate

The industry is already using high volumes of recycled cullet, resulting in significantly increased melting efficiencies. A high-cullet batch is responsible for decreased thermal limits and lowered residence times, resulting in a ~2.5% reduced fuel consumption for every 10% addition of recycled cullet (Madivate, 1998) (for container glass). The treatment of recycled cullet requires 0.18 GJ/t of cullet which is considerably less than the energy needed for raw materials. Therefore, each tonne of primary raw material replaced by cullet saves 1.9-2.35 GJ/t glass, which is also driven by the reduced volumes of soda usage (Drummond, 2011). The reduction in process emissions as a result of the avoidance of heat reactions with the carbonates is also significant, where a cullet-only batch would theoretically result in a 55% emissions reduction compared to batch consisted of raw materials (Butler et al. 2011). This option prerequisites a better quantity/quality check of the cullet, including activities such as the removal of different contaminants and a better separation system.

4.5 Product design

Alternative glass compositions

Variations in the batch mixing formula may have potential benefits in energy costs and environmental impact (Butler et al. 2011). The energy consumption in the chain per packaged unit (i.e. from raw material to glass production) can be considerably reduced, where 0.65% less energy is required for every 1% weight saving per bottle (VNG, 2012; PB & DNV GL, 2015a). Changes in glass composition will result in colour and property changes which are interlinked to the client preferences (Section 3.1). An overview of savings of theoretical energy requirement and CO_2 emissions for alternative glass compositions is presented in Appendix B.

4.6 Residual energy use

Building Heating

Waste heat from the glass furnace or downstream smaller other technologies such as annealing lehrs or air compressors that contain sensible heat may be put to practical use. That way, adequate warehouse temperatures are maintained or general heating applications are facilitated (PB & DNV GL, 2015; Bišćan et al. 2012, cited in Hatzilau et al. 2016). Steam generation and usage incur challenges of fouling and corrosion of the heat exchanger materials due to contaminated exhaust gases, therefore additional costs are required for flue gas cleaning (e.g. desulphurisation) (Worrell et al., 2008; Beerkens, 1986). Investment costs are compensated with lower energy consumption which leads to a reduction in operational costs (TNO, 2019). A continuous heat demand is also necessary for replacing conventional cooling systems with heat applications.

³⁸ Sven-Roger Kahl, Ardagh Glass Dongen B.V., Personal communication.



Figure 22 Waste heat recovery boiler in the copper industry (Fraunhofer, 2019a).

District Heating

District heating is a technical solution to deliver low-grade heat to external thermal uses, such as for tertiary, agriculture, or industrial processes in neighbouring facilities (HREII, 2018). Thermal energy can be provided by the heat recovery system to feed the district heating grid, which has in turn resulted in advanced heat integration on industrial plants (Ricardo-AEA, 2013). The implementation of a district heating project is often impeded by factors such as missing infrastructure (e.g. pipelines, back-up systems), disagreements on where heat should be transferred to and taking up the risk of the investment (Forni et al. 2014).

4.7 Carbon Capture, Storage and Utilisation (CCS/CCU)

Carbon capture techniques depend on the characteristics of flue gases such as concentration of CO₂, purity and volume. Three options are distinguished: pre-combustion, postcombustion and oxy/fuel combustion capturing. Depending on the fuel input and the combustion agent, the capturing involves the removal of particulates, contaminants and water (i.e. SO_x, NO_x, H₂O vapour), as well as the compression of CO₂ into sealed containers (Fraunhofer, 2019a). Cooling down, liquefying and cleaning CO₂ is a cost- and energyintensive operation. Separation technologies include cryogenic process, vacuum pressure swing adsorption (VPSA) and selective membranes (Ming et al. 2003). The cost of available emission emissions control equipment (i.e. particulate and NOx) is estimated at EUR 2.1 million for air/fuel and EUR 0.6 million for oxy/fuel furnaces (Baukal, 2013).



Figure 23 | Options of capturing carbon sorted by combustion (Baukal, 2013).

The deployment of integrated and commercial-size CCS projects has been limited to a few industrial applications. Researchers often assume that carbon capture would be employed on the majority of sites which currently employ oxy-fuel combustion (Fraunhofer, 2019a). However the scale of CO_2 emissions in the glass sector is such that the implementation of carbon capture at a facility is currently insufficient to justify the implementation of a full CCS chain (PB & DNV GL, 2015). A potential proximity of the glass facility to a larger industrial cluster would provide access to a shared CO_2 transportation and storage network while enabling the utilisation of captured CO_2 (CCU). Despite the high specifications of the receiver industries which would increase the cleaning cost of the exhaust gases, the small volumes of captured CO_2 are more likely to align with the CO_2 feedstock requirements of future CO_2 utilisation industries.

5 Discussion

Low-carbon innovations must be selected based on prospects of possessing a place in the future technology mix, given that the process industry in 2050 will have experienced substantial changes (de Pee et al. 2018; Bouman et al. 2017). They can be achieved in part through process integration techniques, allowing for further CO₂-reduction opportunities to be tapped and synergies across sectors to be exploited (Berntsson, 2017). It is important is to avoid lock-in by implementing technologies on the shorter term which create barriers for further innovation (ECN, 2018).

Options exist on a technological base as well as through a shift in energy carrier, while there are synergies and mutually exclusive options such as the combination of electric melting and switching to fuels of higher calorific value (PB & DNV GL, 2015b). To a large extent, the existing furnaces will be the operating ones in 2030³⁹ and much of the growth will be met by upgrading existing plants at scheduled rebuilds (JRC Report, 2013). The oxy/fuel technology

³⁹ Oscar Verheijen, Celsian Glass and Solar B.V., Personal communication.

will be easiest to include for air/fuel furnaces with inefficient or no heat recovery, with high cost electricity or with a need for post-treatment of the flue gases (Baukal, 2013). Hybrid configurations that range from limiting the zones of regenerative or oxy/fuel firing as well as using by-product fuels for regeneration and enriching the air stream with oxygen are steadily gaining momentum (O'Connor, 2015; Reynolds, 2018). Lastly, a combined full-electric option for container glass production would a require more sophisticated control system to allow changing pull rates and new designs to handle the increased erosion and corrosion of the refractories under process changes⁴⁰.

While pursuing the deep decarbonisation of the industry, the Dutch container glass producers are facing the challenge of maintaining a sustainable competitive advantage over international players in the packaging industries. From a transitional perspective, the Dutch companies make efforts to remain sufficiently profitable and position themselves optimally within their organisations to enable reinvestment at their production sites (MEE, 2015). At the same time, both the energy costs and the overall cost effectiveness of glass installations are having a direct impact on investment potential in energy and CO₂ reduction. Investments are expected to be significantly linked to the prices of EU allowances, given the pressure coming from the EU-ETS, and the price volatility of carbon and energy carriers (Hatzilau et al. 2016).

The efforts of the glass industry to achieve cumulative emission reductions by 2050 contribute to the Dutch governmental agenda for attaining national energy goals (MINEZ, 2017). It is therefore imperative that this endeavour is accompanied with a robust policy framework for supporting innovation and long-term industrial investments which are needed for both the society and for a competitive low carbon economy (FEVE, 2016; Eurostat, 2011). In addition to technical initiatives, education, training and international precompetitive cooperation in R&D and knowledge transfer are essential, especially where complex innovation is needed (MME, 2015).

⁴⁰ Sven-Roger Kahl, Ardagh Glass Dongen B.V., Personal communication.

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APPENDIX A – Conventional technologies

End-port regenerative furnaces

Air/fuel furnaces of this type are broadly used in the Netherlands along with oxy/fuel furnaces. Regenerative heating is a cyclic process where exhaust gases pass over and heat up refractory blocks in one of two pre-heating chambers (Meuleman, 2017). Cold combustion air is introduced into the first chamber to be pre-heated by abstracting heat from the refractory while the exhaust gases are diverted to heat the second chamber. Continuous reversal of this process in 20-minute intervals provides a permanent flow of pre-heated combustion air. As a result, preheating the combustion intake air going into the burners can lead to more potential savings (Ming et al. 2013).

Regenerative furnaces help providing higher capacities and producing high quality glass, we assume with a typical energy efficiency of 70%, although in practice 60% would be a more typical value. This results in an energy use of approx. 4GJ/t for a container batch containing 76% cullet. It allows high thermal efficiencies and preheat temperatures at a theoretical maximum of 1480°C, and the hot flue gases exit the regenerator at 450–550°C (TNO, 2007). The capital cost for heat recovery in air/fuel furnaces reaches EUR 1.5 million (Baukal, 2013).



Figure A1 Schematic of an end-port regenerative furnace⁴¹.

Electric boosting and Bubblers

Electric boosting refers to the application of electrical energy as an additional energy source in conventional fossil fuel-fired furnaces (approx. 2–12% of the total energy input). It supports the pull rate of a furnace as it nears the end of its operating life, and provides flexibility by increasing the production capacity (Figure A3a; Worrell et al. 2008). This extra energy is applied in melting areas which are difficult to heat using gas, particularly when melting coloured glass (Eurotherm, 2019). The key is in the electrode arrangement and the energy release pattern that the electrodes create in the furnace, which in turn directly affects temperature profiles, convection currents, flow paths and residence time (Stormont, 2010; Karem, 2018). At current designs, a larger percentage would lead to glass quality issues, and it is also limited by the cost of electricity (CPIV, 2009). Depending on the furnace geometry, the majority of melting boosters have an installed power in the range of 400–1200kVA⁴².

⁴¹ Retrieved from BDF Industries: <u>https://www.bdfindustriesgroup.com/products/melting-furnace-rigenerative-recuperative-furnace/</u>

⁴² Retrieved from Glass Service s.r.I.: <u>https://www.glassservice.it/en/products/booster/49/</u>

An alternative to electric boosting is the fluidised bed combustion (i.e. "bubbling"); a simple method for increasing bottom temperature and assisting the retention of unmelt batch in melting area (Sorg, 2011). Air or other gases are blown through special bubbler nozzles installed in the furnace bottom, hence the upward movement of bubbles produces strong localised convection currents around their path. These currents move bottom glass upwards causing an increase in the glass temperature at the bottom of the tank.





Excess Combustion Air

In a combustion process, the complete burning cannot be accomplished with the exact theoretical amount of air (i.e. stoichiometric conditions). Therefore it is essential to supply an excess amount of air for the combustion of high calorific gases, ideally in a ratio of approximately 10:1 (Boateng, 2016). This corresponds to a 1–2% oxygen measurement in the stack. By contrast, too much excess air leads to lower flame temperature, hence less heat gets into the system. As a result, the best combustion efficiency occurs at the optimum air-to-fuel ratio and controlling this provides the highest efficiency (Figure A3b). With suitable preheaters, the use of excess air can be used on a large scale with economic advantage over oxygen enrichment, although much depends on the availability and price of





Figure A3 (a) Electric boost power input per extra tonne per day by glass type (Stormont, 2017); (b) Relation between combustion efficiency and excess air⁴³.

⁴³ Retrieved from Engineering Toolbox: <u>https://www.engineeringtoolbox.com/boiler-combustion-efficiency-</u> <u>d 271.html</u>

The calculation of energy and mass balance was performed based on values retrieved from scientific literature in order to match energy consumption to the registered emissions, following the method described in Appendix C⁴⁴. Plant-related information was retrieved from interviews with glass experts for verifying the correctness of results. Raw material inputs were assumed to be 62.3% silica, 18.4% soda ash, 7.8% limestone, 10.9% dolomite and 0.6% sodium sulphate. These are not actual values representative for a glass company, as details of the process may differ and result in substantial changes.

Parameter	Regenerative end-port furnace with 8% electric boosting.	Regenerative end-port furnace with 8% electric boosting and batch/cullet preheater	Oxy/fuel furnace with pure oxygen and 5% electric boosting.	Unit/t molten glass
Natural gas	3.95	3.23	3.56	GJ
Electricity ⁴⁵	0.39	0.33	0.23	GJ
Oxygen	0	0	0.29	t
Direct CO ₂	0.26	0.22	0.24	t
Stack loss	1.42	0.64	1.82	GJ

Table A1 Calculated specifications of inputs and outputs for the furnaces under study, expressed per tonnes of molten glass, for batches with 76% cullet.

⁴⁴ The excel model can be found at <u>https://github.com/ioannispapadogeo/MSc_Thesis</u>

⁴⁵ Including 0.048 GJ/t for batch preparation

 $^{^{\}rm 46}$ Natural gas consumption with emissions factor of 56.6 kg/GJ accounts for 85% of emissions, 15% are process emissions

APPENDIX B – Alternative batch compositions

Table B1 Overview of alternative container glass types.

			Chem.
Type			reaction ⁴⁷ –
No.	Chemical Reactions per Glass Type	Description	Melting ⁴⁸
			- Direct CO ₂
1	$CaMa(CO_3)_2(s) + Heat \Rightarrow CaCO_3(s) + CO_2(a) + MaO(s)$	Carbonate	0.82 GJ
		route: reacting at 850-1000°C	5.09 GJ 319 kg
	$Na_2CO_3(s) + CaCO_3(s) \Rightarrow Na_2Ca(CO_3)_2$	and use of	519 Kg
	$Na_2Ca(CO_3)_2 + 2SiO_2 \Rightarrow Na_2O.CaO.2SiO_2 + 2CO_2(g)$	Dolomite	
2	Na₂CO₃(s) + CaCO₃(s) ⇔ Na₂Ca(CO₃)₂	Carbonate route and use of	0.67 GJ 4 52 G1
	Na ₂ Ca(CO ₃) ₂ + 2SiO ₂ \Rightarrow Na ₂ O. CaO.2SiO ₂ + CO ₂ (g)	Limestone	259 kg
3	2SiO₂ (s) + Na₂CO₃(s) ⇔ Na₂O.2SiO₂ (s) + CO₂(g)	Silica route:	0.55 GJ
	$C=M_{2}(C_{2}) + M_{2}(C_{2}) + M_$	860°C and use	3.82 GJ 286 kg
	$Camg(CO_3)_2(S) + Heat \Rightarrow CaCO_3(S) + CO_2(g) + MgO(S)$	of Dolomite	-
	$CaCO_3(s) + Heat \Rightarrow CaO(s) + CO_2(g)$		
	2CaO + (SiO ₂ + Na2O.2SiO ₂) \Rightarrow Na ₂ O.2CaO.3SiO ₂		
4	2SiO ₂ (s) + Na ₂ CO ₃ (s) ⇔ Na ₂ O.2 SiO ₂ (s) + CO ₂ (g)	Silica route:	0.45 GJ
	$C_{2}(C_{2}(c) + H_{2}(c) + C_{2}(c) + C_{2}(c)$	860°C and use	213 kg
	$Caco_3(s) + Heat \rightarrow Cac(s) + Co_2(g)$	of Limestone	
	2CaO + (SiO ₂ + Na ₂ O.2 SiO ₂) ⇔ Na₂O.2CaO.3SiO₂		
5	$SiO_2(s) + Na_2CO_3(s) \Rightarrow Na_2O.SiO_2(s) + CO_2(g)$	Silica route: reacting at 800–	0.33 GJ 2.71 GJ
	$CaMg(CO_3)_2(s) + Heat \Rightarrow CaCO_3(s) + CO_2(g) + MgO(s)$	1100°C and use of Dolomite	178 kg
	CaCO₃(s) + Heat ⇔ CaO(s) + CO₂(g)		
	3CaO + (5 SiO₂ + Na₂O. SiO₂) ⇔ Na₂O.3CaO.6SiO₂		
6	(2) (2) (2) (2) (2) (2) (2) (2) (2) (2) (2)	Silica route:	0.27 GJ
	$SiO_2(S) + Nd_2CO_3(S) \Rightarrow Nd_2O.SiO_2(S) + CO_2(g)$	reacting at 800- 1100°C and use	3.16 GJ
	$CaCO_3(s) + Heat \Rightarrow CaO(s) + CO_2(g)$	of Limestone	244 Kg
	$3CaO + (5 SiO_2 + Na_2O. SiO_2) \Rightarrow Na_2O.3CaO.6SiO_2$		
7	SiO2(s) + Na ₂ CO ₃ (s) ⇔ Na ₂ O.SiO ₂ (s) + CO ₂ (g)	Silica route: reacting at 800-	0.20 GJ 2.51 GI
	Al₂O₃ +5SiO₂ + Na₂O.SiO₂ ⇔ Na₂O.Al₂O₃.6SiO₂	1100°C and use	117 kg
		of Aluminium	
8	CaMg(CO ₃) ₂ (s) + Heat ⇔ CaCO ₃ (s) + CO ₂ (g) + MqO(s)	Carbonate	0.36 GJ
		at 800-900°C	4.31 GJ 278 kg
		and use of Dolomite	-
		or polonite	

⁴⁷ Theoretical Energy requirement for 68% cullet. (excl. primary energy). Calculated based on stoichiometry, ⁴⁸ Assumed only natural gas is used for melting.

APPENDIX C – Material and Energy Flow Analysis

Mass Balance

Material flow analysis helps minimising the assumptions about the primary energy needed to deliver energy to the system and the chemical composition of batches and final products. For this purpose, mass balance is performed for estimating the material requirements and cycles which are directly connected to the material and emissions streams. The batch weight is the aggregate of virgin materials and recycled cullet (i.e. heterogeneous mixture) and equals to the amount of molten glass added by the process emissions (Eq. C1, C2). Approximately 10% of glass is lost in activities downstream of the melting which is re-introduced back to the batch mixing. The reactions taken place can be expressed as (Madivate, 1998):

((1 + b) kg of batch) (Troom) \Rightarrow (1 kg of glass mix) (Troom) + (b kg of gas) (Troom), ΔHR (C1)(1 kg of glass mix) (Troom) \Rightarrow (1 kg of glass melt) (T), $\Delta Hglass$ (C2)(b kg of gas) (Troom) \Rightarrow (b kg of gas) (T), $b\Delta Hgas$ (C3)

where *T*: melting temperature, *b*: gas emitted during reaction, ΔH_R : sum of energy necessary to decompose raw materials to oxides and energy involved in the formation of vitreous phases of glass (approx. 0.3 GJ/t glass), ΔH_{glass} : enthalpy of glass (approx. 0.6 GJ/t glass depending on the glass type), and ΔH_{gas} : enthalpy of gases (approx. 0.28 GJ/t glass). The enthalpies used are presented in Appendix C which originate from standard thermochemical tables.

The batch and glass compositions per glass colour are presented in Table C1, which are normalised by the sum of the most abundant contents to 100%. The end-product contains 70–74% silica, 12–15% sodium oxide, and 10–15% calcium oxide by weight. The heat capacities under constant pressure⁴⁹ (c_p) for raw materials and natural gas compounds were calculated using formulas of Moore et al. (1951; 1958) and Engineering Toolbox⁵⁰, respectively.

Compound	Flint	Dark Amber	Emerald Green	Molar mass (kg/mol)	Heat capac. (kJ/K*mol)	Density (kg/m³)
Batch composit	ion (wt %)					
SiO ₂	62.28	61.98	62.96	0.060	0.111	0.265
Na ₂ CO ₃	18.42	16.79	18.89	0.106	0.115	0.254
CaCO ₃	7.78	9.81	11.33	0.101	0.098	0.271
CaMg(CO ₃) ₂	10.90	11.11	6.31	0.184	0.225	0.286
Na ₂ SO ₄	0.62	0.31	0.51	0.142	0.199	0.266
Glass composit	ion (wt %)					
SiO ₂	73.07	73.39	73.31	0.060	0.283	0.265

Table C1 Chemical composition batch and glass product (Madivate, 1998).

 49 Standard conditions refer to temperature level $T_{room}=25^{\circ}C$ of environment and to isobaric conditions at p=1 bar. The c_p values presented in Table C1 consist of the average of all glass types for the temperature window between the ambient and the furnace temperatures (see Section 2.9).

⁵⁰ Retrieved from Engineering Toolbox: <u>https://www.engineeringtoolbox.com/methane-d_980.html</u>

Na₂O	12.72	12.36	12.74	0.062	0.483	0.227
CaO	9.86	9.91	10.00	0.056	0.254	0.334
MgO	2.84	2.77	2.03	0.040	0.318	0.358
Al ₂ O ₃	1.53	1.58	1.92	0.102	0.278	0.395
Process emission	ns (vol %)					
Process emission CO ₂	ns (vol %) 98.18	99.14	98.44	0.044	1.097	1.977
Process emission CO ₂ SO ₂	ns (vol %) 98.18 1.22	99.14 0.57	98.44 1.04	0.044 0.064	1.097 2.109	1.977 2.630

Theoretical Energy Requirement

The estimation of the material and emissions streams enables the application of the laws of thermodynamics for calculating the energy requirements of the melting activity (Figure 3). Integral to the calculation of the specific energy consumption of the melting activity is the estimation of the temperature windows of the batch melting point and furnace operation. Within these boundary conditions, the simplified methodology proposed by Madivate (1998) is used for calculating the theoretical energy requirement for silicate glasses. This includes the heat involved in the transformation of a certain amount of batch material at room temperature to molten glass and flue gas at temperature T in the glass-melting region.

The estimation of the temperature windows of both the furnace operation and batch melting point are necessary for setting realistic boundaries for the calculation of the specific energy use of the furnace. With the assumption that the trajectories of both the furnace temperature (Eq. C4) and the batch melting point (Eq. C5) have the same slope and are proportional to the cullet added to the batch⁵¹ (see Section 3.2), these temperatures can be approximated using the following formulas:

Furnace Temperature =	T _f * (1+E*(1-C/100)) / (2*10)	(C4)

Batch melting point =
$$T_m^* (1+E^*(1-C/100)) / (2^*10)$$
 (C5)

where T_f : furnace temperature for 100% cullet, T_m : melting temperature for 100% cullet, E: energy savings per cullet (approx. 33% for 100% cullet), C: cullet percentage, C/10: amount of times that a 10% increase is added in the batch. The melting point moves inside the temperature window for sand dissolution and increases after the melting to homogenise the molten glass (i.e. maximum furnace temperature; see Section 2.2).

This study leverages the methodology proposed by Madivate (1998) for calculating the theoretical energy requirement (TER) for the fusion of glass. The energy consumption of the furnace is considered constant in the calculations which, in reality, is dependent on the production level of the equipment. The results generally have a deviation of <2% from results of more-complex approaches (Madivate, 1998, pp. 3305). The theoretical energy requirement (TER) for the fusion of glass is expressed as follows:

$$TER = \Delta H_{glass}(T) + \Delta H_{gas}(T) + \Delta H_{R}(T_{room}) [kJ/(kg \text{ of glass mix})]$$
(C6)

where ΔH_{glass} is the heat required to raise the temperature of the raw materials from 25°C (T_{room}) to the highest melting point and then raise the temperature of the melt until the maximum furnace temperature, ΔH_R is the latent heat required to enable the reactions between the batch components to form the glass as a function of T_{room} , and ΔH_{gas} is the heat content of the gases released from the batch during melting, all expressed in kJ per kg of the

⁵¹ Lower limits are 1500°C for furnace temperature and 1100°C melting point for a cullet-only batch. The furnace temperature is normally affected by the glass quality demands.

heterogeneous glass mix. The mixing enthalpy which is involved in the transformation of the batch and cullet mixture into homogeneous molten glass was neglected (see Section 2.2).

The mass fraction of oxides Na_2O , CaO, and SiO_2 which are found in abundance in the glasses under study are normalised by their sum to 100%. By leveraging the glass compositions of Table C1 for the respective type, the elements of equation (C6) are obtained from the following relations (Madivate, 1998):

$$\Delta H_{glass}(T) = \Sigma(\text{\%oxide})_i * x_i + [\Sigma(\text{\%oxide})_i * y_i] * T$$
(C7)

$$\Delta H_{gas}(T) = b * [\Sigma m_i a_i^* (T-296) + (0.5/10^{-3})^* \Sigma m_i b_i^* (T^2 - 296^2) - ...$$

... - (1/10⁶)* $\Sigma m_i c_i^* (T^{-1} - 296^{-1}) + (0.33/10^6)^* \Sigma m_i d_i^* (T^3 - 296^3)]$ (C8)

$$\Delta H_{R} = (1+b)^{*} (\Sigma \Delta H_{d,i}^{*}m_{i}) + \Delta H_{f,1}^{*} (\% CaO/100) + ...$$

... + $\Delta H_{f,2}^{*} (\text{rest-Na}_{2}O/100) + \Delta H_{f,3}^{*} (\text{rest-SiO}_{2}/100)$ (C9)

where: rest-Na₂O =
$$Na_2O - CaO^*(M_{Na2O}/3*M_{CaO})$$
 (C10)

$$rest-SiO_2 = \%SiO_2 - \%CaO^*(6^*M_{SiO2}/3^*M_{CaO}) - rest-Na_2O^*(2^*M_{SiO2}/M_{Na2O})$$
(C11)

and *%oxide*: percentage of glass content extracted from Table C1, x_i and y_i : factors which have been derived from measured heat contents of glass samples using standard linear regression (see Table C2a), *T*: batch melting point (see equation C5), *b*: gas emitted during reaction, m_i : mass fraction of a given component of process emissions, a_i , b_i , c_i , and d_i : coefficients of heat capacity (see Table C3), $\Delta H_{d,i}$: enthalpies of dissociation of raw materials to their respective oxides for the production of the silicate glasses under study (see Table C2b), $\Delta H_{f,1}$ = -3303 kJ/(kg CaO), $\Delta H_{f,2}$ = -3576 kJ/(kg Na₂O), and $\Delta H_{f,3}$ = 152 kJ/(kg SiO₂), and M_{Na2O} , M_{CaO} , M_{sio2} : molar mass of the respective oxide.

The parameters *rest-Na*₂O and *rest-SiO*₂ in equations (C10) and (C11) denote the respective amounts of Na₂O and SiO₂ which are not consumed in the following reactions:

Table C2 (a) Factors for calculating ΔH_{glass} ; (b) Enthalpies of dissociation of raw

$$Na_2O + 3CaO + 6SiO_2 ⇔ Na_2O.3CaO.6SiO_2, ΔH_{f1}=-3303 kJ/(kg CaO)$$
 (C12)

$$Na_2O + 2SiO_2 \Rightarrow Na_2O.2SiO_2, \Delta H_{f2} = -3576 \text{ kJ}/(\text{kg Na}_2O)$$
 (C13)

Oxides	Xi	y i
SiO ₂	67.67	-0.014
Na ₂ O	-133.30	-0.027
CaO	-264.00	0.148
MgO	-224.40	0.124
AI_2O_3	-365.10	0.250

materials (Madivate, 1998).

Compounds	ΔH _{d,i} (kg/kg raw)	
Na ₂ CO ₃	3040	
CaCO ₃	1796	
CaMg(CO ₃) ₂	1649	
$Na_2SO_4^{++}$	4063	
AI(OH)₃	1188	

Table C3 Coefficients of the heat capacity equation for calculating ΔH_{gas} .

Gases	a _i	b _i	Ci	di
CO ₂	1.162	0.099	0.033	-
SO ₂	0.780	0.074	0.016	-
NO ₂	0.750	0.536	0.009	0.149
02	0.911	0.202	0.006	0.032
H ₂ O	1.910	0.436	0.024	-

Assuming that the cullet does not undergo any transformation during melting, the equation (C6) can be expanded for including the impact of cullet in TER. Therefore the TER value for cullet-containing batches becomes:

$$TER' = ((1-c)*TER + c*(1+b)*\Delta H_{glass})/(1+c*b) [kJ/kg of glass melt]$$
(C14)

Fuel combustion

Combustion emissions per unit of energy are attributed to the fuel input, the most common of which is methane processed from natural gas (Butler et al. 2011). The c_p 's of the natural gas, air compounds and combustion emissions correspond to the temperature window between ambient and furnace temperatures (Table C4). The indirect CO₂ emissions are calculated separately using country-specific CO₂ emission factors related to the electricity production (approx. 0.5kg/kWh) (VNPI, 2018; WEC Report, 2018).

Compound	Composition (vol %)	Gross C.V. (MJ/m3)	Molar mass (kg/mol)	Heat capacity (kJ/K*mol)	Density (kg/m³)	
Groningen Natural gas						
CH₄	81.30	39.82	0.016	4.099	0.668	
C ₂ H ₆	2.88	70.31	0.030	3.398	1.264	
C ₃ H ₈	0.39	101.23	0.044	3.172	1.882	
C ₄ H ₁₀	0.16	133.69	0.058	3.148	2.489	
C ₅ H ₁₂	0.04	169.27	0.072	3.128	3.390	
N ₂	14.33	-	0.028	1.150	1.165	
02	0.01	-	0.032	1.037	1.331	
CO ₂	0.90	-	0.044	1.097	1.977	
Atmospheric air						
N ₂	78.10	-	0.028	1.150	1.165	
02	20.94	-	0.032	1.037	1.331	
Ar	0.93	-	0.040	0.907	1.661	
CO ₂	0.03	-	0.044	1.097	1.977	
Exhaust gases from Natural gas combustion						
N ₂	67.25	-	0.028	1.150	1.165	
02	0.001	-	0.032	1.037	1.331	
Ar	0.78	-	0.040	0.907	1.661	
CO ₂	8.39	-	0.044	1.097	1.977	
H ₂ O	23.57	-	0.018	2.288	0.554	

Table C4 | Composition of natural gas and atmospheric air (Gasunie, 1980).

Heat Balance

Heat balance is considered as a guideline for determining energy conservation in a glassmelting furnace since it can monitor heat losses and furnace efficiency (Tapasa et al. 2012). Factors that have a major influence on energy consumption and emissions are fuel used, furnace age and capacity utilisation, cullet percentage, mixed batch recipe and degree of residual heat recovery. By recovering minimum losses from flue gases, energy savings is obtained regarding a system with process recovery, combustion recovery, and recovery of both. In regenerative furnaces, the heat effects are balanced in the following manner:

$$Q_{\text{fuel}} - Q_{\text{flue}} + Q_{\text{preheat air}} = Q_{\text{U}} + Q_{\text{loss}} - Q_{\text{preheat batch}}$$
(C15)

where Q_{fuel} : primary energy, Q_{flue} : heat carried by exhaust gases, $Q_{preheat air}$: energy reintroduced from the regenerator, Q_U : theoretical energy requirement, Q_{loss} : sum of combustion space and tank losses, $Q_{preheat batch}$: heat carried by batch mix. The left-hand side of Eq. (C15) expresses the heat released in the combustion space and the right part denotes the heat consumption in the furnace (Khoshmanesh et al. 2007).

To maintain a constant temperature in the furnace, a certain rate of heat transfer from the combustion gas is required. Assuming that combustion takes place under adiabatic conditions, the energy balance for the flow through the furnace becomes:

 $CV + H_R = H_P + Q_u + Q_{loss} \xrightarrow{[adiab.]} m_{NG}*LHV + m_R*C_{pR}*(T_{preh} - T_o) = m_P*C_{pP}*(T_{flame} - T_o)$ (C16)

where *CV*: net calorific value⁵², *H_R*: sensible heat of reactants (i.e. fuel and oxidant), *H_P*: heat of products (i.e. flue gas), *T_o*: ambient temperature, and *T_{flame}*: theoretical flame temperature. More specifically, *T_{flame}* represents the practical upper limit of flame temperature when fuel and oxidant is mixed rapidly and ignited (Baukal, 2013). Preheating the reactants (e.g. atmospheric air) leads to higher flame temperatures, hence increased heat transfer in the system and better effectiveness of the regenerative heat exchanger.

If combustion takes place with an oxygen rich gas, high flame temperatures can be reached as the mass of flue gas is considerably reduced. This causes the enthalpy difference between the flame-temperature gas and the exhaust to become higher (Ming et al. 2003). For the same heat transfer, this would lead to a lower required mass flow rate which is proportional to fuel consumption. Therefore, fewer carbon dioxide is formed as a result of burning less fuel.

⁵² Net calorific value (approx. 31.669 MJ/m³ Groningen Natural gas) is appropriate for predicting the temperature reached within the flame. As a result, the enthalpy of exhaust gases will consist of sensible heat terms only.



Figure C1 (a) Flame temperature versus air-fuel ratio (Boateng, 2016); (b) Effect of the increased O_2 concentration on the flame temperature (Hendershot, 2010).

The calculation of energy requirements is iterative, and depends on the assumptions made for both the combustion agent and the heat recovery options. When the modelling paradigm prevents from having a variable energy input, then the exploited wasted heat $Q_{preheat air}$ and $Q_{preheat batch}$ can be subtracted from the primary Q_{fuel} with a deviation <3.5% from the actual situation. More information on combustion with oxygen is provided in Section 4.3.