

Environmental aspects

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12.1 General

The use of metals and other materials has increased greatly in the last century, and this growth is expected to speed up further as the world transitions towards less fossil dependency. The technologies needed to combat climate change require vast amounts of base metals, but also many less common materials, such as lithium and rare earth elements.

Silicon is the raw material for many of the technologies enabling future sustainability and digitalization, such as semiconductors and electronics, batteries, and solar cells. Silicon-based alloys are also important additives in the production of iron and steel, aluminium, silicone rubbers, and many other chemical products. While the volume of silicon alloy production is minor compared to the base metals, the production of additives such as silicon must be included when assessing the total environmental impact of any base metal component or structure.

Nearly all aspects of the production and use of high-silicon alloys are energy-intensive, and most of the process steps in production emit pollutants of some sort, not least greenhouse gases (GHG) and dust. To minimize the environmental footprint, it is important to make use of the Best Available Technology (BAT) for every step of the production process. This chapter discusses some of the most important environmental parameters, with emphasis on the process-specific parameters of the primary production unit, i.e. the smelter.

12.2 Occupational health

Workers in industrial settings may be exposed to various types of stress factors and pollutants, and the silicon industry is no exception. Generally acknowledged stress factors include heat exposure, noise, and night work, which are also common in this industry. More process-specific issues are typically related to personnel exposure to polluting substances from the different processes in the plant.

Fugitive or diffuse emissions are any type of emission that is not controlled or directed. However, these terms are often used only to designate airborne particulate matter. It is important to note, however, that any type of pollutant (gases or particles) can be diffuse or fugitive in nature. This means that they may affect the health of people not only working in a smelter, but also residents in the nearby area of the plant. Nevertheless, where occupational hygiene and local community air quality are concerned, dust (airborne particulate matter) is especially important.

Historically, the main visible effluent from silicon alloy smelters was dust consisting of amorphous silica particles. This dust was generated by the furnace process and emitted through chimney pipes. Unfortunately, fume could also leak into the furnace building and pollute the working atmosphere. This type of dust is still generated under the furnace hood, but today it is effectively collected in filters and often sold as a by-product ('silica fume'), which is used as a filler material, e.g. in concrete. However, furnace fume is just one example of the thermally generated dust commonly encountered in silicon smelters. Similar fumes are also produced by other hot processes, such as oxidative ladle refining, casting, and tapping; these also constitute potential sources of fugitive dust emission.

In many respects, exposure to dust and fume still constitutes the largest health risk and hygiene challenge for silicon smelters. Long-term inhalation of silica fume has been linked to cancer, pneumonia, chronic obstructive pulmonary disease (COPD), and other respiratory and cardiovascular syndromes (Hobbesland *et al.*, 1997a, 1997b; Johnsen, 2009; Johnsen *et al.*, 2008a, 2008b; Søyseth *et al.*, 2013, 2011a, 2011b).

12.3 Pollution control

Process emissions are often divided into three main categories: pollution to air, soil, and water. From the silicon process, air pollution is most significant. On the other hand, emissions to the air may later be converted into soil or water pollution.

As the fate of the various process streams becomes known, a mass balance for each element can be an efficient tool for describing the level of pollution. This method is commonly used for environmental reporting to authorities, for example emissions of heavy metals and sulphur. Sometimes generic emission factors have been established for a process or industrial branch, so that reporting can be simplified. Typically, most production emissions are calculated estimates based on the raw material mix, production tonnage, and emission factors. Control measurements are then carried out to verify the calculated numbers.

However, this approach is not always adequate. Some emissions are a result of the process itself and do not depend on the amount or mixture of elements fed to the process. One example is the NO_x emissions from the silicon process, where NO_x mainly forms from a reaction between the nitrogen and oxygen present in the cooling air drawn into the high-temperature zone of the furnace hood.

The main sources of pollution in the silicon process are:

- Electric energy delivered to the process. A variety of environmental problems may arise from the production of electricity.
- Reduction materials – coke, coal and woodchips. The carbon necessary for the reduction process will inevitably be converted to CO , CO_2 and, to a lesser extent, various hydrocarbon gases. Other elements in the reductants will be distributed between the off-gas, dust, slag, and alloy (as shown in Chapter 4).
- Quartzite minerals. The silica sources normally contain minor elements besides the SiO_2 . These elements will be distributed between the off-gas, dust, slag, and alloy (as shown in Chapter 4).

There are clear dependencies between process parameters and certain types of pollution from the production processes. This means that, in some instances, changing the process parameters may reduce process emissions. The single most important process parameter is the silicon yield. An improved silicon yield will lower the amount of any input material or energy *per tonne product*, for example the *specific* energy consumption. This may lead to a reduction of many other specific parameters.

Emissions to soil and water

Whenever a solid waste or by-product is produced and disposed of, the risk of water or soil pollution must be considered. Before waste materials may go to any form of deposition, the risk of leakage must be assessed. For high silicon alloy smelters, the materials that may be deposited include slag, dust from the filters, spent refractories, and other minor by-products and waste materials. Skull formation in a ladle may also create an off-grade mixture of refractory, which must be disposed of.

Importantly, many airborne pollutants may be carried into soil or water systems by atmospheric deposition and rain.

Emissions to air

The issues of airborne emissions from high silicon alloy production are, in many ways, similar to the challenges of airborne emissions from other metallurgical industries, most notably the inherent energy intensity and the greenhouse gas emissions from both electricity production and the carbonaceous reducing agents. Certain challenges are, however, highly branch-specific, for example the dust types generated and the management of NO_x emissions by furnace design and operation.

Unwanted discharges to the air tend to be the most common environmental effect of silicon production. This type of pollution consists of both gases and airborne particulate matter (PM; dust). This chapter will focus on CO₂, NO_x, SO₂, PAH, PM, and trace elements, which are some of the most important and common types of airborne emissions from high silicon alloy production.

CO₂ and other greenhouse gas emissions

While CO₂ is by far the most important greenhouse gas (GHG) in high silicon alloy production, other GHGs may also be emitted. During combustion of the reductants and electrodes, volatile hydrocarbons may form and contribute to GHG emissions. The most important of these volatile organic compounds (VOC) is methane (CH₄). Since methane has a higher radiative forcing than CO₂, even relatively low concentrations may contribute significantly to the calculated CO₂ equivalents from a plant.

As described in previous chapters, the production of high silicon alloys requires the reduction of quartz using carbonaceous reductants like coal, coke, charcoal, and wood chips. In open or semi-closed furnaces, carbothermic reduction first generates CO gas, which then oxidizes to carbon dioxide (CO₂) at the furnace charge top. Additional CO₂ can also be generated inside the furnace charge, for example by the Boudouard reaction or by the reduction of iron oxides in FeSi production.

The reducing agents coke and coal are the main contributors to the GHG emissions, but other carbon-based materials, such as the electrodes and electrode paste, will also contribute. Hydrocarbon emissions are highly dependent on both alloy type and operation, which in turn leads to large variations and uncertainties in the reported data. Kero *et al.* (2017) found literature discrepancies of the order of a factor of 10.

Greenhouse gas emissions mainly depend on:

1. The type of alloy produced. Reduction of quartz requires more energy than most other oxides, e.g. iron oxides, which means higher consumption of both carbon and electricity. Thus, the higher the Si content, the higher the GHG emissions.
2. The carbon material mix. The levels of fixed carbon and volatile matter depend on the choice of reductants, which in turn affects GHG emissions. Note that emissions from biogenic carbon reductants are often exempt from GHG emission estimates.
3. The furnace operation and charging method strongly influence emissions, in particular of CH₄ and NO_x. More even charging will generally reduce emissions compared with batch-wise charging (Grådahl *et al.*, 2007; Kamfjord, 2012; Myrhaug *et al.*, 2012).

Lindstad *et al.* summarized official GHG emission factors for FeSi/Si products and reductants in the 2006 IPPC Greenhouse Gas Inventory Guidelines (Lindstad *et al.*, 2006).

Sulphur dioxide, SO₂

In the silicon alloy industry, SO₂ gas emissions are caused by the oxidation of sulphur contained in the raw materials, primarily the reducing agents. Reported emission levels are typically calculated based on material balances. Abatement methods for post-filter cleaning of SO₂, mainly limestone based, are available and are increasingly being installed in new silicon plants. While SO₂ is emitted continuously during combustion and reaction of the carbon materials, peaks of SO₂ and CO gas emissions have been shown to correlate with furnace events such as ‘avalanches’, i.e. the collapse of the charge burden near the electrodes (Grådahl *et al.*, 2007).

Nitrogen oxides, NO_x

Nitrogen oxides (NO and NO₂, the sum of which is referred to as NO_x) contribute to a number of environmental problems, including acid rain, ozone smog, eutrophication (stimulated growth of algae and bacteria), and bronchial suffering in animals and humans. NO_x may form by three different mechanisms, known respectively as prompt, fuel, and thermal generation. In silicon furnaces, the fuel and thermal formation mechanisms dominate. Fuel NO_x is formed by oxidation of the nitrogen components present in the solid raw materials. Thermal NO_x, on the other hand, is formed by direct oxidation of nitrogen from the air at temperatures above 1400°C. Such temperatures are frequently observed in the furnace hood (Kamfjord *et al.*, 2012; Ravary and Johansen, 1999; Solheim *et al.*, 2012).

Combustion of gaseous SiO above the charge surface and in the taphole may also locally increase the temperature. The amount of SiO(g) released from the charge will therefore influence NO_x formation, while measures that reduce SiO also seem to reduce NO_x emissions. NO_x production is therefore inversely proportional to the silicon yield (since low Si yield means high SiO losses), at least up to a certain level of silica fume formation. Furnace design and furnace operating procedures, such as stoking and charging, heavily influ-

ence NO_x emissions (Brede, 2013; Kamfjord *et al.*, 2012; Panjwani *et al.*, 2011; Ravary *et al.*, 2007; Ravary and Johansen, 1999; Solheim *et al.*, 2012; Solheim and Kamfjord, 2013). NO_x formation also correlates with the moisture content of the furnace gas and seems to decrease with increasing moisture content (Slizovski *et al.*, 2014).

The effect of the furnace hood design and the inlet of false air are key to understanding NO_x generation. The amount of air and its flow path throughout the hood determines when, where, and whether oxygen and nitrogen are mixed for a sufficiently long time in a sufficiently hot zone to produce thermal NO_x. Optimization of furnace hood designs can therefore decrease NO_x emissions but has proven to be a very complex task. As the trial-and-error approach is both time- and cost-consuming, modelling capabilities and tools for NO_x prediction are very desirable. It is possible to reduce NO_x emissions by means of furnace operation, process management, and/or by modifying the SAF system itself (Echterhof and Pfeifer, 2012, 2011; Kirschen *et al.*, 2005). For high silicon alloy production, this typically implies:

- Active cooling of the primary flame zone to control the combustion temperature (Tveit *et al.*, 2004).
- Avoid avalanches and other furnace events which may allow SiO-rich gas to ‘blow’ through the charge surface.
- Frequent stoking and continuous or semi-continuous charging. Grådahl *et al.* (2007) found that implementation of best practices could reduce NO_x emissions by 50%.
- Recycling the flue gas to reduce excess air above the charge.

Particulate matter (dust) emissions

Almost all processes involved in silicon alloy production produce particulate matter (PM; dust) in some form. In this chapter, the terms particulate matter and dust are used as synonyms and primarily describe airborne solid particles. The term fume relates to thermally generated particles. The largest source of dust is the furnace, where combustion of escaping SiO gas from the furnace hot zone results in the formation of SiO₂ particles, known as silica fume, above the charge surface. Modern ventilation and filter systems have enabled efficient collection of this type of furnace dust, which also constitutes

a profitable by-product, sometimes called microsilica. Furnace fume consists of agglomerates of amorphous SiO_2 spheres and includes both fine (FP) and ultrafine particles (UFP), i.e. particles with aerodynamic diameters of $< 2.5 \mu\text{m}$ and $< 0.1 \mu\text{m}$, respectively (Dingsøyr *et al.*, 1992; Myrhaug and Tveit, 2000; Næss, 2013; Tveit and Myrhaug, 2000).

All the major sources of fugitive dust emissions, both dust remaining inside the plant and dust escaping out of the plant, involve the liquid alloy. Tapping, refining, casting, and other operations where high-temperature liquid alloy is in contact with air produce silica fume with many similarities to that produced during the oxidation of SiO originating from the reduction of quartz in the furnace (Kamford, 2012; Kero *et al.*, 2015, 2013; Næss, 2013). During oxidative ladle refining (OLR), the main mechanism for dust formation is the active oxidation of the liquid silicon alloy, while splashing (droplet expulsion) may contribute a smaller amount ($< 1\%$) of coarser particles (Næss *et al.*, 2012a). Active oxidation occurs in two steps: surface silicon first reacts with oxygen to form SiO gas, which then oxidizes further to SiO_2 (Næss *et al.*, 2013, 2012b, 2012a, 2011). The kinetics of this process are governed by oxygen access to the alloy surface and are therefore highly dependent on the dynamics and temperature of the alloy surface exposed to air (Olsen *et al.*, 2012, 2011).

Dust from the handling and transport of solid materials, such as the products and the raw materials, is fundamentally different from dust generated by active oxidation. Dust from handling solid materials is typically coarser, and its physical and chemical properties depend on the material from which it was generated. Raw material handling and transport can, for example, produce airborne crystalline alpha-quartz, which is a health hazard in its own right. Likewise, metallic dust (as opposed to oxide dust) can become explosive under certain circumstances, which constitutes a safety risk (see section on safety).

Particle-bound organic compounds including Polycyclic aromatic hydrocarbons

Polycyclic aromatic hydrocarbons (PAH) are organic compounds characterized by molecular structures with more than two aromatic (benzene) rings. Lighter compounds, with fewer aromatic rings, are gaseous at room temperature, whereas heavier compounds are liquid or solid. Hence, the larger molecules often follow the particulate matter in the off-gas system and are

collected in the filters. Particle-bound PAH may be either condensed PAH or PAH adsorbed onto particles (such as silica fume or soot).

From an occupational health point of view, PAH receive special attention among the organic emissions because some PAH compounds are linked to cancer. The term PAH is often used to denote only a particular subset of PAH compounds, more correctly known as the EPA-16. These 16 compounds have been identified by the United States Environmental Protection Agency (US EPA) due to their potential to induce adverse effects on the environment and health. Although the EPA-16 are the standard for emission reporting, determination of an extended list of PAH in the off-gas from silicon production has indicated that the EPA-16 account for only a fraction of those emitted (Arnesen *et al.*, 2024, 2023; Kero and Indresand, 2022).

Man-made PAHs are typically formed by incomplete combustion of organic materials. In silicon alloy production, the combustion of reductants in the furnace is the main source of PAH. Another important upstream source is the baking of electrodes. PAH formation is linked to soot formation, which in turn is influenced by furnace design and operation and varies throughout the charging cycle (Tveit *et al.*, 2004). As PAHs are destroyed at high temperatures, emissions can be significantly reduced by increasing off-gas temperatures. It is noteworthy, however, that increasing the off-gas temperature may have other unwanted side-effects, such as increased NO_x emissions (Grådahl *et al.*, 2007; Kero *et al.*, 2017).

There are several other types of complex, toxic, organic pollutants that may form in the furnace process. The single most important strategy to reduce emissions of complex organic compounds like dioxins, furans and PAH is to optimize furnace operation to ensure high enough temperatures and sufficient oxygen for complete combustion. Today's modern, semi-closed SAFs allow very efficient destruction of these types of compounds.

Particle-bound inorganic compounds: Trace elements and heavy metals

Trace elements, including heavy metals, enter the production process as low-concentration constituents in the raw materials and electrodes. In the process, the elements become redistributed to metal, slag, dust and gas. Emissions thus depend on the raw material mix, the alloy composition and the process

temperatures. At temperatures of 1600°C or higher, certain metals such as Zn, Pb, Cd, Na, Mn and Fe go into the gas phase and may escape as metal vapour. When the off-gas temperature drops, the metal vapours condense and may be collected with the dust. Myrhaug and Tveit (2000) showed that a boiling point model can be used to predict the redistribution of an element in the furnace, as discussed in Chapter 4. Næss *et al.* (2014) showed that the model is also applicable to the refining ladle, with some modifications due to the oxidation of elements.

Regulation of industrial emissions of heavy metals and other trace elements varies between countries and regions. The United Nations Environment Programme (UNEP) focuses on lead, cadmium and mercury, but many national authorities require emission control and reporting of many more elements, for example As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Se and Zn. Reported emissions of trace elements are often based on material balances and may vary greatly between plants.

Among airborne heavy metal emissions, mercury is of particular environmental concern, and international legislation has long been stringent with respect to Hg emissions. In silicon alloy plants, particulate control devices (e.g. fabric filter or wet scrubber) capture particle-bound mercury. Elemental, more volatile mercury may, however, be emitted. Emission estimates to air through the filter systems should cover both gaseous and particle-bound heavy metals, but the low concentrations of these elements represent a major challenge with respect to analysis. The detection limit (DL) for Hg is often in units of parts per billion (ppb), whereas the other heavy metals typically have DLs of the order of magnitude of parts per million (ppm). Large uncertainties are also related to sampling and representativeness of samples. The total uncertainty for elements such as Co, Hg and Mo is usually around or above 100% (Næss *et al.*, 2014, 2013). The very significant measurement uncertainties often make it impossible to ‘close’ the material balance for individual elements.

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