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Editorial: Upgraded metallurgical grade silicon: quality, applications, and process economics

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KEYWORDS

silicon, metallurgical grade, solar cells, photovoltaics, feedstock

Editorial on the Research Topic
Upgraded metallurgical grade silicon: quality, applications, and process economics

This special edition of FRONTIERS was intended to review the use of *upgraded metallurgical grade silicon* (UMG-Si, UMGS, or UMG) in solar cells. Let us start with some remarks on the terminology and look back on history. Silicon, the most abundant element on the Earth`s crust after oxygen, is never found in mother nature as native (Si^0) but always associated (Si^{4+}) with oxygen (or in some cases, fluorine) in quartz (SiO_2) and silicates (e.g., Na_2SiO_4, $CaSiO_4$, $MgSiO_4$, and $Al_2(SiO_4)_3$ and fluorosilicates ($M_nSi_2F_6$). Silicon and silicon-rich alloys, such as ferrosilicon and silicomanganese, are industrially produced by the carbothermic reduction of silica (quartz and quartzite). Silicon with a purity of 98%–99% can be obtained in an electric arc funace with submerged electrodes by this overall reaction:

$$SiO_2 + 2C \rightarrow Si + 2CO.$$
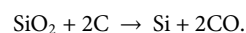
The process is more than a century old, well proven, and optimized with respect to many performances. The product tapped from the furnace is commonly called *silicon metal* (98%–99% purity) because of its appearance and despite the fact that silicon, by chemical classification, is not a metal but a metalloid. Its first commercial and industrial application was as an alloying element to aluminum, a metal discovered simultaneously with silicon in the second part of the 19th century. Massive use of aluminum alloys started around the Second World War, with the demand being stimulated by the aircraft industry. Referring to application in metallurgy, the product was designated as *metallurgical grade silicon* (MGS or MG-Si), the main impurities (1%–2% in total) being Fe, Ca, and Al. The latter impurity mentioned has no significance when silicon dissolves in molten aluminum and, therefore, is never counted in the specifications by this industry.

Approximately at the same time as the Second World War emerged the *silicones* or polysiloxanes, a new class of inorganic polymers forming oils, elastomers, and resins exhibiting remarkable and unique properties. The backbone of the polysiloxanes consists of silicon atoms alternating with oxygen atoms, each silicon atom carrying two alkyl groups, most commonly methyl groups. The manufacture of silicones starts with silicon metal as the main raw material. The purity required for silicon is 98%–99%, that is, the same level as for

aluminum alloys, but in the case of siloxanes, Al, Ca, and the ratio between the concentration of both elements (Al/Ca) must be carefully controlled to achieve high productivity and reactivity in the direct process of making the monomer precursor (dialkyldichlorosilane). The exact composition of impurities is achieved by oxidative refining of the tapped molten silicon in the ladle before casting/solidifying and crushing. The silicon grade dedicated to the silicones is sometimes designated as *chemical grade silicon* by vendors and buyers in this industry. However, this distinction is not made in the semiconductor and photovoltaic industry, making use of silicon of far higher purity, and all silicon grades below 99% are, by extension, designated as MGS, regardless of whether the application is metallurgy or chemistry.

With the invention of the transistor and the development of solid-state electronics in the 1950s and onward, ultra-high pure silicon was required. Impurities had to be at the part per trillion (ppt) level. Early in the 1960s, a process was widely adopted to achieve this challenging target. In this process, silicon (MGS grade) is reacted with chlorine (or hydrogen chloride) to form chlorosilanes (preferably trichlorosilane), easily volatile compounds that can be purified by distillation and thus enable the required purity for semiconductor silicon (i.e., between 99.99999% and 99.999999%). The chlorosilane is then reduced to elementary silicon of ultra-high purity by chemical vapor deposition (CVD) at a high temperature (approximately 1,150°C) on a heated filament (preferably of silicon). This process is commonly known as the *Siemens process*, according to the name of one of the companies developing it around 1960. The resulting product is shaped as long rods, which can be used in float zone (FZ) single-crystal pullers or as broken chunks and chips in Czochralski (CZ) single crystal pullers. The product is commonly called *polysilicon* due to its polycrystalline structure and in opposition to the monocrystalline structure obtained through the FZ and CZ pulling. It is also designated as *electronic grade silicon* (EG-Si or EGS) by comparison with MG-Si and other silicon grades. The single crystals successfully grown and qualified are further processed to wafers. From these wafers, both solar cells and integrated circuits can be obtained.

The oil crisis in the first part of the 1970s pushed the development of alternative energy sources to petroleum. Solar energy was identified as a major challenger to oil. The first solar cell was manufactured in Bell Telephone Laboratories (now Bell Labs) in the United States in 1954 with an efficiency of approximately 6% (Chapin et al., 1954) and started the development of photovoltaics (PVs). Silicon solar cells made with the silicon technology known from the semiconductor industry were initially produced for aerospace satellites with little attention paid to costs. With the oil crisis striking in the 1970s, ambitious programs to develop commercial terrestrial PVs were launched in the United States (by the Department of Energy/ DoE), in Japan (Ministry of Economy, Trade and Industry/METI), and in selected European countries (European pioneers were Italy and France) to make solar electricity competitive with oil. In the mid-1970s, a symbolic cost target assigned by the DoE to the developers was 1 US dollar per watt-peak (W_P) on the module level, an objective that took 30 years to reach. These ambitious programs and objectives triggered the development of low-cost silicon of purity suitable for solar cells, that is, not as high purity as

for semiconductors but much purer than MGS. The concept of *solar grade silicon* (SGS or SoG-Si) arose without an exact definition of purity. Fifty years later, the question remains quite open; 1 ppm (weight) was indicated as a possible target (i.e., 99.9999% Si), but most important was the content of donors (mainly phosphorus) and acceptors (mainly boron). One avenue to achieve this goal (moderate purity at low cost) was to use large-scale, well-proven metallurgical techniques, which are supposed to be cost-efficient and scalable to the level needed for the future deployment of PVs. The demand was calculated in millions of tons at the time EG silicon was produced in less than 10,000 tons a year. It was argued that the only way to achieve large volumes was by the metallurgical route. SGS produced by the metallurgical route, as opposed to polysilicon by the chemical/ CVD/Siemens process, was designated as *upgraded metallurgical grade silicon* (UMGS or UMG). The chemical route through volatile silicon precursors was also challenged to develop alternative variants compatible in terms of purity and cost with the demand of terrestrial PV deployment. Fluidized bed reactors (FBRs) as an alternative to hot filament CVD in bell-jar Siemens reactors and new silicon precursors (e.g., monosilane SiH_4) were proposed and became the topics of advanced R&D programs conducted in parallel with UMG for the benefit of the PV industry. Remarkable results have been achieved over the past 50 years. For more information on the history of silicon science, industry, and market, the interested reader is invited to consult our publications and the multiple references herein (Ceccaroli and Lohne, 2003; Ceccaroli et al., 2012a; Ceccaroli et al., 2016).

Regarding the upgraded metallurgical route (or UMG), several strategies were pursued. Combining several steps appeared necessary to reach the optimal goal of purity and cost. Most consideration has been devoted to the following:

- Selection of the raw materials (various types of silica ranging from ultra-pure quartz to silica-rich rice husk) and reductants (including synthetic carbon black). This allowed for a low content of donors (P) and acceptors (B) in the silicon.
- Alternative furnaces to adjust to high-purity raw materials, including two-step furnaces going via silicon carbide as intermediate.
- Advanced refining techniques in the ladle to remove by oxidation all elements less noble than silicon (e.g., Ca, Al, and C). This includes gas, slag, plasma, electron beam treatment, and more.
- Aluminothermic reduction replacing carbothermic reduction.
- Alloying silicon with other metals, for example, Al, Ca, Ba, Mg, and forming intermetallic phases or eutectics with silicon able to absorb and concentrate the impurities, dissolving these intermetallic phases and impurities by acid leaching. This technique was proven efficient in removing Fe and transition elements, which are all detrimental to solar cell performance. It was also proven to partially remove P.
- Directional solidification (DS) playing on the distribution of elements with low segregation coefficients. Obeying mechanisms similar to those for the formation of intermetallic phases in alloys, DS is shown to be particularly efficient for removing metallic (transition) elements as well as P and C.

The metallurgical route never achieved the same level of purity as the chemical route to polysilicon, especially regarding P and B, which are, respectively, a charge donor and an acceptor to silicon. However, playing on "*compensation*" (neutralizing excess of one charge carrier by its opposite until the net concentration of the suitable charge carrier is reached) and mitigating imperfection by "*defect engineering,*" the industrial developers, hand in hand with academic researchers, developed materials (wafers) and cell architectures performing at par with solar cells made of higher-purity polysilicon. The best results were achieved when combining UMG with multicrystalline cast silicon, a far less costly and energy-consuming technique than monocrystalline CZ silicon. Regarding UMG performance and limitation, in addition to the chapters in books referred by Ceccaroli and Lohne (2003), Ceccaroli et al. (2012a), and Ceccaroli et al. (2016), more specific information is gathered in some others of our publications (Øvrelid et al., 2012; Ceccaroli et al., 2012b; Coletti et al., 2011; Bye and Ceccaroli, 2014; Forniés et al., 2019).

This Research Topic of FRONTIERS delivers two review articles showing great details on this strategy.

- *Production of UMG silicon for low-cost, high efficiency, and reliable PV technology* by Novoa et al. In this article, the authors present an innovative technology for purifying silicon, specifically designed for solar applications. This technology spans the entire value chain, from the raw material feedstock to the final photovoltaic (PV) system. At its core, the process involves several metallurgical steps, that is, slagging, evaporation, and solidification, that upgrade metallurgical silicon into upgraded metallurgical silicon (UMG-Si). This purified material contains low levels of impurities, including boron, phosphorus, and metals, all of which are compatible with solar cell production. The obtained multicrystalline Si ingots show uniform resistivity along their height due to the addition of gallium for compensated resistivity. Solar cells using this material have achieved efficiency levels of 18.4% for Al-back surface field (BSF) and 20.1% for passivated emitter and rear cell (PERC) technologies. These cells also show promise for future advancements, with potential efficiencies of up to 22% in next-generation solar cell designs. Beyond performance, UMG technology stands out for its environmental benefits. Compared to conventional polysilicon, it delivers a significant reduction in climate change emissions by over 20% and cuts the energy payback time by 50%. These advantages make UMG-Si a cost-effective, low-CAPEX, and environmentally friendly alternative for producing PV silicon.
- *A review of defect mitigation strategies for UMG-Si wafers* by Basnet and Macdonald. In this work, the authors illustrate different defect mitigation strategies like tabula rasa, gettering, and hydrogenation for improving the performance of UMG-Si wafers and UMG-Si-based solar cells. While UMG-Si still lags behind conventional silicon, some of these strategies can be applied together with further advancements in purification and ingot growth, which are expected to boost UMG-Si-based solar cell performance. Continued research and detailed techno-economic studies will be key to fully realizing UMG-Si's potential as a cost-effective, competitive alternative in the solar industry.

A third article proposes the use of magnesium as an impurity-dissolving agent, completing the knowledge on more common calcium and aluminum as alloying metals.

- *Effect of Mg-alloying and cooling rate in the microstructure of silicon* by Zhu et al. They report on the effects of magnesium alloying (5.5 wt% and 9.0 wt%) and cooling rates (3°C/min, 10°C/min, 25°C/min, 40°C/min, and 80°C/min) on the microstructure of MG-Si. Faster cooling and higher magnesium concentrations reduced the primary silicon grain size, with faster cooling leading to more equiaxed grains and slower cooling producing elongated platelet shapes. The microstructure investigation shows an increased crystallographic orientation heterogeneity with higher cooling rates. This study emphasizes optimizing cooling rates and alloying content for efficient silicon purification, balancing cost and process effectiveness.

Production of wafers from silicon ingots (both multi- and monocrystalline) generates much silicon loss in the form of small particles commonly called *kerf*. It is assumed that 35%–40% of the incoming silicon feedstock ends up as kerf. The last development in sawing, that is, abrasive-free sawing using diamond-coated steel wire and an organic lubricant, generates kerf of high chemical purity that is relatively easy to clean by acid washing and more uniform wafer thickness. The widely used former technique based on uncoated steel wire entraining SiC abrasives in a liquid suspension (slurry) generated a heavily contaminated silicon mud of low commercial value. The abrasive-free kerf can be used in high-value applications for silicon, such as Li-ion battery anodes.

The fourth article in this Research Topic illustrates this recent opportunity and shows how to exploit major knowledge acquired through UMG development.

- *Silicon kerf-loss as potential anode material for Li-ion batteries* by Søiland et al. This study shows that industrial silicon kerf particles, with a size of ~700 nm and a 1–2 nm oxide layer, can serve as an effective alternative to nano-sized silicon (40–100 nm) in lithium-ion battery (LIB) anodes. The kerf particles demonstrated comparable initial capacities, efficiencies, and cycling stability, maintaining performance beyond 120 cycles. The findings suggest that silicon kerf could offer a low-carbon footprint alternative to nano-silicon, with the potential for further improvements in cycling stability through methods like silicon–carbon composites or particle coatings.

Initiating this special edition, we invited several scientists from academia and industry who we knew had been active and productive in the UMG field. However, the number of enthusiastic responses was rather disappointing. We regret that the present edition does not reflect the tremendous efforts devoted to the research topic during the past five decades. Although regrettable, it nonetheless reflects its current commercial status well. In 2014, we published the list of the most advanced UMG projects. The companies we referred to were JFE (Japan), Dow Corning (Brazil), Timminco (Canada), Elkem Solar (Norway), Jaco (China PR), Silicor Materials (Canada, United States), Photosil (France) Ferroatlantica/Ferrosolar/

Ferroglobe (Spain), and Evonik/Solsic (Norway). Several of these projects were already either terminated or on hold (Bye and Ceccaroli, 2014).

Ferroatlantica/Ferrosolar/Ferroglobe (Spain) and especially Elkem Solar/REC Solar (Norway) were, after 2014, among those pursuing the most continuous efforts in R&D and industrialization. We supported a publication of Ferrosolar as late as 2019 (Forniés et al., 2019). Ferrosolar was planning a plant in Spain, which was unfortunately never completed. In 2010–12, Elkem Solar commissioned a commercial UMG plant in Norway with a capacity of 5,000 tons, which was temporarily shut down and reshaped to different process concepts several times. Elkem Solar sold the plant to REC Silicon, who further sold it to Reliance of India. In 2020, REC Solar in Norway made a last full attempt to make use of its UMG technology and plant equipment, turning some of the huge amounts of silicon kerf generated by wafer manufacturing into single-crystal feedstock. Announcements by the company concluded that the full validity and overall benefit of the method, energy consumption, and carbon footprint were advantageous and had positive environmental and economic impacts. The project demonstrated that recycling a significant part of the silicon waste into a semi-closed loop was possible and suitable. Conservatively assuming that 30% of the feedstock ends up as kerf (worldwide roughly half a million tons per year as of today), it is a golden opportunity for the industry to show social responsibility and reduce this way its carbon footprint by a significant factor. Nevertheless, it did not stop the owner of the company from stopping all these efforts. The new owner shut down the plant in 2023–24, putting an end to the long-lasting development of UMG in Norway. To our best knowledge, no one on our list of 2014 is currently producing solar-grade silicon via the UMG route, and no newcomer has made any attempts to enter this market segment since 2014.

It is symptomatic that the only article in the present edition dealing with kerf aims at serving a totally different business application, that is, anodes for lithium-ion batteries. The article makes a point of achieving a low carbon footprint by using recycled silicon from the solar industry. It must be noted that the silicon market for battery anodes is still at the exploratory stage, and until it really takes off, millions of tons of valuable solar-grade silicon continue to be wasted by the solar industry.

The development of UMG between 1975 and 2020 has taken place in Europe, the United States, and Japan, three regions that have fostered the PV pioneers. China showed interest in PVs after 2000 and in a very short time (5–10 years), it built up a world-class industry surpassing all its Western competitors in terms of momentum and effectiveness. To move into the market quickly, the Chinese enterprises adopted mature and straightforward technology that they could purchase from the West. Regarding the silicon feedstock, the new entrepreneurs made the choice of producing polysilicon by using the classical Siemens process, which then gave the best guarantees in terms of critical purity to make the cells. India is now making similar attempts to penetrate the PV market, adopting a strategy similar to that of the Chinese 20 years ago.

Recycling materials from silicon solar cell manufacturing is, however, important and will be increasingly necessary in the coming years. This includes the recycling of silicon, crucibles, cells/ wafers, metallic contacts, PV module glass, and PV module structural materials. Recently, a review of the status of silicon solar cells and PV market development was published (Di Sabatino et al., 2024), which also gives some perspectives on the use of machine learning in the PV process and solar cell manufacturing and recycling.

In general, it is recommended that the industry considers more use of UMG silicon where its economic and environmental benefits are clearly demonstrated. We fully recognize the merits and the need for other technologies, such as polysilicon via the Siemens and the fluidized bed reactors, as well as single-crystal ingots, for making diversified and market-tuned solar systems. However, we think that abandoning solutions like UMG, multicast, and kerf recycling to solar feedstock is counterproductive when these proposed solutions have proven outstanding performances on environmental impacts.

In conclusion, we call for a rethinking of solar silicon feedstock strategy considering a holistic approach and not only circumstantial interests.

Author contributions

MD: conceptualization, methodology, validation, writing–original draft, and writing–review and editing. BC: conceptualization, methodology, validation, writing–original draft, and writing–review and editing.

Funding

Conflict of interest

Author BC was employed by MARCHE AS.

The remaining author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Generative AI statement

The author(s) declare that no Generative AI was used in the creation of this manuscript.

Publisher's note

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