Global Flow Analysis of Crystalline Silicon

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1. Introduction

Silicon is a unique material. Next to oxygen, it is the second most abundant element in the Earth’s crust. Its abundance is one of the reasons it is used for a wide range of purposes. One of the most important uses of silicon is as a core element of microchips. To manufacture microchips, the microelectronics industry requires silicon with an impurity level of $10^{-11}$. Since silicon forms a stable compound with oxygen (silicon oxide, SiO₂), the deoxidization of silicon oxide needed to reach this high level of purity consumes a substantial amount of energy, which, in turn, affects the environment through emissions of carbon dioxide (CO₂).

In the past decade, there has been a dramatic increase in the global supply and demand of crystalline silicon. This is because of a drastic increase in the demand for crystalline silicon for photovoltaic (PV) cells. While a core element for renewable energy, the purification process of crystalline silicon is energy-intensive. Therefore, the sustainability of crystalline silicon feedstock is an interesting area for research. The effective use of crystalline silicon could contribute to the sustainability of global environmental systems.

Reflecting its importance to sustainability, there has been a growing literature on material flows of crystalline silicon. Some studies highlight energy use and environmental impacts in the process of crystalline silicon production. For instance, Williams conducted a quantitative systems analysis of global production chains for silicon (Williams, 2003). He estimated global material flows of silicon in 1998 and then forecast how these flows would project out to 2020. In analyzing material flows, he chose different physical units such as weight and area, according to the characteristics of different outputs. Using this approach, Williams et al. were able to determine the energy required for a microchip based on the calculation on energy use at each production stage (Williams et al., 2002). Their analysis further was able to show in a quantitative manner that the purification to electronic grade silicon (EG-Si) is an energy intensive process.

Other studies have focused on the supply of crystalline silicon for solar cells. Woditsch and Koch expressed concern about the shortage of crystalline silicon for solar cells which were dependent on off-grade silicon from the EG-Si production process (Woditsch & Koch, 2002). They concluded that new routes for solar-grade silicon production are urgently needed. Sarti and Einhaus proposed short- and long-term solutions to respond to the demand for polycrystalline silicon (pc-Si, also called multicrystalline silicon) for solar cells. Their recommendations included a reduction in the amount of crystalline silicon in the short-term...
and the establishment of solar-grade silicon production from metallurgical-grade silicon (MG-Si) (Sarti & Einhaus, 2002). As for major metals such as iron, copper and nickel, the Yale Stocks and Flows project conducted a material flow analysis of substances on national, regional and global scales (for example, Graedel et al., 2004). Their approach focused on illustrating anthropogenic metal cycles through four processes: production, fabrication and manufacturing, use and waste management. They then offered useful suggestions on the benefits and challenges of a material flow analysis.

In a previous paper, Takiguchi and Morita presented a material flow analysis of silicon in Japan from 1996 to 2006 (Takiguchi & Morita, 2009). The analysis tracked the input and output of silicon in a series of purification process in units of weight and found that rapid growth in demand for pc-Si and single crystalline silicon (sc-Si, also called monocrystalline silicon) changed the structure of the crystalline silicon supply. Takiguchi and Morita also developed the Resource Effective-use Index (REI) to demonstrate how effectively crystalline silicon is used. The analysis of the REI indicated that the effective use of pc-Si had reached its outer limits. At the same time, the paper found a domestic analysis was limited in what it could demonstrate because crystalline silicon is traded globally.

This chapter analyzes and discusses the global flow of crystalline silicon to assess the sustainability of silicon feedstock. The chapter begins by reviewing how crystalline silicon is produced as well as recent trends in crystalline silicon supply. The next section provides a material flow analysis of silicon on a global scale, focusing mainly on crystalline silicon for solar cells. The “results and discussion” section describes the results of the material flow analysis, followed by a discussion of the sustainability of silicon feedstock.

2. Crystalline silicon and solar cells

Before conducting a material flow analysis, this section observes the recent trend of solar cell production and explains how crystalline silicon is produced, followed by sources of crystalline silicon for solar cells.

2.1 Expansion of solar cell production

Nowadays, many countries have promoted the use of renewable energy to reduce carbon dioxide (CO₂) for climate change mitigation and diversify energy sources. In particular, the installation of PV systems is expanding in many parts of the world. Under these circumstances, the production of solar cells, core elements of PV systems, is increasing rapidly. Fig. 1 illustrates trends in solar cells production by types. The total production of solar cells in the world sharply increased from 126 MW in 1997 to 10,660 MW in 2009. The share of sc- and pc-Si solar cells grew the most at 32% and 45%, respectively. This can be attributed to the ease of mass production using highly developed silicon technology, a good balance between energy conversion efficiency and cost, and the fact that their products are non-toxic. These advantages suggest that crystalline silicon (pc- and sc- Si) solar cells will be likely to continue to expand in the future.

2.2 Process for crystalline silicon production

A typical production process for crystalline silicon for semiconductors is illustrated in Fig. 2. In the first step, the reduction of silica (quartz, SiO₂) produces metallurgical-grade silicon (MG-Si) with a purity of approximately 98% in electronic furnaces.
The second step is purification from MG-Si to pc-Si. Usually, a chemical gaseous purification technique known as the Siemens method is used for purification. This method involves reacting MG-Si with hydrochloric acid (HCl) to convert purified trichlorosilane (SiHCl$_3$) by distillation (Si + 3HCl $\rightarrow$ SiHCl$_3$ + H$_2$). The trichlorosilane is then decomposed with hydrogen on pure silicon surfaces and converted back into pc-Si (SiHCl$_3$ + H$_2$ $\rightarrow$ Si + 3HCl). After the reaction, pc-Si with an impurity level of $10^{-11}$ is obtained. In this process, the formation of silicon tetrachloride (SiCl$_4$) as a by-product of the production of trichlorosilane lowers the yield of pc-Si.

Fig. 2. Typical process flow for purified Si production.
The third step is to convert pc-Si to sc-Si. For the conversion process, the semiconductor industry usually employs the Czochralski (CZ) method. In the CZ method, sc-Si grows on a seed crystal drawn up from molten pc-Si in a crucible, producing a sc-Si ingot. The cylindrical ingot of sc-Si is sliced into wafers followed by a series of processes such as wrapping and etching. Finally, the wafers are processed into integrated circuit (IC) chips. Table 1 provides the energy consumption at each stage from quartz to wafers (Frankl et al., 2004). It is obvious that the process of producing pc-Si from MG-Si consumes the most energy of all of the processes. While it is the established and most frequently used method of producing pc-Si, the production method presents several issues that need to be resolved, such as the limited velocity of the chemical reaction, the considerable initial investment required to enlarge the process, and the high cost in proportion to high consumption of energy.

### Table 1. Energy required for Si purification.

| Product                  | Electrical energy input/ kg silicon out (kWh/kg) | Silicon yield |
|--------------------------|-------------------------------------------------|---------------|
| Quartz → MG-Si           | 11                                              | 0.79          |
| MG-Si → pc-Si            | 149.9                                           | 0.76          |
| pc-Si → sc-Si ingot (Czochralski) | 31.1                                             | 1             |
| sc-Si ingot → wafer      | 42.5                                            | 0.37          |

2.3 Crystalline silicon for solar cells

Crystalline silicon for solar cells does not require a purity level as high as the level required for semiconductors. While the impurity level required for EG-Si is $10^{-11}$, the level for pc-Si solar cells is $10^{-7}$ to $10^{-6}$. Therefore, off-grade silicon produced as a by-product of the EG-Si production process has been used for solar cells. The following describes the process typically used to generate off-grade silicon. In the process of crushing pc-Si, approximately 5-10% of the produced pc-Si is lost; this is used as off-grade silicon. In the CZ process of producing sc-Si, nearly 40% of the pc-Si is lost as pot scrap, tops and tails, kerf loss and test wafers (Fig. 3) and used as off-grade silicon (New Energy and Industrial Technology Development Organization [NEDO], 2001). Scrap wafers are also used as off-grade silicon. The off-grade silicon is melted and turned into a pc-Si ingot via castings in a crucible mold or a sc-Si ingot via another CZ process. Polycrystalline silicon or sc-Si produced from the off-grade silicon ingot is sliced into wafers with a wire saw and used for solar cells. Depending on its purity level, off-grade silicon is used for other low-grade purposes such as making aluminium alloy.

In the 1990s, off-grade silicon obtained from the EG-Si production process satisfied the demand for silicon for solar cells. The recent increase in demand for solar cells, however, resulted in production of crystalline silicon independently, not as scraps of EG-Si. Since the purification level of pc-Si for solar cells is lower than that for semiconductors, pc-Si for solar cells is produced by a simplified Siemens method which increases the speed of trichlorosilane decomposition. It should be noted that the simplified method still consumes considerable energy.

Thus, crystalline silicon for solar cells is currently obtained from two sources: 1) off-grade silicon produced as a by-product of the EG-Si production process and 2) silicon produced independently for solar cells.
3. Global flow analysis of crystalline silicon

This section presents a material flow analysis of crystalline silicon. After explaining the scope and methodology of the analysis, the material flows are shown and discussed.

3.1 Scope of material flow analysis

A material flow analysis tracks flows of materials at a particular scale in a quantitative manner. Possible scales include the global, regional, national, community, or factory scale. A material flow analysis for a specific material is called a “substance flow analysis.” This chapter analyzes the material flow of crystalline silicon, clarifying the input and output of the material at each phase of the production process.

In undertaking a material flow analysis, the scope of the material flow should be clarified. This chapter analyzes the material flow of silicon on a global scale from 1997 to 2009. The material flow also focuses on the stages of production, fabrication and manufacture (Fig.4), because it aims to demonstrate the sustainable supply of crystalline silicon. The global scale was chosen as the geographical boundary for the material flow analysis since the objective of this analysis is to understand the sustainability of flows. Nowadays, material flows at the national level would not be closed due to the export and import of the materials. A global material flow can offset export and import and hence capture the entire flows. On the other hand, a drawback of the global material flow is the quality of data. A global material flow requires global data. While some countries have robust data, others do not, which can influence the accuracy of the analysis. Therefore, these advantages and disadvantages should be considered carefully in analyzing global material flows.
The timeframe for the material flow analysis is 1997 to 2009. This twelve year period starts when relevant solar cell first became available and goes to the latest available data. Whereas a material flow in a single year is like a “snapshot,” a time series analysis of material flows over the period enables illustrates the changes of flows over time.

In the material flow analysis in this chapter, silicon used for making thin-film and amorphous types are not considered, because silicon used for these types seems to be less than 1% of that used for making the crystalline types. This point is discussed in greater detail in the “results and discussion” section.

3.2 Methodology
To determine the material flow of silicon on a global scale, data used for the analysis and the respective sources are listed in the Table 2. The assumptions made in developing the material flow and methodologies to estimate values are based mainly on Takiguchi and Morita (Takiguchi & Morita, 2009). Most of the data for the material flows are gathered from the Japanese journal Industrial Rare Metal, which reviews annual trends in industrial materials (Industrial Rare Metal, 1998-2010).
Table 2. Data sources and calculation methods.

### 3.2.1 Metallurgical-grade silicon

MG-Si is supplied mainly by China, Norway, and Brazil with China increasingly recognized as the dominant supplier. MG-Si is used for various purposes: production of crystalline silicon; deoxidization of steel; and production of aluminium alloy and silicon resin etc. Data on the supply of MG-Si on a global scale are available (Industrial Rare Metal, 1997-2010). It is assumed that the supply of MG-Si is equal to the figures in that data. While data regarding the amount of MG-Si used for crystalline silicon were not available, it is estimated that nearly 1.3 kg MG-Si is used to produce 1 kg pc-Si.

### 3.2.2 Polycrystalline silicon

Polycrystalline silicon as the primary products is divided into electronic-grade pc-Si (EG pc-Si, \( P_{pc,e} \)) and pc-Si produced independently for solar cells (\( P_{pc,s} \)). Data on the production of \( P_{pc,e} \) and \( P_{pc,s} \) are available (Industrial Rare Metal, 1998-2010). In 2009, 92,100 tons of pc-Si was produced, the ratio of \( P_{pc,s} \) to \( P_{pc,e} \) being approximately 3:1. The data are the sum of production by manufacturers located mainly in the United States, Japan and Germany. Off-grade pc-Si (\( O_{pc} \)) is generated in the pc-Si production process and the CZ process.

The total demand for pc-Si for solar cells (\( D_{pc,s} \)) is satisfied by two sources: off-grade pc-Si for solar cells (\( O_{pc,s} \)) and pc-Si produced independently for solar cells (\( P_{pc,s} \)). Based on the assumption that there is no loss in casting, this supply and demand relationship is expressed as:

\[
P_{pc,s} + O_{pc,s} = D_{pc,s}
\]  

(1)

Because pc-Si (\( P_{pc,s} \)) has been produced independently for solar cells since around 2000, the chapter assumes that \( P_{pc,s} \) was zero before the year 2000. Demand for pc-Si for solar cells (\( D_{pc,s} \)) has been calculated by multiplying the global production of pc-Si solar cells (in Watts) by the amount of pc-Si used for the production of 1 W. Since the amount of pc-Si consumed per Watt steadily decreased from 20 g in 1995 to 15 g in 2000 to 10 g in 2005 (Industrial Rare
Metal), it is estimated from this linear relationship from 1997-2005. After 2006, it is estimated at 9 g per Watt.

It should be noted that there is a loss of crystalline silicon in the wafer saw process. Given 15% for the cell efficiency and 180 micrometers for the wafer thickness, 1 W of the solar cell includes 2.8 g of pc-Si inside. The difference between the demand of pc-Si for solar cells (9 g/W in 2009) and 2.8 g is the loss.

Although the amount of off-grade pc-Si for solar cells is not given, it can be estimated by equation (1). Data for the average price of EG pc-Si per ton each year was available (Industrial Rare Metal, 1998-2010).

### 3.2.3 Single crystalline silicon

Single crystalline silicon as the primary product is divided into EG sc-Si (P_{sc,e}) and sc-Si produced independently for solar cells (P_{sc,s}). While data on these variants of silicon are not available, EG sc-Si can be estimated by multiplying the amount of EG pc-Si (P_{pc,e}) by the yield rate of EG sc-Si. In this chapter, the rate was set at 0.7. Single crystalline silicon is produced independently for solar cells, partly using pc-Si for solar cells. Off-grade sc-Si (O_{sc}) is generated in the wafer production process or as wafer waste and presumably used for solar cells. Data on the off-grade sc-Si are also not available. As well as pc-Si for solar cells, off-grade sc-Si was used as a proxy for the demand for sc-Si for solar cells until around the year 2000.

The same approach to estimating the demand of pc-Si for solar cells (D_{pc,s}) is applied to estimating demand of sc-Si for solar cells (D_{sc,s}), assuming that consumption of sc-Si per Watt is identical to that of pc-Si for solar cells. The amount of sc-Si per Watt for the combined type of sc-Si and amorphous silicon in heterostructures was assumed to be 60% of other sc-Si cells.

### 3.2.4 Wafers

In Fig. 4, “wafers” refers to those used for semiconductors not solar applications. Data regarding the global shipments of wafers for semiconductors are available in units of area (Semiconductor Equipment and Materials International [SEMI], 2011) and are assumed to be equal to production of the wafers (P_w). The calculation of weight of the wafers requires currently unavailable thickness data. Therefore, composition of wafers in diameter has been assumed to be identical to the case in Japan (Ministry of Economy, Trade and Industry, Japan, 1997–2010). Using this assumption, the composition of wafers in 2009 is 4.2% for a wafer under 5 inches (in.), 10.4% for 6 in., 22.6% for 8 in., and 62.7% for 12 in. The weight of products, in turn, has been calculated based on the assumptions that the density of a wafer is equal to that of silicon (2,330 kg/m^3) and that the thickness is 0.625 mm for a wafer under 5 in., 0.675 mm for 6 in., 0.725 mm for 8 in., and 0.775 mm for 12 in.

### 3.2.5 Resource effective-use index

As in Takiguchi and Morita, the chapter uses the resource effective-use index (REI), which is the ratio of resource input to output required for a given product (Takiguchi & Morita, 2009). The REI enables one to measure quantitatively the extent to which resources are used effectively. The trend in the REI values, therefore, explains how effectively the materials in
question have been used over time. The increase of the REI value means that the resource in question is being used more effectively.

In this chapter, the input is the sum of production of electronic-grade pc-Si (P_{pc,e}) and pc-Si produced independently for solar cells (P_{pc,s}), while the output is the sum of demand of pc-Si and sc-Si for solar cells (D_{pc,s} and D_{sc,s}) and production of wafers (P_{w}). The REI can be defined as follows:

\[
REI = \frac{P_{w} + D_{pc,s} + D_{sc,s}}{P_{pc,e} + P_{pc,s}}
\]  

(2)

To be exact, reuse of wafers as off-grade silicon should be added into the inputs in the calculation of the REI. However, data on the amount of reuse are not available, and therefore they are excluded.

### 3.3 Results and discussion

In the period of interest, the amount of crystalline silicon supply has expanded. Fig. 5 illustrates that the growing demand of crystalline silicon for solar cells brought about the increase in pc-Si supplies. The supply increased from 16,050 tons in 1997 to 92,100 tons in 2009. The production level of wafers decreased in 2009 from the previous year, probably due to the financial crisis and global economic slump. Nevertheless, it merits attention that the supply of pc-Si did not decline despite of downward trends in the global economy.

![Fig. 5. Production of pc-silicon and wafers.](data:image/png;base64,iVBORw0KGgoAAAANSUhEUgAAAAIAAAAgCAMAAAAD303vAAAACXBIWXMAAAsTAA

As Fig. 6 clearly shows, the growing demand for crystalline silicon boosted its price. The price of pc-Si increased after 2004 and approached nearly 10,000 Japanese Yen (JPY) per kg (approximately, 100 US dollars, using the exchange rate: 1 US$ = 100 JPY). In 2008 and 2009, the trend remained stable mainly because of sufficient pc-Si supply.

The main objective of this chapter is to track the material flow of silicon on a global scale. Figs. 7 and 8 show the global material flows of silicon in 1997 and 2009 respectively.
Comparing the two figures, there is a remarkable increase in the amount of silicon at each stage. The 1997 material flow is relatively simple, because demand for crystalline silicon for solar cells was covered by the off-grade silicon from the EG-Si production process. In 2009, however, pc-Si produced independently for solar cells was much larger than the off-grade silicon.

![Fig. 6. Price of pc-silicon.](image)

While the global material flow expanded over the period of interest, the question is how effectively crystalline silicon had been used. An analysis of trends in the REI can help answer this question. Fig. 9 describes the trends in the REI. From 2001 to 2008, the values of REI consistently increased. This trend implies progress in the effective use of crystalline silicon. In 2009, however, the REI fell to 0.92 from 1.14 in 2008, partly because the global economic downturn created some slack between supply and demand.

Effective use of crystalline silicon was probably achieved by improvements in the yield rate at each stage, reductions in wafer thickness and kerf loss, and enhanced use of off-grade silicon. Off-grade silicon, which was used as a cheap additive to aluminium alloy in the past, is now used for more valuable products—i.e. solar cells.

It is interesting to note that the REI value exceeded 1.0 in 2008. This implies the mass balance was not achieved in that year, partly because of the reuse of test wafers, changes in stocks, and inaccuracy of data. Nevertheless, the upward trend in the REI values is apparent.

Fig. 10 plots the REI values as a function of the price over the period of interest. The REI values seem to rise in response to the pc-Si’s increasing price, because the increasing value leads to the more efficient use of materials. This applies to the second half of the analyzed period. In the first half of the period, the REI values increased despite price fluctuations. This is not surprising, given the fact that advanced technologies for effective use of pc-Si would be used regardless of price fluctuations once they were built. According to Tilton, the supply of scraps generated in the course of producing new goods is unresponsive to changes in the market price because of the ease of its collection, high quality, and low recycling cost (Tilton, 1999).
Fig. 7. Global silicon material flow (1997).
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Fig. 8. Global silicon material flow (2009).
Although it is important to pay attention to trends in 2010 and after, concerns over the shortage of crystalline silicon for solar cells are rarely raised recently, due to an expansion in the supply. Progress in the effective use of crystalline silicon has been demonstrated by a material flow analysis of silicon on a global scale. However, pc-Si for solar cells is produced independently by conventional energy-intensive methods. Taking into consideration the continuous expansion of solar cells, a sustainable supply of crystalline silicon should be achieved by low-energy and low-cost methods.
In Takiguchi and Morita, four solutions are proposed to ensure a sustainable supply of silicon feedstock (Takiguchi & Morita, 2009): (1) production of solar-grade pc-Si by a less costly and less energy-intensive method, (2) reduction of the amount of pc-Si per Watt in solar cells, (3) acceleration of the development and deployment of other PV types, and (4) reuse and recycling of solar cells in the future. With the exception of the third recommendation which is predicated on diversifying the materials used for solar cells into non-silicon, the other three suggestions are applicable to global supply of crystalline silicon. Less costly and less energy-consuming silicon refining processes for solar cells are currently being developed, including a process that develops the refining solidification of silicon using the Si-Al solvent under low temperatures (Morita & Yoshikawa, 2007). Furthermore, in Japan, the JFE steel company produces solar-grade silicon directly from MG-Si using a pyrometallurgical process at a production scale of 400 tons per year (Yuge et al. 2001).

There have been achievements thus far in reducing the amount of crystalline silicon per Watt in solar cells. More significant reductions of silicon could be realized by new types of silicon solar cells. For example, thin-film silicon has been introduced for solar cells, typified by tandem-type silicon composed of stacked amorphous silicon and microcrystalline silicon, also known as nanocrystalline silicon. Tandem-type silicon with a thickness layer less than one hundredth that of bulk types can contribute to meaningful reductions of silicon used for solar cells. In this regard, the material flow in unit of weight may not be the best indicator of resource efficiency since small but important flows, such as development of thin-film silicon, are likely to be neglected. In analyzing the material flow, therefore, attention should be paid to important trends behind the flow.

The reuse and recycling of solar cells will gain in significance in the near future. “Reuse” implies the second use of end-of-use PV modules, while “recycling” refers to use of the material recovered from decomposed PV modules. Needless to say, the reuse of PV modules would reduce energy consumption and CO₂ emissions, compared to newly produced modules. With regard to recycling end-of-use PV modules, a quantitative analysis showed that the recycling can reduce energy and CO₂ emissions when inputting recovered silicon into the process after purification (Takiguchi & Morita, 2010). According to the NEDO report, modules of crystalline silicon did not show any deterioration in performance even after being in use for more than 15 years (NEDO, 2006). In the reuse and recycling of PV modules, a robust system to collect end-of-use modules will be a key to success, because unintentional incorporation of impurities into the reuse and recycling process will make reuse more difficult. Recycling is not limited to PV modules. As described in 3.2.2, there is loss of crystalline silicon in the wafer saw process. Dong et al. conducted a beneficial and technological analysis for solar grade silicon wastes demonstrating it is feasible to recycle silicon ingot top-cut scraps and sawing slurry wastes (Dong et al., 2011).

Overall, the material flow analysis on a global scale was found to be a useful approach to examine the sustainability of crystalline silicon supply. As described in the sub-section of methodology, uncertainty of the data on a global scale is a drawback to the analysis. Nevertheless, global flow analyses are meaningful to overview a worldwide picture.

## 4. Conclusions

This chapter discussed the sustainability of crystalline silicon supply. The discussion focused on the material flow analysis of silicon on a global scale. The results showed
significant changes in crystalline silicon supply due to growing demand for solar cells. The global supply chains not only expanded but became more complicated. While the analysis of the REI values showed progress in the effective use of crystalline silicon, pc-Si for solar cells is being produced through an energy-intensive method. To ensure a sustainable supply of silicon feedstock, three recommendations were made: 1) solar-grade pc-Si should be produced through a less costly and less energy-intensive method; 2) the amount of pc-Si per Watt in solar cells should be reduced; and 3) solar cells should be reused and recycled. The demand for solar cells is still strong. Crystalline silicon supply in the future will be integral to the sustainability of global environmental systems.

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The exciting world of crystalline silicon is the source of the spectacular advancement of discrete electronic devices and solar cells. The exploitation of ever changing properties of crystalline silicon with dimensional transformation may indicate more innovative silicon based technologies in near future. For example, the discovery of nanocrystalline silicon has largely overcome the obstacles of using silicon as optoelectronic material. The further research and development is necessary to find out the treasures hidden within this material. The book presents different forms of silicon material, their preparation and properties. The modern techniques to study the surface and interface defect states, dislocations, and so on, in different crystalline forms have been highlighted in this book. This book presents basic and applied aspects of different crystalline forms of silicon in wide range of information from materials to devices.

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