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LIFE CYCLE ASSESSMENT OF AN INGOT MANUFACTURING PROCESS USING SILICON POWDERS FROM RECYCLING

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Abstract: Within the Horizon 2020 innovation action “Implementation of a circular economy based on recycled, reused and recovered indium, silicon and silver materials for photovoltaic and other applications” with the acronym “CABRISS” opportunities for innovative photovoltaic (PV) materials are assessed. In this paper an innovative process for the production of silicon (Si) wafer equivalent and highly doped wafers from Si powders coming from the PV modules recycling process or from kerf of the diamond wire sawing process is presented. The core motivation is the reuse of Si waste as pure Si powder (purity of 99.5 – 99.9% and with tailored particle size distribution) as a basis for the production of new Si-based PV wafer material.

The results of the investigation show promising values for life cycle costing (LCC) and life cycle analysis (LCA) attributes if the production is up scaled to an industrial size. The current key figures for one 156 x156 mm^2 standard wafer sawed from a sintered ingot are 0.4 kWh_{el} cumulated final energy and costs of 0.48 €. Assuming a cell efficiency of 15 % and Q_G=1400 kWh/(m^2*a) the contribution of these innovative wafers to the total energy payback time (EPBT) of PV systems would be 0,08 years what is a very promising result. Therefore sintered wafers could compete with conventional ones, if the following refining processes to produce PV cells will not be too expensive from a LCC and LCA point of view.

Further research and development has to focus on an optimal production process and technology design, an optimization of the sintering process parameters and the probably necessary finishing of the wafer surface to get it ready for PV cell production.

Keywords: PV silicon recycling, hot pressing of silicon powders, innovative silicon processing, life cycle assessment of PV recycling, CABRISS

1 Description of the investigated PV recycling system

In CABRISS two major pathways are investigated: a) recycling of end of life crystalline Si PV systems and the production of new crystalline Si PV and b) recycling of end of life thin film PV systems and the production of new crystalline thin film PV. In view of the specific topic of the present paper a flow chart of the recycling system for crystalline Si PV is shown in **Figure 1** below.

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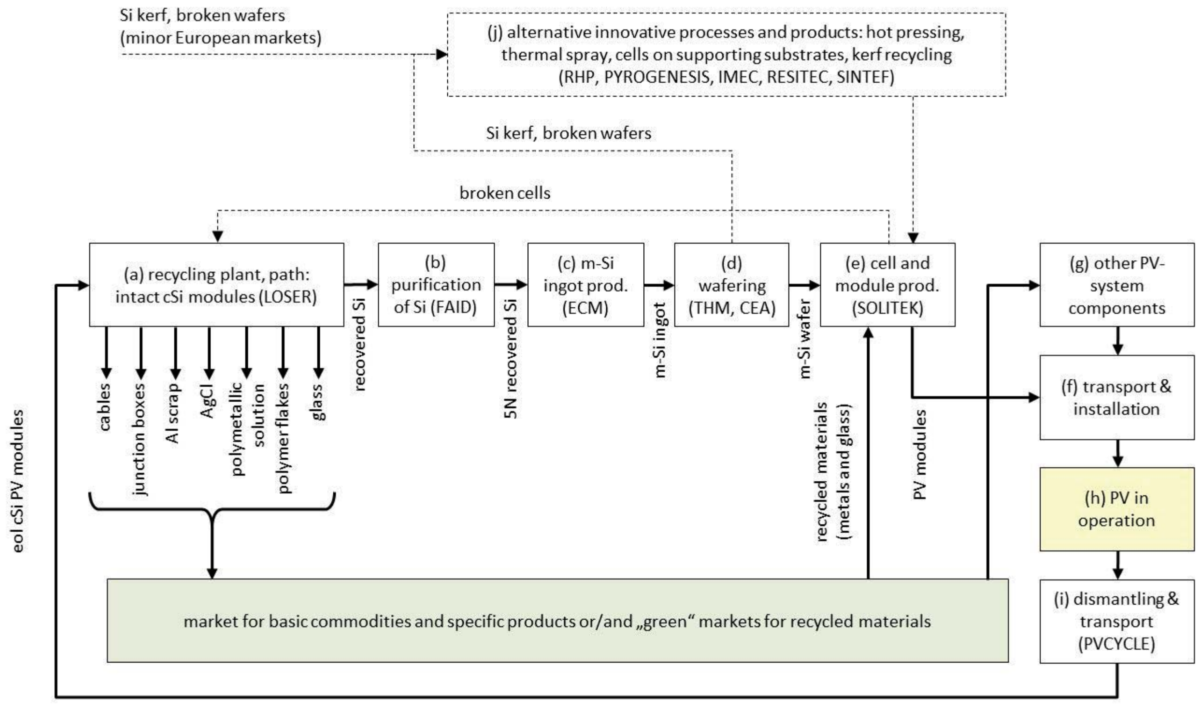

Figure 1: flow chart of the PV recycling system for crystalline Si PV systems. Source: research project CABRISS. Involved organizations: RHP (AT), PYROGENESIS (GR), IMEC (BE), RESITEC (NO), SINTEF (NO), LOSER (DE), FAID (ES), THM (DE), CEA (FR), SOLITEK (LT), PVCYCLE (FR) and TU-Vienna (AT), responsible for LCA and LCC aspects of the recycling and innovation system.

The investigated system is a closed loop (cradle to cradle) and starts with the dismantling and transportation of end of life PV systems to the recycling plant, see box (i) in **Figure 1**. Because of distinct economic scale effects and low transportation costs a few recycling plants will probably manage the European PV recycling market. The optimal locations for these plants are influenced by the geographic distribution of the present and future stock of PV systems and the vicinity of downstream industry like glass industry.

The output of the recycling plant mainly goes into the market for basic commodities like copper, aluminium, other valuable metals, polymers and glass. If it is financially attractive or the (future) recycling plant covers additional links of the value chain, the recovered Si can be purified and put into the production of new crystalline Si PV cells and modules. The technical results of the research project CABRISS show that the recycling system for crystalline PV systems is technically feasible.

Beside this main recycling path there are several innovative approaches which investigate specific questions for the exploitation of side products of the silicon and PV industry. One of these topics is the question how to use silicon from wafer and cell breakage and kerf from the sawing process which is the major material loss in the value chain up to now. From the LCC and LCA point of view it is best to use these material losses in an unmodified shape. Especially the Si powders from the sawing processes but also milled Si residuals from other process steps along the value chain are predestinated for sinter processes.

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2 Wafer production via hot pressing process

The investigations of the usability of kerf focus on kerf from cutting of silicon ingots, silicon blocks and silicon wafers from solar grade silicon. The sawing technology was a water based system with diamond wire cutting and wafering. Traditional cutting methods using glycol and silicon carbide as an abrasive grain were not analyzed. Two different qualities of kerf are described in the following by their chemical composition and particle size distribution see **Figure 2** and **Figure 3**.

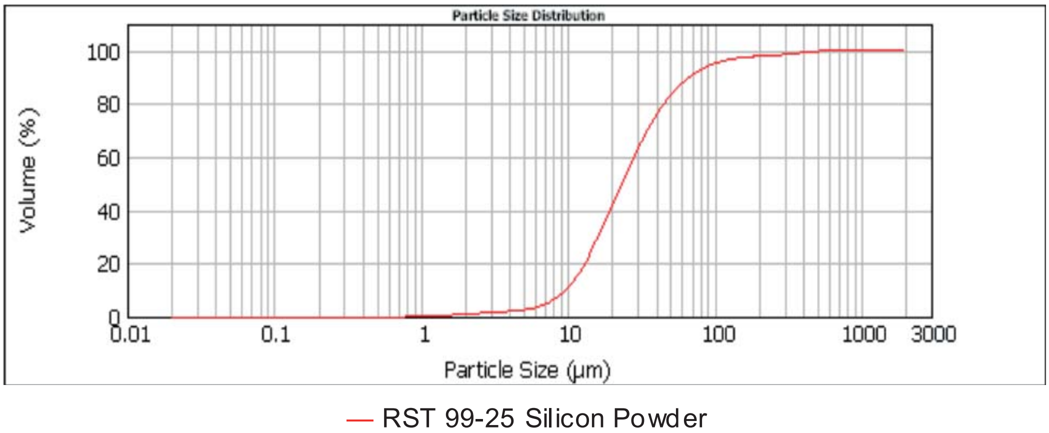

Figure 2: Characterization of sample 1: Si99-25: Si: >99,2%, Fe: ≤0,06%, Al: ≤0,7%, Ca: ≤0,02%, Cr, Cl, Cu, Ti: ≤0,001%; PSD: D10: 10µm, D50: 25-30µm, D90: 100-150µm;

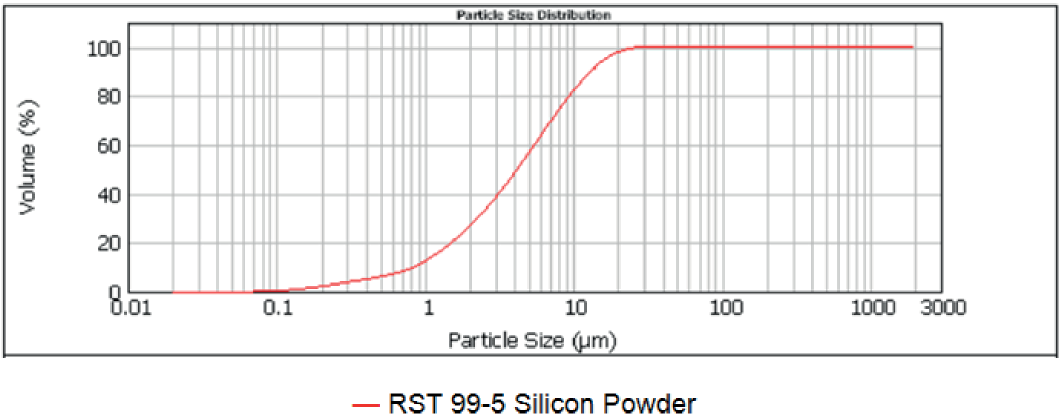

Figure 3: Characterization of sample 2: Si99-50: Si: >99,5%, Fe: ≤0,05%, Al: ≤0,15%, Ca: ≤0,02%, Cr, Cl, Cu, Ti: ≤0,001%; PSD: D10: 1µm, D50: 5µm, D90: 15µm;

The input material of sample 1 and 2 was fed in the sintering process which is shown in a flow chart in **Figure 4** respectively in images in **Figure 5 to 12**. After doping the Si powder with Boron with a mixing process the doped powder is filled into a pressing mold and it is pre-compressed in the cold state. The hot pressing process follows at a temperature of 1350 °C during approx. 4 hours whereby the result depends on the adjustment of several process parameters. The most important are:

- Temperature and temperature regime over the process duration
- Pressure and pressure regime over the process duration

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- Dimensions and technical design of the graphite pressing mold
- Particle size distribution in the doped Si powder
- Chemical impurities of the doped Si powder
- Process atmosphere (argon and hydrogen)

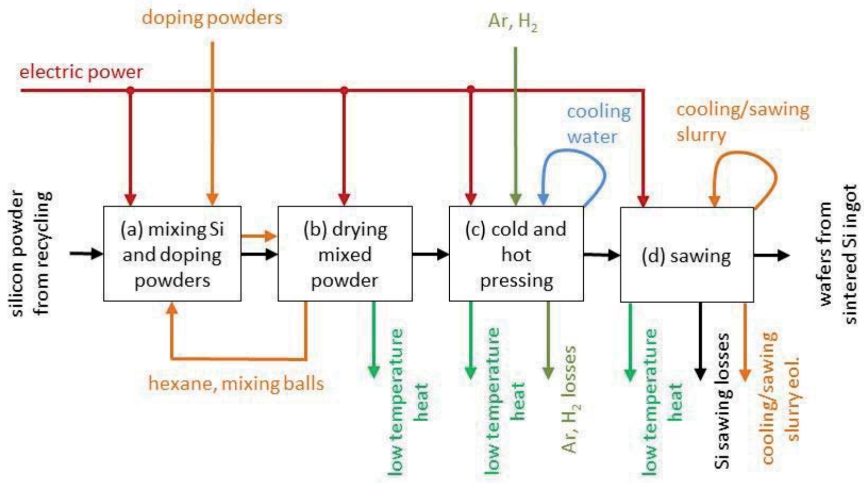

Figure 4: flow chart of the investigated Si powder sintering process including wafering via diamond wire sawing process.

The material density of the probes described above reached 97.1 % in case of the finer powder (sample 2) and 92.0 % in case of sample 1. The diamond wire sawing process was successful in both cases but inhomogeneous material structures resulting from partially melting regions are a big problem for the sawing process. A major challenge is to avoid the reaction between the pressing mold made from graphite and the very reactive silicon during the hot pressing process. In addition it was decided to manufacture the ingots not with sharp edges due to mold cracking but with a pseudo-square shape similar to monocrystalline wafers.

It was also tested to produce wafers via hot pressing process directly so that the sawing process can be omitted. It was possible to produce square shaped wafers of 156 x 156 mm^2 and a thickness of 1.5 mm and circular shaped wafers with a diameter of 65 mm and a thickness of 0.5 mm. But the quality of the surface was low and would need further process steps of grinding and polishing to get it ready for further refinement.

Reasons for the damage of pressing molds are mostly melting processes where the silicon gets in contact with the graphite mold. The high reactivity of the silicon causes a diffusion process and the ingot weld with the mold which is damaged beyond repair. Silicon melting processes are facilitated by chemical impurities of the powder and inhomogeneity caused by the filling process of the mold which is carried out manually up to now.

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Figure 5: powder mixing machine

Figure 6: powder drying machine

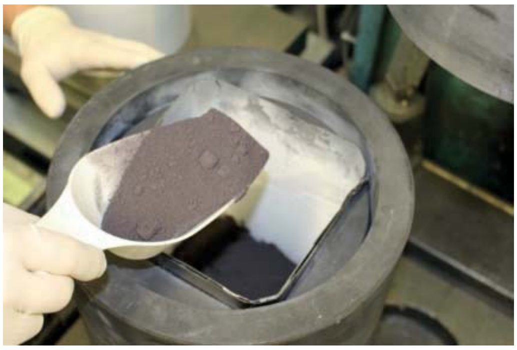

Figure 7: manually filling of Si powder into the graphite pressing mold

Figure 8: cold pre pressing of Si powder in the completed pressing mold

Figure 9: hot pressing machine in progress

Figure 10: hot pressing machine with completed pressing mold

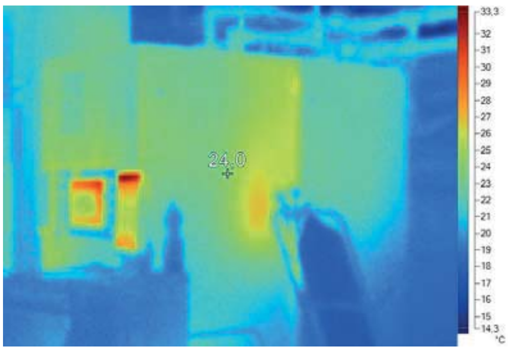

Figure 11: IR image of hot pressing machine surface at max. cycle temp. of 1350 °C

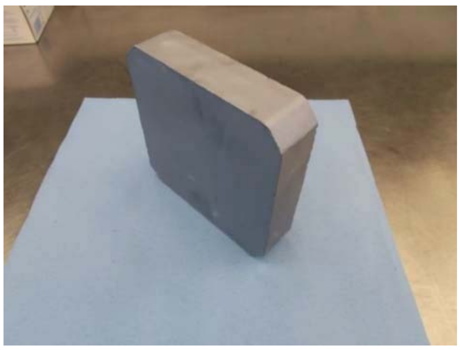

Figure 12: hot pressed Si ingot 156 mm x 156 mm x 40 mm (📷 E. Neubauer)

All photos, if not marked differently, by Peter Biermayr

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3 Industrial process design

At the moment, the entire process is done on laboratory scale. From an LCA and LCC point of view, the hot pressing process is most important and it is also the bottleneck in the present process chain. The duration of the hot pressing of a 2200 gram 156 mm x 156 mm x 40 mm cSi ingot produced in the related experiment was nearly 5 hours and caused an electricity consumption of 277,5 kWh.

The height of the ingot (40 mm up to 100 mm) has only a marginal impact on process duration and energy consumption. The major problem of the investigated process is the combined heating, pressing and cooling down in one and the same machine. The duration of the cycle could be decreased significantly by splitting the process in 3 different machine components: i) pre-heating to the process temperature; ii) hot pressing process; iii) cooling down.

In addition to the much lower cycle time, heat losses are also much lower and of higher exergy. That should allow the usage of remaining heat losses in the process (e.g. high temperature cooling unit heat goes to pre-heating unit) and in the surrounding area (e.g. drying processes, space and water heating). A draft of an industrial scale ingot production line with hot pressing process is shown in **Figure 13**.

The second promising efficiency potential is the design of the pressing mold. Currently, one ingot with the maximum measures 156 mm x 156 mm x 100 mm can be produced in a circular graphite pressing mold per cycle. But the dimensions of the hot pressing machine also allow the usage of a more complex pressing mold for 4 ingots of the same size which can be hot pressed at the same time. The biggest challenge is to handle the radial forces in the square mold. It could be solved by the usage of a stiff metal outside case which is able to withstand the high process temperature. In any case this is a topic for further research and development.

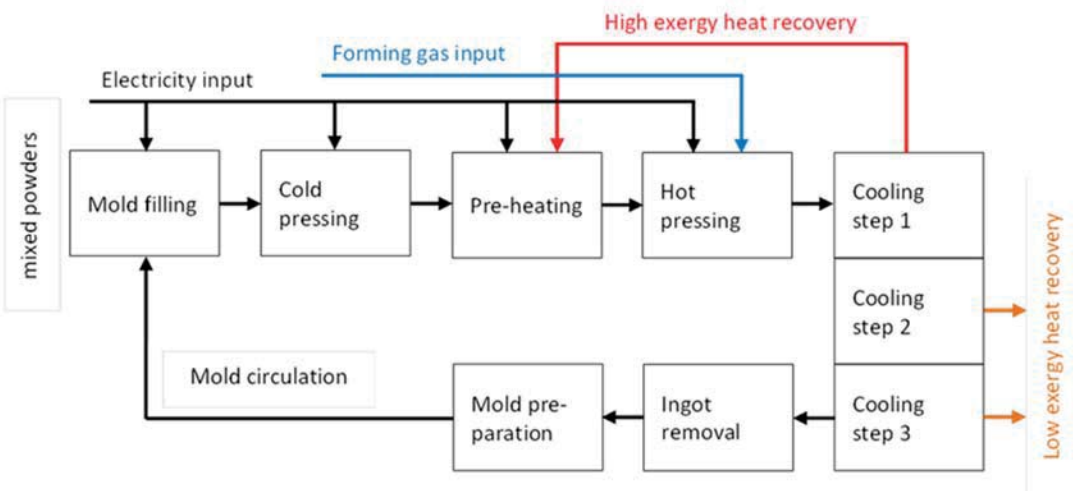

Figure 13: Draft of an industrial scale ingot production line with hot pressing process.

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4 LCC and LCA aspects

The total wafer costs and the cost structure can be seen in **Table 1**. Lab scale production costs amount to 1.48 € per wafer and in case of industrial scale costs are 0.48 € per wafer.

The cost structures of the two production scenarios are significantly different, see **Figure 14** and **Table 2**. Especially investment costs and labour costs are much lower in case of industrial production. A sensitivity analysis shows that the specific costs of the Si/B powder mix have the highest impact on the total wafer price in case of industrial scale wafer production. A comparison with the current cSi wafer spot market price (0.62 €/wafer) shows that the lab scale hot pressing process will not be competitive even if the input Si powder costs nothing.

Table 1: Notation of LCC key figures for Si ingot and wafer production via hot pressing. If there is no other information, the figures refer to 1 kg cSi ingot (kg_i).

Description	lab scale (experiments)	industrial upscaled	unit
Process productivity			
Cycle time (absolute)	5	0.5	h
Cycle time (relative)	0.93	0.02	h/kg_i
Cycles per year	1,402	16,644	1/a
Maximum productivity in wafers per year	400,457	19,021,714	1/a
Energy prices			
Electricity price	0.12	0.06	$€/kWh_{el}$
Material prices			
Si powder (2nd grade powdered, estimation)	14.18	12.99	€/kg
B powder (-200 mesh)	400	400	€/kg
H_2 (gas in 50 liter / 200 bar bottles)	5.20	5.20	$€/m^3$
Ar (gas in 50 liter / 200 bar bottles, purity 99.996 %)	14.50	14.50	$€/m^3$
Investment costs			
Capital costs (adequate target rate = 5 %)	33,706	49,178	€/a
Depreciation period (in general)	5	5	a
Workforce			
Workforce per cycle (technician)	2.5	0,5	h
Hourly rate (absorbed cost)	35	35	€/h
Product costs			
Wafering	0.13	0.11	€/wafer
Investment	0.39	0.01	€/wafer
Material	0.41	0.32	€/wafer
Energy	0.17	0.03	€/wafer
Workforce	0,38	0.01	€/wafer
Total costs	**1.48**	**0.48**	**€/wafer**

The calculated industrial scale wafer production costs are lower by 0.14 €/wafer than the present spot market price for multicrystalline Si wafers. This difference between the production costs of wafers from the sinter process and the spot market prices can be seen as a potential profit of the innovative producer if it can be proven that additional measures for

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the preparation of the wafer surface are not necessary or very cheap. Therefore these cost values must not be seen as final values.

On the one hand the calculated wafer costs are based on relatively high Si powder costs and on the other hand the investigation shows a high sensitivity in this area. Thus further investigation should focus on using cheap Si powders and should investigate further areas of economies of scale in the industrial production design model. Silicon powders from eol PV recycling could be held in the recycling process if they fulfill exactly the minimum requirements for hot pressing or spark plasma processes (quality and price are too high for e.g. metallurgical use and too low for e.g. Cz-process).

Table 2: Cost summary for wafer production via hot pressing process.

Process	Scale	Total costs*	% of total costs for			
			invest, capital & deprec.	material input	energy input	labour
wafer production via hot pressing	lab	1.48 €/Wafer	26 %	28 %	11 %	26 %
	industry	0.48 €/Wafer	3 %	67 %	6 %	2 %
*including costs for wafering (% of total lab scale: 9 %; % of total industrial scale: 24 %)						

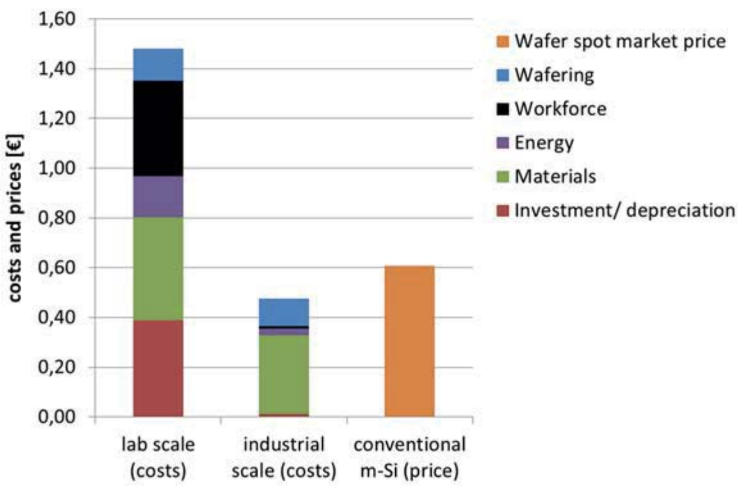

Figure 14: Comparison of wafer costs (lab scale and industrial scale) and spot market price of multi-Si wafers; source: TU-Vienna

The investigated LCA aspects focus on cumulated final energy consumption (to be independent from site location and local primary energy mix) and the contribution to the EPBT of PV systems produced from innovative components. The final energy consumption for pressing and slicing one kg ingot was 72.9 kWh_{el} in case of lab scale and 23.4 kWh_{el} in case of industrial scale process, see **Table 3**. Thus specific final energy consumption for the lab scale process is 3.1 times higher than for the optimized process. Under consideration of a technically feasible heat recovery in the industrial process (at the minimum useful heat > 600 °C can be recovered) the factor increases to 3.5. Therefore the production of one wafer

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needs 1.4 kWh_{el} in case of lab scale process and 0.4 kWh_{el} in case of industrial process. Assuming a cell efficiency of 15 % and Q_G=1400 kWh/(m^2*a) the contribution to the EPBT of this wafer material would be 0,08 years what is a very promising result.

Table 3: Selected empirical LCA input data for the production of wafers from sintered Si materials in case of lab scale and industrial scale process.

Description	laboratory	industry	unit
Material input			
Mixed powder input	1.05	1.01	kg/kg_i
forming gas (Ar, H2) input	0.28	0.05	m^3/kg_i
Ingot mass which can be produced per cycle	5.4	21.6	kg_i
Energy (=electricity) input			
Electricity consumption for powder mixing	0.7	0.2	kWh/kg_i
Electricity consumption for powder drying	0.6	0.3	kWh/kg_i
Electricity consumption for cold pressing	0.04	0.01	kWh/kg_i
Electricity consumption for hot pressing	54.1	15.4	kWh/kg_i
Electricity consumption for auxiliary drives	4.6	0.1	kWh/kg_i
Electricity consumption for wire sawing	12.9	7.3	kWh/kg_i
Total electricity consumption of the process	**72.9**	**23.4**	**kWh/kg_i**
Characterisations			
Wafer thickness	200	200	µm
Kerf loss	150	150	µm
Number of wafers per cycle	286	1143	1
Total wafer area per cycle	7.0	27.8	m^2
Total wafer area per year	9746	462912	m^2
wafer mass output	0.6	0.6	kg/kg_i
kerf loss mass	0.4	0.4	kg/kg_i
Heat recovery			
Useful heat > 600 °C	0	2.3	kWh/kg_i
Useful heat > 150 °C … 600 °C	0	5.8	kWh/kg_i
Useful heat > 60 °C … 150 °C	1.5	3.5	kWh/kg_i
Total useful heat (heat recovery)	1.5	11.7	kWh/kg_i
Electricity consumption per wafer	**1.4**	**0.4**	**kWh**

5 Conclusions

The innovative approach of processing Si wafers from Si kerf or PV recycling sources via hot pressing is technically feasible and it shows some promising results for LCC and LCA attributes. But it also shows some challenges for process engineering, material improvements and the need of an empirical proof of a PV cell production on the original or modified surface of the sintered wafer. The optimization of the sintering process is a multi-parameter optimization problem which requires further empirical research and development to make it insensitive for changing input material quality features like particle size distribution or chemical impurities what can be expected in case of the use of recycling materials.

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