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Projekt
Solare Herstellung von Kalk
(Solar Production of Lime)

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Zusammenfassung

Ein neuartiger, indirekt beheizter Solarreaktor zur Herstellung von solarem Kalk (CaO) wurde konstruiert, gebaut und im Solarofen des *PSI* getestet. Der kontinuierlich betriebene 10-kW Reaktor besteht aus einem geneigten Drehrohr mit einem speziell geformten, gut wärmeleitenden und hochtemperaturbeständigen Schwarzkörperabsorber aus SiC sowie einer Vorheizkammer, in welcher die feinkörnigen Kalksteinpartikel beinahe auf die Reaktionstemperatur vorgeheizt werden. Der Übergang zu einem indirekt beheizten Reaktor, verbunden mit zusätzlichen Konstruktionsänderungen, verlief erfolgreich: (1) Dank des verbesserten Fördersystems konnte die maximale Kalkproduktionsrate von 1.5 kg/h auf etwa 4 kg/h erhöht werden; (2) Wärmeverluste wurden signifikant reduziert, und es gelang, den thermischen Wirkungsgrad auf über 30% mit einem Maximum bei 35% zu erhöhen, und zwar unter Beibehaltung des hohen Kalzinierungsgrads von teilweise über 98%; (3) Der SiC Absorber überstand thermische Schocks bei Temperaturen bis 1600 K.

SUMMARY

A novel indirect-heated solar reactor for the solar production of lime (CaO) was designed, built and tested in a solar furnace at PSI. This 10-kW reactor is operated in continuous mode and consists of a tilted rotary kiln with a specially designed high-temperature resistant blackbody absorber made from SiC and a preheating chamber where the small-grained limestone particles are preheated almost to the reaction temperature. The transition to an indirect-heated reactor including additional design changes was successful: (1) With the improved particle feeding system, the maximum CaO production rate was increased from 1.5 kg/hr to about 4 kg/hr; (2) The heat losses were significantly reduced, and consequently the reactor efficiency was increased to more than 30% with a maximum near 35%, while still maintaining a high degree of calcination exceeding 98%; (3) The SiC absorber withstood thermal shocks at temperatures up to 1600 K.

Projektziele - Project Goals

Substituting concentrated solar energy in place of carbonaceous fuels, as the source of high-temperature process heat for the thermal decomposition of limestone ($CaCO_3$) to produce lime (CaO), is a means to eliminate the dependence on conventional energy resources and to reduce emissions of CO_2 and other pollutants. Solar processing offers a clean and sustainable path for reaching these goals, and the industrial solar lime project with *QualiCal* [1] and *ETH* [2] is a pioneering attempt to address this challenge.

The specific purpose of the project is to establish the technical and economic feasibility of a 0.5 MW thermal input solar calcination plant for the production of lime, e.g. as building material in a developing world setting or for applications in specific market sectors of the chemical industry where high quality standards are required. Indeed, using solar energy instead of heavy fuel oils means that no contamination of the end product occurs during the calcination phase, since no combustion gases (namely, no contaminating agents) are produced.

The current work is based on the experience from previous studies showing that $CaCO_3$ can be calcined with concentrated sunlight and that a high degree of chemical conversion can be achieved [3]-[6]. Furthermore, an internal study done at *PSI* along with several leading players in the cement industry concluded that solar calcination is a feasible process [7]-[9].

The current project started in the year 2000 with preliminary calcination experiments using thermogravimetry, electric furnaces, and the *ETH* Solar Simulator [10]. In the year 2001, a 10-kW solar reactor was designed, constructed, and tested during a first solar experimental campaign at the *PSI* Small Solar Furnace [11], [12]. Within the Ph.D. program, a numerical model is being developed.

The **Project Goals for the Year 2002** were [11]:

1. **Further develop and validate the numerical model to assist in the modification and optimization of the solar lime reactor.**
2. **In a second experimental campaign, establish the thermal performance of the 10-kW solar lime reactor with a *PSI* solar furnace. The thermal efficiency shall be increased significantly to about 30-40% while maintaining the high quality standard of the produced quicklime.**
3. **Begin the conceptual design and economic analysis of a small (ca. 0.5-1 MW) industrial solar lime plant.**
4. **Present the results of the Solar Lime Project at conferences and in peer-reviewed journals.**

Durchgeführte Arbeiten und erreichte Ergebnisse - Results

Within the first half of the year 2002, a novel 10-kW solar lime reactor was designed, constructed, assembled, and pre-tested. A second solar experimental campaign was conducted in the *PSI* Small Solar Furnace between June and September 2002. Data evaluation was done between September and December 2002. Within the Ph.D. program, a numerical model is further being developed and validated. A conceptual design for a 0.5-1 MW solar lime plant is currently being performed. An economic assessment and an ecological study have been conducted. Results of the Solar Lime Project have been presented at conferences and in journals.

DESIGN AND CONSTRUCTION OF A 10-KW SOLAR LIME REACTOR

The first solar lime reactor concept developed within the current project was a 10-kW **direct-heated solar rotary reactor** [11] comprising a conical reaction chamber with a fixed cone angle of 5 degrees. In contrast to the conventional tilted cylindrical rotary kiln, this reactor was operated in a horizontal position. The reaction chamber was lined with refractory concrete and insulated with porous ceramic fiber. This first solar lime reactor prototype produced high purity lime that meets industrial standards [13]. A typical solar energy to chemical energy conversion efficiency was about 13%, the maximum efficiency reaching 20% [14].

The direct-heated reactor was operating for more than 100 hours under concentrated solar irradiation. However, some problems were encountered: (1) Clogging of the feeding tube at high $CaCO_3$ feed rates due to unfavorable shape and sizing of the reaction chamber, thus limiting the maximum CaO production rate to about 1.5 kg/hr; (2) Outflow of fine calcined particles through the open aperture, forming a white powder cloud that reflects and absorbs a significant amount of the incident sunlight; (3) Cracking of the relatively thin refractory lining at temperatures above 1500 K.

To overcome these problems, we decided to change the reactor concept and to develop a novel 10-kW **indirect-heated solar rotary reactor** (Fig. 1), for which a patent application is pending [15]. It comprises a specially designed high-temperature resistant blackbody absorber and a preheating chamber where the small-grained limestone particles are preheated almost to the reaction temperature. The concentrated solar radiation enters through the circular aperture of a water-cooled aluminum front shield and is heating the cavity absorber that delivers the heat indirectly through large surfaces to the limestone material. The rotary reactor is tilted and works in continuous mode of operation. The reactants flow rate is controlled by the feed rate and by mutually adjusting the tilt angle and the speed of rotation of the reactor. The rotation provides a good mixing of the feed material moving through the reactor.

Fig. 1: *The indirect-heated 10-kW solar lime prototype reactor mounted on the experiment table in the Small Solar Furnace at PSI. The experimental set-up comprises the rotary kiln, which is protected with a water-cooled shield, a white target for solar flux measurements, a reactants feeding and a product discharging system.*

The indirect-heated rotary reactor consists of an outer cylindrical steel drum (thickness 1 mm). The inner part of the reactor was manufactured by *INSULTECH AG* [16]. The blackbody absorber is made from *RSiC* (Re-crystallized Silicon Carbide, maximum operating temperature 1600°C). The entire reactor is lined with ceramic insulation material (*INSULBOARD 1600*, maximum operating temperature 1600°C; *INSULTHERM 1000*, maximum operating temperature 1000°C).

For temperature measurements, the reactor is equipped with a *MT32 Mini Telemetry* system from *KMT GmbH* [17] allowing for wireless data transmission from thermocouples (TC) placed inside the rotary reactor. The system consists of eight miniature TC modules, an encoder and a transmitter that are mounted on a plate at the rear of the rotary reactor

(Fig. 2), as well as a stationary receiver-decoder connected to a data acquisition system. All the electronic components on the hot back plate are cooled with air jets to keep their temperature below 60°C.

Fig. 2: *Close view of the mini telemetry system consisting of eight modules mounted on a plate at the rear of the rotary reactor. The electronic components are cooled with air jets.*

Optionally, the reactor may be operated with any conventional external heat source, which is introduced through the aperture into the blackbody cavity in place of concentrated solar radiation. Possible radiation heat sources comprise oil and gas burners including plasma burners, light arcs, induction heating, direct and indirect electric heating elements, etc. As long as the reactants are heated indirectly, the type of the heat source does not influence the conditions inside the burning chamber, i.e. the quality of the end product remains the same.

EXPERIMENTAL TEST OF THE 10-KW SOLAR LIME REACTOR

Prior to manufacturing the indirect-heated solar lime reactor, the novel reactor concept was tested with a model reactor made from steel. It comprised the reaction chamber consisting of a pre-heating zone and a burning zone. Cold tests were performed to verify the correct dimensioning of the reactor geometry, especially of the feeding and discharging systems to avoid clogging. In addition, the operating conditions (drum speed of rotation and tilt angle) for maximum material feed rates and control of the residence time were checked.

After mounting the complete reactor system on the experimental table in the *PSI* Small Solar Furnace, all system components were validated. The data acquisition system *MessHaus* from *Delphin Technology AG* [18] was programmed to record simultaneously the thermocouple temperatures (some of them transmitted from the mini telemetry system), the feeder speed of rotation and the drum speed of rotation, the shutter position (opening angle), and the direct normal solar irradiation. The solar flux distribution on a white target was measured with a *CCD* camera, and the solar power entering the circular aperture was computed using calibration data obtained with a *Kendall* pyrometer.

The raw material feed rate was determined by calibrating the dozing system for two different grain sizes (1.5-2 mm and 2-3 mm) of very pure *Carrara marble* ($CaCO_3$ content close to 98%), provided by *QualiCal*.

The indirect-heated 10-kW reactor prototype was operated with two different external power sources, namely:

- With **solar energy** in the *PSI* Small Solar Furnace (Fig. 3). This solar concentrating system consists of a focusing heliostat plus a parabolic dish concentrator that delivers solar power close to 20 kW with a peak concentration of about 4000 Suns (1 Sun = 1 kW/m^2) on a focal spot of 8 cm diameter. The solar power input into the reactor and, therefore, the limestone burning temperature inside the reaction chamber can be controlled using a shutter, i.e. a Venetian blind flux attenuation device situated between the heliostat and the parabolic dish.

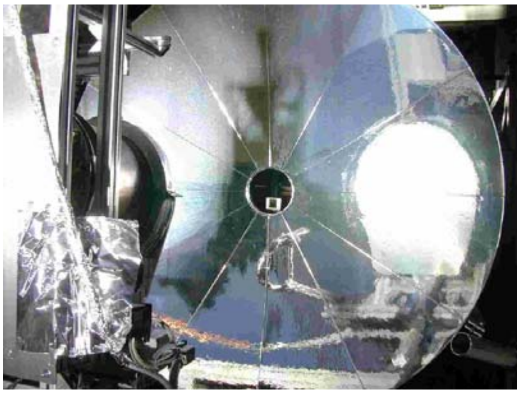

Fig. 3: *The indirect-heated solar lime reactor operated with concentrated solar energy. Front view of reactor with open shutter (left); rear view of reactor with parabolic dish (right).*

- With an **electric heating** element from *KANTHAL* [19] (made from SiC, as used in standard electric ovens) that is inserted into the cavity and completely closes the reactor (Fig. 4a). The cavity temperature is measured with a platinum thermocouple (Fig. 4b) and can be set to a maximum of 1200°C by controlling the power supplied to the SiC heating element. The maximum electric power that can be supplied to the reactor is close to 7 kW.

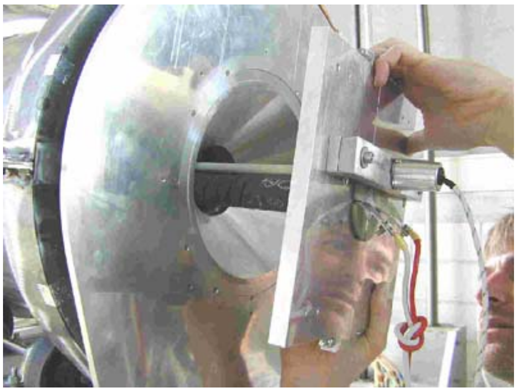

Fig. 4a: *Indirect-heated solar lime reactor operated with electric energy. Inserting the SiC heating element into the aperture.*

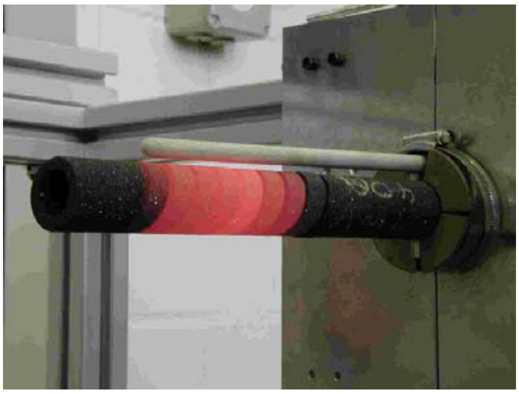

Fig. 4b: *SiC heating element working "in air" during a test. On top of the red glowing coil, the white platinum thermocouple is placed.*

A typical solar experiment was performed according to the following **procedure**:

(1) Set the experiment conditions (particle size, burning temperature, drum speed of rotation, feeder speed of rotation, tilt angle fixed to 5 degrees).

(2) As soon as steady state conditions are reached inside the reaction chamber, take a quicklime sample (minimum 50 g) to determine the degree of calcination; if needed, take also a bigger sample (more than 200 g) to perform reactivity tests.

(3) Determine the degree of calcination by further processing the quicklime sample in the electric furnace at 1100°C until complete calcination is achieved, according to the method described in [20].

(4) Run the reactivity test (only for selected samples with acceptable degree of calcination exceeding 95%) in a Dewar flask by mixing the quicklime with water according to the method described in [21].

The **Main Results of the Solar Lime Experimental Campaign 2002** [22],[23] are summarized as follows:

1. Using electric heating (maximum available power 6.5-7 KW), the maximum CaO production rate with an acceptable degree of calcination (>95%) was about 2.9 kg/hr for 2-3 mm *Carrara* marble (Fig. 5); using solar energy (maximum power ca. 10 kW), the maximum CaO production rate with an optimum degree of calcination (98%) was about 4 kg/hr for 2-3 mm *Carrara* marble.

2. For electric heating experiments, the thermal efficiency of the solar lime reactor, defined as the ratio of process heat used for the chemical reaction to the electric power input, was near 30% with a maximum of 35% (Fig. 5).

   - Remark: Obviously, the available electric power input was not sufficient to completely calcine the limestone material with an acceptable degree of calcination (>95%) for a CaO production rate exceeding 2.9 kg/hr.

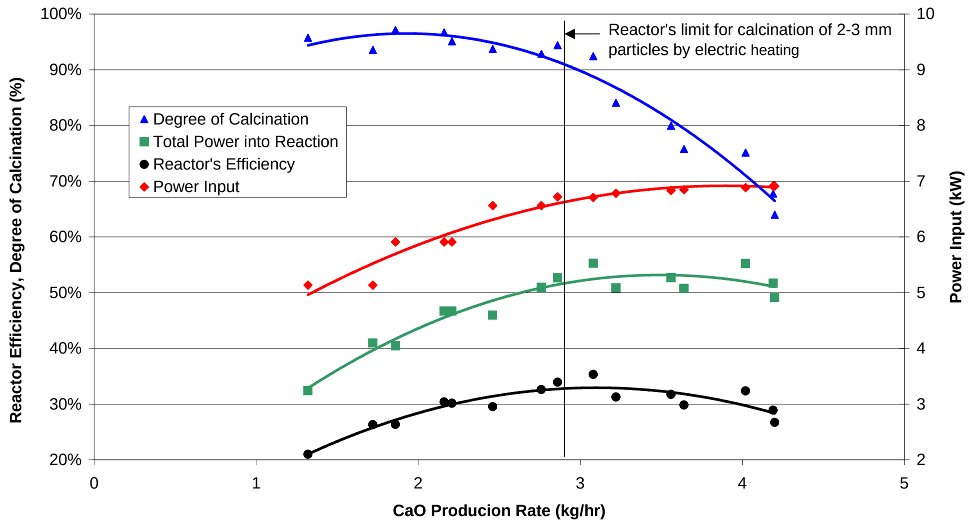

***Fig. 5:** Degree of calcination and reactor efficiency depending on the CaO production rate using an electric heating element as external radiant heat source for the indirect-heated reactor.*

Similarly, for solar experiments the thermal efficiency was more than 30% with a maximum near 35% for a CaO production rate of 4 kg/hr, while still maintaining a high degree of calcination (>95%). Preliminary results from an energy balance yield the relative energy fractions for a variety of solar experiments performed at different operating conditions (Fig.6).

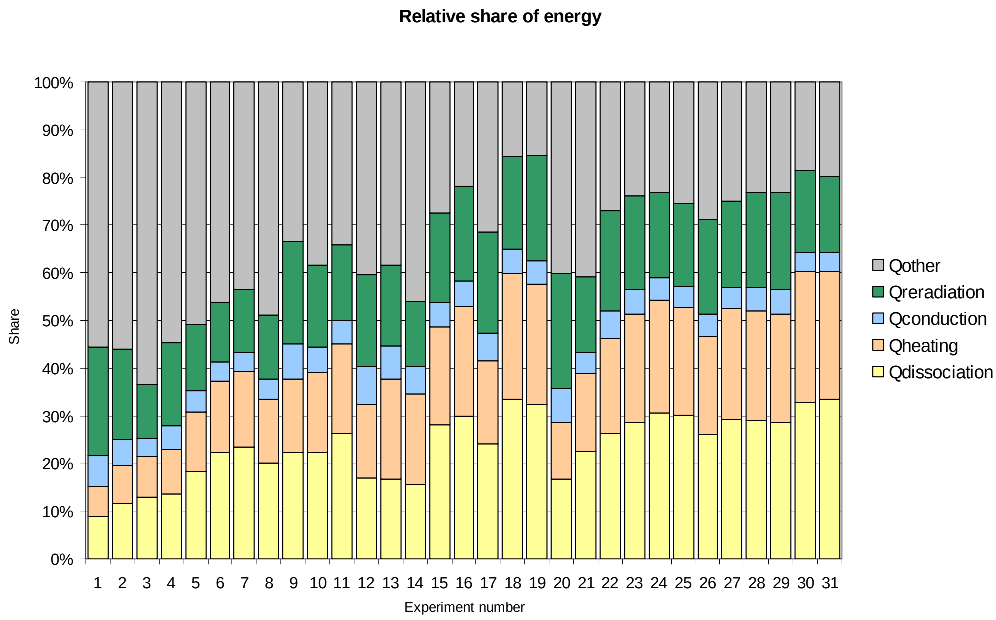

Fig. 6: *Relative energy fractions for a variety of solar experiments. The total energy consumption corresponds to 100%. The higher the CaO production rate, the bigger the fraction of the energy used for heating and dissociation, and the smaller the heat losses.*

3. Quicklime with a wide range of reactivity was produced in the solar lime reactor. T_{60} ranged from 15 seconds (CaO production rate: 2.5 kg/hr; drum speed: 10 rpm) to 12 minutes (CaO production rate: 1 kg/hr; drum speed: 3 rpm), where T_{60} indicates the time needed for quicklime to be heated from 20°C to 60°C when reacting with water. The highest reactivity was reached for short particle residence time (high drum speed) and, consequently, higher production rate (Fig. 7), as well as at lower temperatures (Fig. 8).

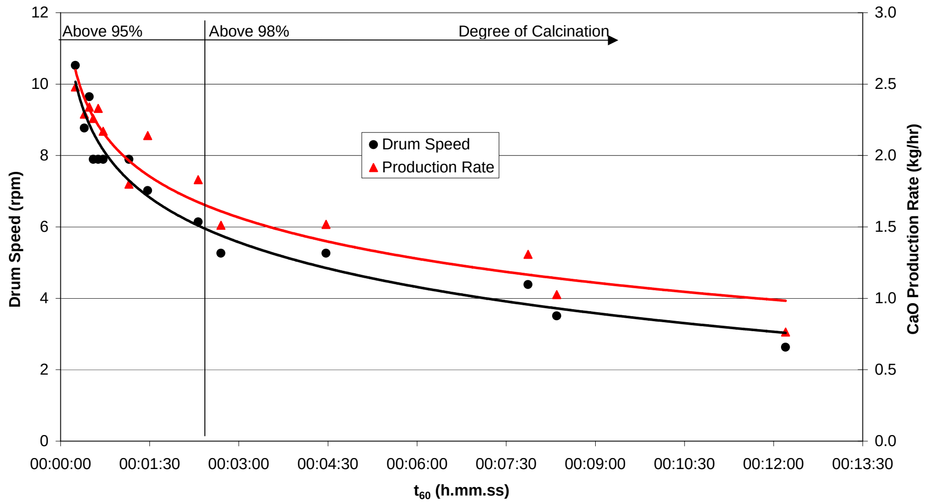

Fig. 7: *Reactivity of quicklime ranging from high (15 seconds) to low (12 minutes) for some specific samples with high degree of calcination, depending on drum speed and CaO production rate (electric heating experiments).*

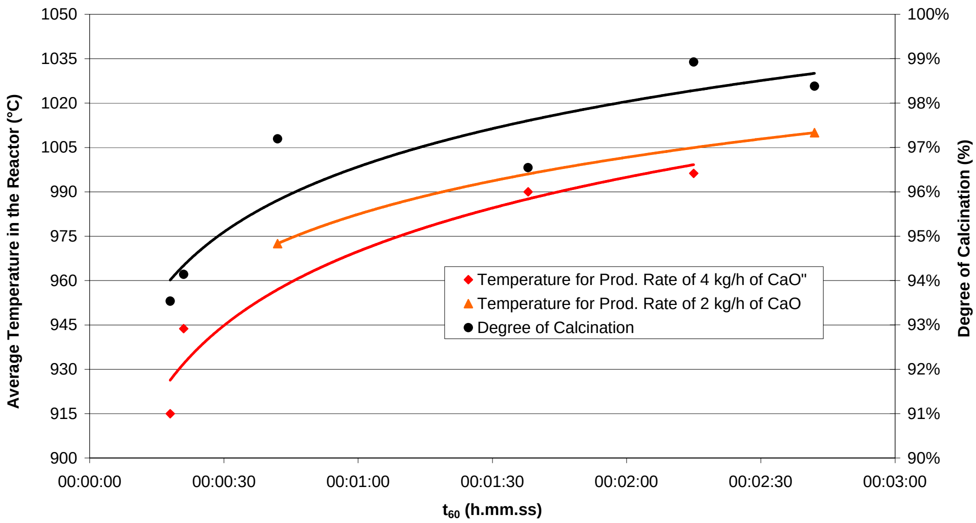

Fig. 8: *Reactivity of quicklime ranging from 40 seconds to 2 min 40 seconds for some specific samples with high degree of calcination (>95%), depending on the average reactor temperature (solar experiments).*

NUMERICAL MODEL FOR THE 10-KW SYSTEM

Within the Ph.D. program, progress was made in numerical modeling:

- A numerical model has been developed for the purpose of understanding the process and the kinetics of the calcination reaction and for being able to scale-up the solar lime reactor. The model takes into account various heat transfer modes (conduction, convection, and radiation) as well as chemical reaction kinetics. So far, the program calculates the temperature distribution and the degree of decomposition as a function of time in a simple geometry.
- The numerical model has been validated with data from a set of experiments performed at *ETH*'s High-Flux Solar Simulator [2]. For different time periods and flux intensities, a thin layer of 2-3 mm $CaCO_3$ particles was placed on a SiC plate and directly irradiated by the artificial concentrated solar radiation. Although quite a good agreement was found between the numerical model and the experimental results, it is suggested to simplify the rather complex experimental set-up in order to be able to better determine the relevant experimental parameters and some of the material properties [24].
- Currently, a method for modeling unsteady systems including chemical reactions is being developed. It is suited for the simulation and calculation of chemical processes in different types of solar reactors, but it can also be applied to combustion processes. Emphasis is given to the coupling of thermal radiation with chemical kinetics.

In addition, a CFD (Computational Fluid Dynamics) study was performed to investigate the convection heat losses in the direct-heated reactor with the conical reaction chamber [25]. Preliminary results suggests that only 6-10% of the unaccounted heat losses are due to natural convection through the open aperture. However, the effects of the CO_2 release were not yet fully taken into account. The CFD model included conduction and radiation (Monte-Carlo) heat transfer as well as chemical kinetics. It will be extended to the new indirect-heated reactor geometry.

CONCEPTUAL DESIGN OF A 0.5-1 MW SOLAR LIME PLANT

A conceptual design of a 0.5-1 MW solar lime plant is currently being developed. It comprises the solar lime reactor concept including feeding and heat recovery system. The reactor will be integrated in the solar lime plant consisting of a heliostat field and a tower. The design and dimensioning of the solar concentrating system will be done with the program WinDELSOL that is based on DELSOL3 [26] and distributed through SolarPACES [27].

ECONOMIC AND ECOLOGICAL ASSESSMENT

A preliminary economic assessment concludes that the economic feasibility of an industrial solar lime plant is not guaranteed *a priori* [28]. However, if certain prerequisites are fulfilled, the solar production of lime may have the potential of being economically viable:

- Only geographical regions with sufficiently high annual solar irradiation should be considered as potential locations for solar lime plants.
- The heliostat costs need to be lower than the actually predicted costs (current assumption: 150 $\$/m^2$). In this respect, one can expect a high economy of scale. Also, high reactor efficiency results in a smaller heliostat field and, hence, lower costs.
- Extremely pure solar produced lime will allow for a much higher selling price than the actual market price for lime.
- The potential for saving fossil fuels and reducing CO_2 emissions may become an incentive for the lime and cement industry to invest in industrial solar lime technology.
- Governmental subsidies and regulations like the CO_2 tax may help introducing the solar lime technology in the market.

The economic analysis is not related to a specific solar lime plant, but presented in a general form. An Excel-File allows cost calculations (NPV=net present value; PBT=pay back time; IRR=internal rate of return; specific lime costs and price; fuel savings and advantages of CO_2 tax) for different input parameters (plant design and size; annual performance; plant capital and O&M costs; specific costs and savings).

A preliminary ecological evaluation concludes that the industrial solar lime production is ecologically beneficial [29]. For example, the savings of greenhouse gas emissions may be as high as 95% of the emissions from an average vertical shaft kiln for conventional lime production. An Excel-File allows calculating CO_2-equivalents (material, transport, fuels, electricity, heliostat, tower) and the CO_2 reduction potential for different input parameters taken from [28].

PUBLICATIONS IN 2002

The results of the project "The Solar Production of Lime" obtained in the year 2001 have been presented at two conferences:

- *10th International Lime Association Congress*, Washington D.C., USA, May 7-10, 2002 [13];
- *11th SolarPACES International Symposium on Concentrated Solar Power and Chemical Energy Technologies,* Zurich, Switzerland, September 4-6, 2002 [14].

Furthermore, first results from the years 2000 and 2001 have been published in the *PSI Scientific Report 2001* [12], and the progress in the year 2002 will be reported in the *PSI Scientific Report 2002* [30].

Nationale Zusammenarbeit – National Cooperation

A close cooperation between the *Solar Process Technology Group* at *PSI* and the *Professorship for Renewable Energy Carriers* at the *Institute of Energy Technology* at *ETH Zurich* [2] is well established. The access to the solar simulator in the laboratory of Prof. Steinfeld is an important prerequisite for the successful work within the Solar Lime Project.

Prof. Dr. A. Steinfeld is the scientific supervisor of the Ph.D. thesis of W. Lipinski, which is an integral part of the project "The Solar Production of Lime". In the year 2002, several *ETH* students of Prof. Steinfeld have accomplished their Semester Thesis within this project, supervised by W. Lipinski, E. Bonaldi, and A. Meier, respectively ([23],[24],[25],[28],[29]).

The new indirect-heated rotary reactor was partly built by *Insultech AG [16]*, a manufacturer of refractory material.

Internationale Zusammenarbeit – International Cooperation

The research activities within the Solar Lime Project are implemented in the framework of the *IEA SolarPACES* Program [27],[31], thus ensuring international coordination and scientific knowledge exchange.

The *PSI Solar Technology Laboratory (STL)* and its industrial partner*, QualiCal* [1], jointly explore the potential of an Industrial Solar Lime Plant. *STL* brings its expertise in developing the science and technology of high temperature solar chemical processes. *QualiCal* brings its expertise in developing the technology for the lime industry.

At the International Lime Association (*ILA*) Congress in Washington D.C. [13], several lime producing companies were contacted. Especially one of them was extremely interested in the technology for the solar production of lime: *Carmeuse*, Belgium [32], actually the second largest lime producer in the world. After performing an internal economic study, they decided to postpone a possible participation in a future follow-up project for a pilot solar lime plant.

Concerning the contact established last year with *Cemex*, Mexico [33], actually the third largest cement company in the world, we have not yet reached an official agreement for a joint collaboration.

Bewertung (Evaluation) 2002 und Ausblick (Outlook) 2003

In the year 2002, a novel 10-kW solar reactor for the production of lime was designed, constructed, assembled, and tested. The thermal performance of the indirect-heated rotary reactor was established during an extended experimental campaign in the Small Solar Furnace at *PSI*, using both solar and electric heating.

From a **chemical** point of view, the performance of the solar lime reactor was excellent. Starting from very pure $CaCO_3$, we were able to calcine the material with a degree of calcination exceeding 95% and to produce high quality quicklime with any desired reactivity (low, medium, and high), depending on the reactor operating conditions.

From an **energetic** point of view, the performance of the reactor was remarkable. The thermal efficiency, defined as the ratio of process heat used for the chemical reaction to the solar power input, reached 35% for both solar and electric heating of the reactor. We expect that the thermal efficiency of this non-optimized solar reactor prototype may be increased to 40-50% by better matching the solar power input with the material feed rate. Beyond that, minimizing the heat losses through better insulating the rotary kiln, and recovering at least part of the sensible heat stored in the hot products (CaO and CO_2) may further improve the reactor efficiency.

From a **mechanical** point of view, the performance of the reactor was good, although some parts need to be modified and improved:

- Due to a manufacturing problem, the SiC blackbody absorber was not built according to the original design. Unfortunately, this inconvenience prevented us from operating the reactor with higher material feed rates, i.e. we could not explore the limits of the reactor with respect to the CaO production rate and the efficiency.
- Due to a manufacturing error, the pre-heating zone was not built according to the specifications. This resulted in small cracks in the refractory lining and in limestone material loss and formation of a powder curtain in the aperture plane. However, this problem was not serious and could be solved by repairing the cracks from time to time.
- Another manufacturing error resulted in a gap between the front insulation and the SiC absorber. Again, this problem was not serious and could be solved by repeatedly closing the gap with SiC coating.

In summary, we have developed a solar reactor that is capable to efficiently calcine limestone particles in the range of 1-5 mm and to produce high purity lime that is not contaminated with combustion products. The quality of the produced solar lime meets industrial standards.

The indirect-heated 10-kW rotary reactor was operating reliably for more than 60 hours under concentrated solar irradiation, and an additional 50 hours using an electric heating element providing 7 kW input power. Both heating modes yielded compatible results, confirming that the reactor may be operated with any conventional external heat source in place of concentrated solar radiation, in order to continuously operate the reactor in case of missing or insufficient solar irradiation.

Our latest results suggest that the novel indirect-heated solar rotary reactor is mature for a scale-up. A conceptual design of a 0.5-1 MW reactor is being developed, partly with the help of a numerical model and CFD simulations [25].

Preliminary results from an economic assessment [28] and an ecological study [29] indicate that the production of solar lime may become an economically viable process that may significantly reduce CO_2 emissions in the lime and cement industry.

The **Project Goals for the Year 2003** are 0:

1. **Further develop and validate the numerical model to assist in the optimization and scale-up of the solar lime reactor.**
2. **Develop a conceptual design of a small 0.5-1 MW industrial solar lime plant.**
3. **Establish costs and CO_2 reduction potential of a 0.5-1 MW solar lime plant.**
4. **Find funding for a 0.5-1 MW pilot and demonstration solar lime plant.**
5. **Present the results of the Solar Lime Project at conferences and in peer-reviewed journals.**
6. **Write the Final Report to BFE.**

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