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# Design and demonstration of a high temperature solar-heated rotary tube reactor for continuous particles calcination

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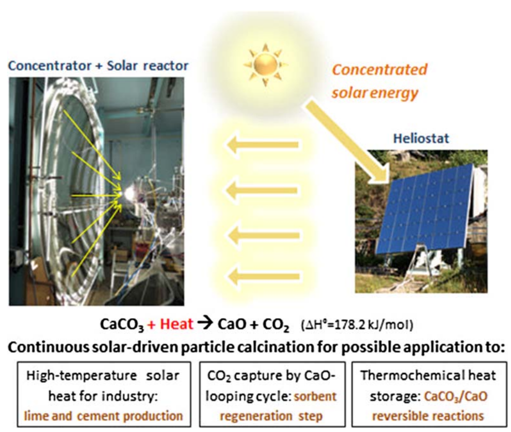
## HIGHLIGHTS

- A new solar chemical reactor for continuous particles processing was developed.
- A rotary tube concept was used for moving bed circulation and uniform tube heating.
- Reliable solar reactor operation during limestone ($CaCO_3$) calcination was demonstrated.
- Pure lime (CaO) was produced at 1000 °C with suitable tube tilting angle and rotational speed.
- The scalable reactor concept can be applied to various solid-gas thermochemical reactions.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

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## ABSTRACT

This study aims at developing a novel solar reactor concept for the continuous processing of reactive particles involved in high-temperature thermochemical reactions (500–1600 °C). The reactor is composed of a cavity-type solar receiver for radiation absorption and heat transfer to a rotary tube in which the reactive particles are continuously injected. This type of reactor shows several advantages in comparison with existing solar thermochemical reactors and the main key characteristics are: (i) external heating by concentrated solar energy, (ii) indirect heating of reactants (reacting zone separated from the zone receiving solar radiation) thus avoiding products deposition on the optical window, (iii) continuous injection of solid reactive particles, (iv) rotation of the tube enabling particles transport and circulation to the outlet, (v) uniform heating of the reactive zone, (vi) direct contact between particles and inner tube wall, enabling optimal heat transfer, (vii) long residence time of particles controlled by the adjustable tube tilting angle, tube rotational speed and particle feeding rate, (viii) reactor adapted to various solid-gas reactions and possible large-scale extrapolation. This versatile solar reactor can be operated for a large variety of thermochemical processes involving solid reactants such as calcination reactions (e.g. decarbonation of limestone for lime or cement production). In this study, proof-of-concept experiments were performed to demonstrate the feasibility of continuous solar calcination of limestone particles ($CaCO_3 \rightarrow CaO + CO_{2(g)}$), which for example could be associated to a cement production solar process, but also applied to CaO-based sorbent regeneration in a $CO_2$ capture process or thermochemical energy storage via $CaCO_3$/CaO reversible reactions.

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## 1. Introduction

The thermal treatment of reactive particles using solar energy consists in substituting fossil fuels combustion (heat supply for endothermic reactions) by concentrated solar energy as the external source of high-temperature process heat for driving the endothermal reaction. Thermochemical solar processes inherently combine several advantages: (i) the use of fossil fuels is avoided, (ii) the $CO_2$ emissions and other harmful pollutants ($SO_x$, $NO_x$) are reduced or suppressed with respect to conventional processes, (iii) solar energy is stored as high-value chemical fuels or commodities for the power, transportation and chemical sectors, and (iv) the reaction products are not contaminated by the combustion by-products.

Nevertheless, most of the previously developed solar reactors for solid particles processing [1–7] suffer from one or several limitations and/or technological constraints, such as:

- Limited maximum temperature of operation because of materials instability at high temperature.
- Low residence time of the particles in the high-temperature zone (for example, entrained flow, aerosol flow or cyclonic reactors), resulting in low conversion yields or incomplete conversions.
- Sensitivity of the chemical conversion to particle size.
- Elevated inert gas flow-rates necessary to ensure the transport or the stirring of the reactive particles (fluidized-bed reactor, particle suspension reactor, entrained flow reactor) and to screen the window in order to avoid particle deposition, which further increases the thermal losses (energy required for heating the excess gas), increases the dilution of reaction products, and decreases the gas residence time.
- Batch processing of reactors (packed-bed reactors) which cannot be compatible with continuous operation and requires reloading of the feedstock after the reaction, thereby inducing additional thermal losses during time-consuming cooling and heating stages.
- Semi-continuous reactors enabling particle injection but precluding continuous extraction and collection of the converted particles or solid residues.
- High thermal gradients and non-homogeneous conversion of the feedstock because of heat and mass transfer limitations (e.g., packed-bed reactors).
- Particles deposition on the optical window, which may cause overheating and damage (case of directly-irradiated solar reactors).
- Formation of dust from particles attrition because of particles mixing and friction, and clogging issues (fluidized-bed, aerosol flow or cyclonic flow reactors).

New solar reactor concepts need to be developed to tackle these numerous limitations and accommodate to industrial processes involving continuous particle flows. Industrial needs in process heat are commonly fulfilled using technologies mainly based on fossil fuels combustion or electrical power occasionally (electrical resistance heat, plasma, etc.). This concerns all the industrial processes using high-temperature heat for the treatment of solid reactive particles (for example, cement, lime, phosphate or clay processing, pyro-metallurgy, thermochemical gasification processes, or any kind of solid-gas reactions). Such energy intensive industries need the major part of their energy input as thermal heat currently provided by combustion and are, after the power and petrochemical industry, the biggest energy consumers and $CO_2$ emitters. The power industry already applies different alternatives to implement renewable energies in their processes. Unfortunately, this option is currently not yet available for energy intensive industries. Therefore, integrating concentrated solar thermal energy in existing industrial plants represents a novel renewable pathway to supply this thermal heat without $CO_2$ emissions.

In this study, the developed solar reactor, based on the rotary-kiln concept [8–13], is aimed at conducting various types of high-temperature solid-gas reactions with continuous reactive particle injection, transport and treatment at high temperature. The rotary-tube solar reactor is designed for the continuous thermal treatment of powders in controlled atmosphere. The possible reactions may encompass for instance the solid/gas decomposition reactions (e.g., limestone calcination for lime and cement production), the metal oxides reduction as part of two-step thermochemical redox cycles for either $H_2$/CO production or thermochemical energy storage, the pyrolysis/gasification of carbonaceous materials (coal, biomass, waste) producing syngas, etc. The utilization of high-temperature solar heat to drive such endothermic reactions is an emerging field that requires developing novel efficient and reliable solar reactors able to realize the continuous conversion of reactive particles.

Calcium carbonate decomposition is the most representative reaction in this type of application of solar process heat for industry. A few lab-scale solar reactor concepts have been previously developed for limestone calcination including mainly fluidized beds [14,15], rotary kilns [14–17] or cyclone [18] reactors. For example, an indirect heating multi-tube rotary kiln prototype was operated reliably at 10 $kW_{th}$ scale for limestone (1–5 mm) calcination [17]. Combined $CaCO_3$ calcination and $CH_4$ reforming was also tested in a particle flow reactor [19].

In the field of solid-gas reactions, calcination is a highly endothermic reaction that can be used for the production of hydraulic binders and that generates greenhouse gas emissions representing about 10% of anthropogenic emissions, among which 40% come from fossil fuel combustion used for production of reaction energy in conventional processes (the remaining 60% come from the chemical reaction). Thus, the use of solar energy in industrial processes represents a major challenge given the expected benefits related to the reduction of fossil fuels consumption and greenhouse gases emissions.

The solar calcination process may be applied to the cement industry and more generally to the thermal treatment of ores as it was shown economically viable [20]. The limestone calcination ($CaCO_{3(s)} \rightarrow CaO_{(s)} + CO_{2(g)}$, $\Delta H^{\circ} = 3184$ kJ/kg CaO) is a key step in the clinker production process for cement production [21,22]. This highly endothermic reaction requires temperatures of ~900 °C to produce quick lime and $CO_2$. With the addition of alumino-silicates (in particular clays for providing silicium, aluminium and iron), the reaction of cement (clinker) production can basically be performed at a higher temperature (around 1450 °C). The energy required for the reaction (between 3200 and 5500 MJ/t clinker [21]) comes from fossil fuels such as coal, natural gas or wastes. The process thus generates large amounts of $CO_2$ emissions (around 1 t $CO_2$/t clinker) coming from both the fossil fuel combustion but also the decarbonation of limestone. The cement industry must thus save the raw resources while minimizing energy consumptions and greenhouse gases emissions. The integration of the solar process in the cement plant consists in substituting the combustion by the external solar energy source. The whole reaction heat for calcination can thus be supplied by the concentrated solar heat source. The converted particles (lime) can be stored for being subsequently processed in the flame furnace of the cement kiln for clinker firing. Another alternative may consist in operating the solar reactor at higher temperatures (1450 °C) in order to directly convert the raw meal into clinker. The process could be operated in a continuous mode, if the cement kiln of the cement plant takes over during off-sun periods (hybrid process). Heat recovery systems can be implemented to preheat the raw meal by the hot exiting gas stream. The whole $CO_2$ exiting the solar calcination process (originated from $CaCO_3$ decarbonation) can be recovered as a pure stream for being valorized later in the form of solar fuels. The solar-driven calcination further eliminates the need for a $CO_2$ capture unit to separate the $CO_2$ from the combustion gases (thereby favoring process economics), since the outlet gas (consisting of pure $CO_2$ if $CaCO_3$ alone is injected) from the solar reactor can simply be collected for being stored. The advantage is that the collected $CO_2$ can be recycled instead of being released into the atmosphere [23–26]. This valorization aims to use sunlight to convert $CO_2$ into transportation