



## Solid particles' recirculation distribution in calcium looping post-combustion carbon capture



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### ABSTRACT

Post-combustion capture of CO<sub>2</sub> from the flue gas of power plants by calcium looping, in which lime is used to capture CO<sub>2</sub>, is currently a promising technology. The decay in CO<sub>2</sub> uptake capacity of natural limestone is an intrinsic disadvantage of this technology, and the design of more stable sorbents has become an important goal. Techno-economic calculations that examine calcium looping must take into account this decay in capacity, and this has to date been accomplished using a formula for the population distribution of particles in the cycling system. However, the formula has some limitations that make it less suited to very stable sorbents, making it difficult to incorporate newer materials in such calculations. We developed a more general formula for the population distribution of solid particles in processes involving recirculations, and compared it with the widely used equation in the calculation of several process metrics. Particular consideration was given to conditions relevant to enhanced or tailored sorbents, for which our modified formula was well suited. In addition, the impacts of calcium looping process configuration on this generalized population distribution of solid particles were studied from mathematical and operational perspectives.

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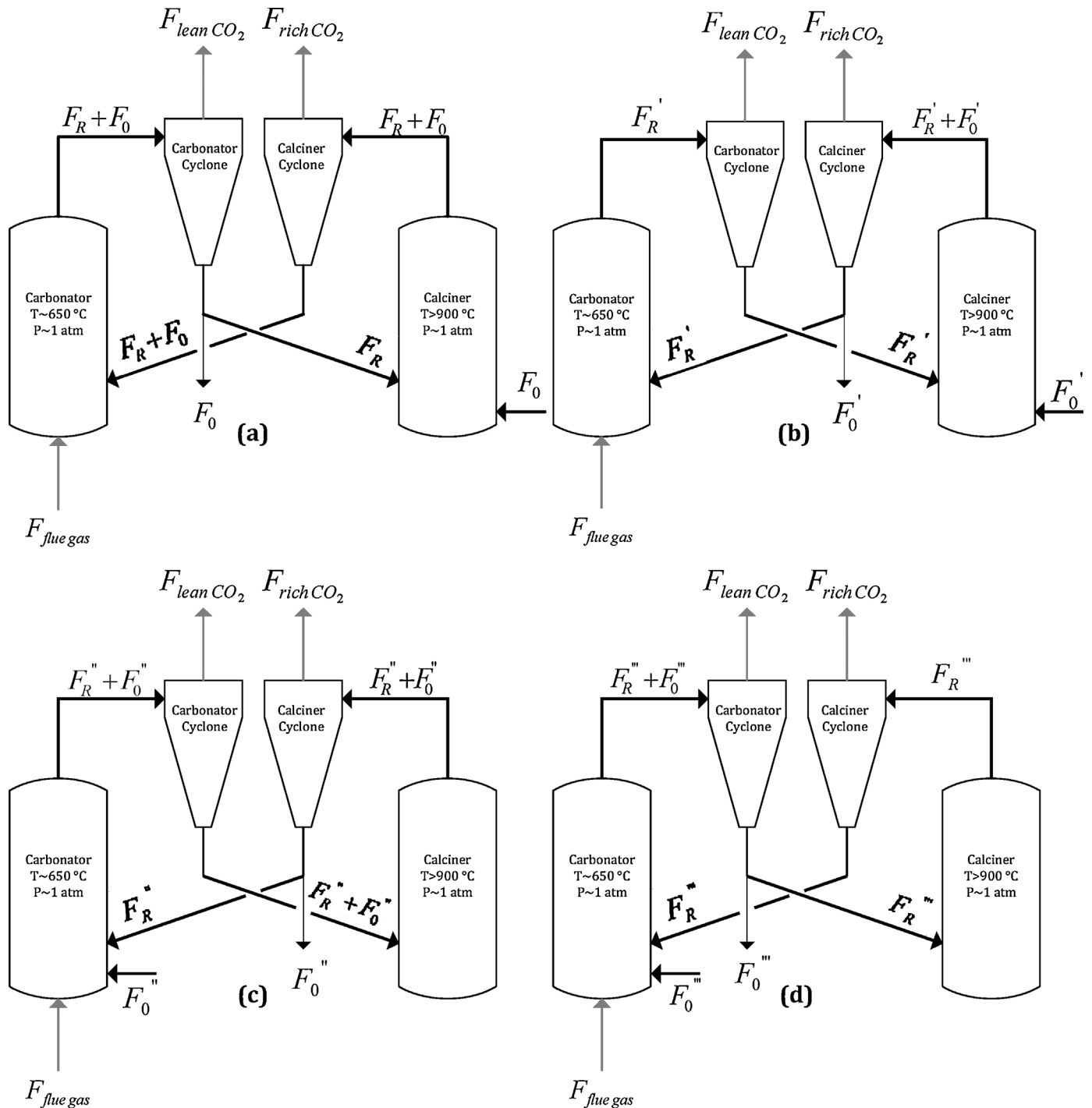
### 1. Introduction

Carbon dioxide has been identified as the main contributor to anthropogenic climate change (Oreskes, 2004). The most important stationary point sources of CO<sub>2</sub> emissions are fossil-fuel-fired heat and power plants where coal provides ~72% of the energy produced, contributing to 41% of global CO<sub>2</sub> emissions (IEA, 2012). As a result, finding efficient and economically attractive methods to capture CO<sub>2</sub> from such plants is an active area of research (Zhao et al., 2013; Pires et al., 2011; Schach et al., 2010). One of the most promising methods of post-combustion CO<sub>2</sub> capture is calcium looping (Zhao et al., 2013), in which lime, composed primarily of CaO, is contacted with a CO<sub>2</sub>-containing flue gas stream in a carbonator, which is typically a circulating fluidized bed (CFB) working at ~650 °C. The solid is then sent through a calciner, typically a CFB working at ≥900 °C, where carbonated CaO (i.e. CaCO<sub>3</sub>) is decomposed to regenerate the sorbent (Fig. 1a), which is then recirculated to the carbonator. The technology is currently being tested on large scales in Spain (Arias et al., 2013), Germany (Ströhle et al., 2014), and Taiwan (Taiwan, 2015). The main advantages of

the process are high working temperatures, which enable heat integration, steam/power generation, and the availability of the low-cost solid sorbent (derived from natural limestone) (Zhao et al., 2013). However, the CO<sub>2</sub>-carrying capacity of limestone decays rapidly over multiple recirculations as a result of sintering at elevated temperatures (Sun et al., 2007). Although there have been many efforts to improve the decay behaviour of CaO, either by designing more stable sorbents (Chang et al., 2013; Coenen et al., 2011; Feng et al., 2006; Liu et al., 2010, 2013; Yang et al., 2009) or by pretreating limestone (Kierzkowska et al., 2013; Manovic and Anthony, 2008; Manovic et al., 2009; Valverde, 2013; Wang et al., 2013), an immediately available option to overcome the problem is to continuously replace some of the spent sorbent with a make-up flow of fresh sorbent ( $F_0$ ), as proposed by Abanades (2002). The addition of a make-up flow and spent-sorbent purge produces a system that can be constructed in four different configurations (Fig. 1), depending on where these components are located. The rate of sorbent replacement (i.e. the ratio of make-up to circulating sorbent) will affect the capture efficiency and the rate at which circulating solids flow, and consequently the heat requirements of the calciner and material requirements of the process. Therefore, a simple mathematical representation of the process enables one to identify reasonable process parameters at which the intended emission reduction target can be reached.

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**Fig. 1.** Schematic of CO<sub>2</sub> capture by calcium looping ( $F_0$  = sorbent make-up stream/spent sorbent purge stream,  $F_R$  = sorbent solid circulating stream); (a) make-up to calciner, purge from carbonator cyclone; (b) make-up to calciner, purge from calciner cyclone; (c) make-up to carbonator, purge from calciner cyclone; (d) make-up to carbonator, purge from carbonator cyclone.

Abanades derived a mathematical relationship between the sorbent replacement rate ( $F_0/F_R$ ) and the maximum CO<sub>2</sub>-capture efficiency (Abanades, 2002); however, that formula was more useful for systems with high sorbent-replacement rates (vide infra). With the advent of higher-performance sorbents and pretreated sorbents, a revised formula that better suits lower replacement rates is desirable. In this paper, an effort to develop a more general model is presented. Furthermore, some key process performance parameters that have previously been examined (e.g. capture efficiency, solids flow rates, streams costs, etc.) are recalculated using

the modified model and compared with the results of Abanades' model.

## 2. Experimental methods

In order to obtain conversion data under industrially relevant conditions, the sorbent (CaCO<sub>3</sub> (Sigma-Aldrich) or CaO/meso-Ca<sub>x</sub>Al<sub>y</sub>O<sub>z</sub>) was exposed to 50 cycles of calcination (3 min at 900 °C under N<sub>2</sub> atmosphere) and carbonation (3 min at 650 °C in 15% CO<sub>2</sub>

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