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# The Mechanism of Self-Reactivation of Cao-Based $CO_2$ Adsorbents

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## Abstract

The self-reactivation phenomenon of CaO-based CO2 adsorbents produced by high-temperature thermal pretreatment improves the durability of adsorbents in calcium looping process. The self-reactivation mechanisms of limestone and Nano-CaO-based adsorbents were explained as the conversion of hard skeleton to soft skeleton and the formation of cracks between Nano-grains respectively. The summarization of the recent researches about the triggering conditions is to provide reference for grasping and utilizing the self-reactivation of adsorbents.

**Keywords:** $CO_2$ adsorption; Calcium looping; CaO-based adsorbent; Self-reactivation

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## Mini Review

Calcium looping (CaL) includes carbonation and regeneration process (Equation 1) has a wide use in flue gas and chemical process for $CO_2$ capture [1-4]. Improving the cyclic stability of CaO-based adsorbents is a research hotspot in this field. Many researches focused on optimizing the pore structure [5-8], doping additives [9-11] and improving the skeleton stability of CaO-based adsorbents to reduce sintering [12].

$$CaO + CO_2 \rightarrow CaCO_3, \Delta H_{298} = -178.8kj / mol$$

Manovic et al. [11]; [13,14] found that limestone exhibited an abnormal 'self-reactivation phenomenon' after thermal pretreatment at 900-1200 °C for 6-48h, in which the sorption capacity of the adsorbent rose with the increasing of initial cycle number (Figure 1). They proposed a 'pore skeleton' model to explain the mechanism of self-reactivation of limestone (Figure 2); [13]. The external skeleton in adsorbents which could react with $CO_2$ was defined as 'soft skeleton' and the internal skeleton without carbonation was defined as 'hard skeleton'. The pre-calcination enhanced sintering of the nascent CaO soft skeleton and the formation of a hard skeleton. During the subsequent CaL cycles, part of CaO in the hard skeleton could be converted into soft skeleton due to the deep carbonation reaction, which caused the increase in the number of active sites. It can be concluded that the self-reactivation mechanism of limestone is the conversion of hard skeleton to soft skeleton in adsorbents.

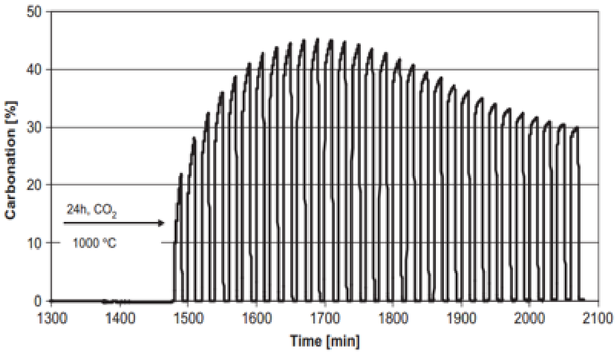

**Figure 1:** The self-reactivation phenomenon of limestone [11].

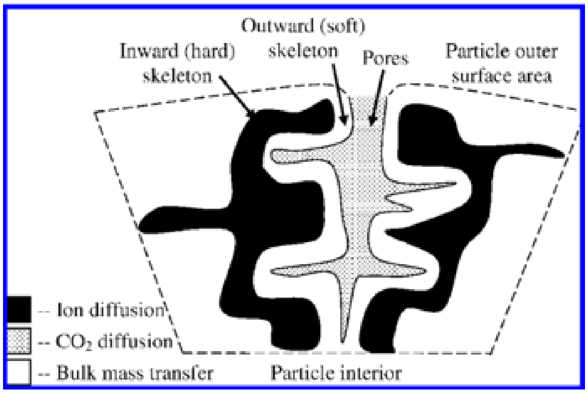

**Figure 2:** Pore-skeleton model of the self-reactivation mechanism of limestone [13].

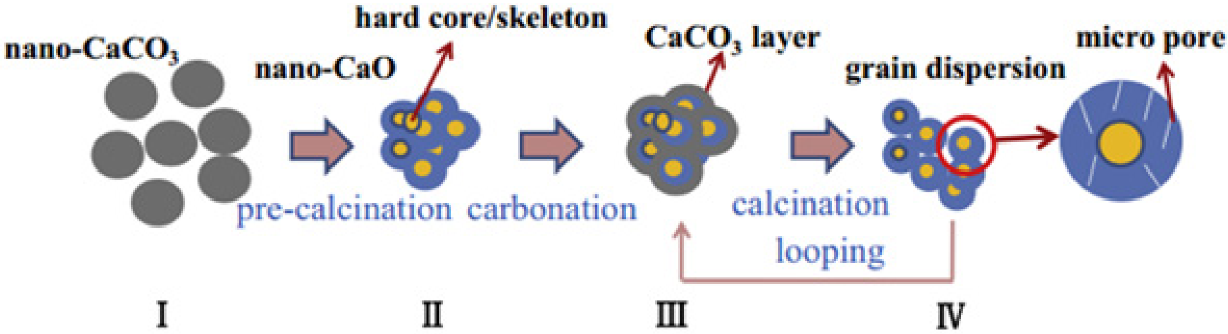

**Figure 3:** Grain-core-pore model of the self-reactivation mechanism of nano-CaO-based adsorbents [15].

The self-reactivation mechanism of Nano-CaO-based adsorbents is different from that of limestone due to the different size of calcium precursors. S.F. Wu et al. [15] proposed a 'grain-core-pore' model to explain the self-reactivation mechanism of Nano-CaO-based adsorbents (Figure 3). After nano-$CaCO_3$ (I) was pre-calcined at high temperature for a prolonged period, the Nano-CaO particles were sintered and accumulated into larger particles with a hard core (II). During the first carbonation, larger CaO particles (II) were carbonated with $CO_2$ and the reaction formed a thick $CaCO_3$ layer (III). At the subsequent calcination stage, the decomposition of $CaCO_3$ layer leaded to the release of $CO_2$ and the return of part of Nano-CaO grains (IV), thereby increasing the number of active sites. In summary, the self-reactivation mechanism of Nano-CaO-based adsorbents is the formation of cracks between Nano-grains.

Based on these mechanism models, there are many researches focusing on the triggering conditions for self-reactivation of adsorbents. First, the self-reactivation triggering of the limestone adsorbent required severer thermal pretreatment operation conditions (high temperature, long time and $CO_2$ atmosphere) to bring more hard skeletons [11], while the Nano-CaO-based sorbent was not resistant to sintering, requiring milder thermal pretreatment conditions [16]. Moreover, the increase of carbonation temperature and the prolongation of carbonation time as well as the reduction in the regeneration temperature were beneficial to the self-reactivation triggering of adsorbents [15]. Finally, the anti-sintering dopant was favorable for the self-reactivation triggering of adsorbents [13,15].

The self-reactivation phenomenon of CaO-based adsorbents induced by thermal pretreatment can effectively improve the sorption stability of adsorbents in CaL process, which has a practical application value. However, there are still some shortcomings in the recent studies of self-reactivation of CaO-based adsorbents. First, the reason for different effects brought by different kinds of dopants is not clear. Besides, in addition to focusing on the self-reactivation triggering conditions, there are few researches paying close attention to what factors affect the self-reactivation performance, which includes the degree of increase in sorption capacity during self-reactivation and the cyclic stability after self-reactivation.

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