



Original Research Paper

Attrition resistance of calcium oxide–copper oxide–cement sorbents for post-combustion carbon dioxide capture

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ARTICLE INFO

Article history:

Received 12 November 2015

Received in revised form 8 March 2016

Accepted 10 March 2016

Available online 17 March 2016

Keywords:

Attrition

Calcium looping

CLC

Cadomin

Copper oxide

ABSTRACT

Integrating calcium looping (CaL) and chemical looping combustion (CLC) is potentially more energy efficient than existing technology for post-combustion CO₂ capture. Mixtures of CaO, CuO and cement (cem) are promising sorbents but attrition losses due to abrasion and fragmentation could incur an unacceptable economic penalty. We assessed the attrition resistance of many fluidized bed powders with a jet mill at orifice velocities from 180 m s⁻¹ to sonic velocity. The attrition resistance of calcium oxide–copper oxide–cement sorbents and crushed cadomin limestone are poorer than vanadyl pyrophosphate (VPP) and fluid catalytic cracking catalyst (FCC). The attrition rates of VPP and FCC at an orifice gas velocity of 180 m s⁻¹ were 5 mg h⁻¹ and 7 mg h⁻¹, respectively; it was 10 mg h⁻¹ for powders with a mass fraction of 50 % CuO 40 % CaO and 10 % cement. The (CaO)₉₀cem fragments to smaller sister particles and produces fines but the fines collected overhead in the thimble filter decreases with temperature. The (CaO)₄₀(CuO)₅₀cem₁₀ attrition resistance improved above 500 °C and the d₅₀ of the pellets remaining in the jet cup increased from 330 μm to 422 μm: the Cu agglomerated the pellets. Attrition rates increased with the square of the orifice diameter, the cube of the gas velocity and to the power 0.4 with gas molecular weight.

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1. Introduction

Two thirds of global anthropogenic carbon dioxide emissions are due to burning fossil fuels [1]. In 2012, fossil fuels contributed 82 % of the global energy demand and will increase 37 % by 2040. Since 1958, the CO₂ concentration has increased by almost 100 ppm [2]. It has risen exponentially and will reach 450 ppm (Eq. (1)) by 2040 if it continues on the same trend.

$$ppm_{CO_2} = 317 + 0.28(y - 1958)^{1.4} \quad (1)$$

It is imperative to mitigate the impact CO₂ emissions might have on the environment. Substituting fossil fuels with renewable sources – wind, solar, geothermal and hydroelectric is the preferred option but the economics remain uncompetitive in many applications—particularly for automotive.

Carbon capture and sequestration (CCS) adds to the cost of CO₂ emissions from fossil power generation plants. Three steps make up CCS: CO₂ capture (15\$/t to 75\$/t), transport (1\$/t to 8\$/t) and

storage (0.5\$/t to 8\$/t) [3]. Currently, amine scrubbing and oxy-fuel combustion are the only technologies practised commercially. Retrofits to existing fossil-fuel combustion plants reduce their overall efficiency, which is related to the energy required to regenerate the solvent [4–6].

Dry solid sorbent technology is an alternative to aqueous solvent processes to capture CO₂. The calcium oxide–calcium carbonate couple (CaL) has superior economics and energy efficiency compared to solvent technologies [7–10]. In this process calcium oxide scavenges CO₂ from the flue gas and forms calcium carbonate. The carbonate calcines in a second vessel to CaO and releases CO₂ and then the CaO returns to the first vessel.

In Chemical Looping Combustion (CLC), a metal oxide cycles between a net reducing environment where a carbon compound reduces the metal oxide to a net oxidizing environment in which oxygen regenerates the solids. Both the CaL and CLC technologies require two vessels. Combining CaL and CLC replaces the air separation unit (for the regeneration of the CaCO₃), with an additional vessel to regenerate the metal oxide. The flue gas from the plant contacts CaO in the carbonator and forms CaCO₃. The carbonate goes to a second vessel (calciner/reducer) where methane reacts with oxygen from the CuO and generates heat to calcine the CaCO₃ to CaO. At the exit of this vessel a fraction of

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Nomenclature

Alphabetical symbol

A	cross-sectional area of the fluidized bed (m^2)
Ar	Archimedes number ($d_p^3 \rho_g (\rho_p - \rho_g) g \mu^{-2}$)
cem	cement
d_o	orifice diameter (mm)
d_p	particle diameter (μm)
d_{sv}	mean surface-volume diameter (μm)
d_{50}	particle diameter based on weight fraction (μm)
k_j	specific attrition rate constant for material, j
M	mass (g)
\dot{m}	mass flow rate, (g s^{-1})
Q	volumetric flow rate (STP – L min^{-1})
R_{aj}	attrition rate of component j (mg h^{-1})
t	time (h)
T	temperature ($^\circ\text{C}$)

U_c	critical velocity (m s^{-1})
U_{mf}	minimum fluidization velocity (m s^{-1})
U_o	orifice velocity (m s^{-1})
U_s	orifice design velocity (m s^{-1})
W	mass of solids in the fluidized bed (kg)
x	mass fraction
y	year

Greek symbol

β_i	Coefficients in Eqs. (6), $i = 0, 3$
ρ_b	bulk (Scott) density (kg m^{-3})
ρ_g	gas density (kg m^{-3})
ρ_p	particle density (kg m^{-3})
μ	viscosity (Pa s)
ξ	voidage

the solids are returned to the flue gas vessel and the other fraction enters the air reactor (regenerator) where oxygen restores the copper to its oxide form (and generates more heat) (Fig. 1).

CANMET developed CaO–CuO sorbents with calcium aluminate cement binders [11,12]. These sorbents must be reactive, stable and able to resist mechanical stresses due to abrasion (particle–wall contact in cyclones, for example) and particle–particle and particle wall collisions (particularly at high velocity nozzles.) Attrition can render the capture process economically unattractive. Moreover, sorbents with metals such as Ni poses a health and environmental problem risk.

For commercial fluidized beds, we define the attrition rate as the mass of powder hoppers collect downstream of cyclones as a function of time, R_a . The cut-off particle diameter for catalytic reactors is 20 μm at an efficiency of 99.99 %. Attrition rates depend on material properties: particle size, surface structure, shape, density and porosity and process conditions: gas and solids velocity, wall hardness, solids residence time, system temperature and pressure. Typically, catalytic fluidized bed reactors operate with Geldart Group A powders with a d_{50} of 70 μm . Sorbents and metal oxides in CaL and CFB process can be an order of magnitude larger - Geldart Group B powders.

Accelerated attrition tests guide the catalyst design process and indicate the amount of binder required to achieve mechanical stability suitable for fluidized beds [13,14] and include: air jet mills [15,16], jet cups (Davison test) with a tangential entry [17–19], cyclones [20], particle impact [21–23] and fluidised beds [24–26]. Ghadiri et al. [27], Boerefijin et al. [28] proposed a general relation for FCC attrition rates based on a single particle impact test that represents the jetting region of a fluidised bed (Eq. (2)).

$$R_{aj} \propto U_o^2 \quad (2)$$

Air jet mills attrit particles with a jet up to sonic velocity that impinges on the particles and causes them to collide. A jet mill measured the VPP attrition resistance catalyst during the development of DuPont's CFB process [29]. The catalyst spray dried in a 1.4 m dryer had a 18 mg h^{-1} attrition rate; powder from an 8 m spray dryer had an attrition rate of 4 mg h^{-1} [30]. Rather the 343 m s^{-1} (or sonic velocity), which is standard for the ASTM-D-5757-00 test, they measured the VPP attrition rate at 232 m s^{-1} ; the higher gas velocities shattered the core–shell catalyst microspheres resulting in unacceptably high rates. VPP would fail the ASTM test but in the commercial reactor, the attrition rate was only 1 kg h^{-1} , whereas the design basis allowed for 10 kg h^{-1} to 25 kg h^{-1} .

In CaL processes, limestone fragments due to chemical–thermal stresses [31], CO_2 pressure that develops when it heats, surface abrasion and particle–particle and particle–surface collisions [21]. Particles can chip (cleave asperities—typical of FCC), split (fragment—characteristic of limestone sorbents) or disintegrate (shatter—core–shell catalyst like VPP).

Werther and Xi [20] related jet induced attrition and the surface energy created with the energy expended (Eq. (3)).

$$R_{aj} \propto n_o \rho_g d_p^2 U_o^3 = k_j d_p \dot{m} U_o^2 \quad (3)$$

where k_j is the material specific attrition constant, ρ_g is the gas density, d_o is the orifice diameter, U_o is the orifice gas velocity and n_o is the number of orifices. They tested three fluidized bed geometries with fluid cracking catalyst (FCC). Thon et al. [32] added the surface area fraction of the particles in an interval j ($\Delta Q_{2,i}$) to this equation.

McMillan et al. [33] measured coke and sand attrition rates due to gas injected through sonic nozzles into fluidized beds. They reported that the particle breakage frequency (attrition rate) was independent of the mass of solids in the bed and that it depended mostly on the nozzle geometry, operating conditions and fluidized bed velocity.

$$R_{aj} = k_j \alpha \beta d_N^{1.131} U_N^{0.55} (\rho_g U_N^2)^{1.635} \left(\frac{U_g - U_{mf}}{U_{mf}} \right)^{0.494} \quad (4)$$

where α is an index related to type of solid ($\alpha = 1$ for silica and $\alpha = 0.8$ for coke), β is a geometry coefficient and the index, N , relates to nozzle diameter (Eq. (4)).

Knight et al. [34] adopted the ASTM-D-5757-00 and tested many sorbent compositions produced by CANMET Energy and the University of Laval. The mechanical resistance of the calcium aluminate cement bound sorbent was better than the crushed limestone but it was inferior to the silica coated sorbent. The attrition rates at 500 $^\circ\text{C}$ were two to three times higher than the experiments at ambient conditions.

The exponents on the factors for the jet attrition mill correlation (Eq. (3)) are much different than for the nozzle immersed in a fluidized bed (Eq. (4)). In the attrition mill, $R_{aj,i}$ is proportional to the orifice diameter squared whereas it is linear for the nozzle; it is third order with respect to gas velocity for the orifice and almost 4th order for the nozzle; finally, it increases linearly with gas density in the orifice and to the power 1.6 in the nozzle. Lin and Wey [35] summarized correlations derived from attrition tests run at up to 800 $^\circ\text{C}$ with various materials including sand, limestone, carbon and alumina. They measured the attrition resistance of silica sand

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