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Dry scrubbing of acid gases in recirculating cyclones

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Abstract

This paper describes a laboratory-scale study on the use of recirculating cyclones as reaction chambers for dry scrubbing of gaseous HCl with solid slaked lime particles. This gas cleaning system combines a numerically optimized reverse flow gas cyclone (RS_VHE geometry) with a straight-through cyclone concentrator, which simultaneously increases the capture of the solid particles and promotes their partial recirculation. A laboratory-scale study was undertaken to test this technology and to compare its performance to a modified Stairmand HE reverse flow cyclone without recirculation. The experimental conditions were: reaction temperature ≈326 K, gas flow rate $\approx 2.9 \times 10^{-4}$ N m^3 s^{-1} and relative humidity of the gas ≈8.5%. The experimental variables tested were the solids load ($1.0–9.2 \times 10^{-7}$ kg s^{-1}) and HCl concentration ($0.4–2.8 \times 10^{-2}$ mol m^{-3}) in the inlet gas.

The experimentally obtained particulate removal efficiencies with the recirculating cyclones (≈98%) were higher than those obtained with the Stairmand HE cyclone (≈93%), with the additional advantage of having significantly lower pressure drop. As for the acid removal efficiencies (≈10–96%), no significant differences were found between the two systems tested under the same experimental conditions.

The possibility of using optimized recirculating cyclones for gas cleaning in a dry scrubbing process is very promising, since this is a low cost technology, highly efficient both for the removal of acid gases and for the capture of solid particles, which has the advantage of not requiring a post-reaction de-duster.

Keywords: Recirculating cyclones; Gas cleaning; Dry-scrubbing; HCl; Slaked lime; Cyclone reactor; Municipal waste incinerator

1. Introduction

Solid waste incineration processes generate large quantities of atmospheric pollutants: acid gases (HCl, SO_2, NO_x, etc.), volatile organic compounds (VOCs), heavy metals and particulate matter [1]. Acid gases are harmful to public health since when inhaled they can cause respiratory illnesses, alteration in the lungs' defences, and aggravation of existing cardiovascular or chronic lung diseases [1,2]. Furthermore, in the presence of ambient moisture, acid gases produce acid rain, which is a well-known global environmental menace.

The removal of these toxic compounds, namely HCl and SO_2, from waste gases can be done through dry, semi-dry or wet scrubbing processes. Among these, dry scrubbing processes are most commonly used [3,4], and have the main advantage, compared to wet processes, of having lower capital and operating costs and requiring less space [1], with acid removal efficiencies higher than 90% [5,6]. Dry-scrubbing is performed through the injection of a finely divided dry powder of an alkaline reactant into the gas stream. Calcium based sorbents (namely $CaCO_3$, $Ca(OH)_2$ and CaO), are widely used due to their low cost and high efficiency for acid gas scrubbing [4,5]. The acid gases are removed through adsorption and reaction at the solid's surface. Downstream, some dust removal equipment is required to capture the solid product formed together with the un-reacted solid powder.

It would be of great practical and economical relevance to promote the dry-scrubbing and the dust removal in the same equipment, which is possible using a cyclone as reaction chamber. Cyclones are gas–solid centrifugal separators, characterized by simple designs, low investment and maintenance costs and with the ability to operate at high loadings, high temperature and pressure. Several studies have proven that cyclones are also powerful gas-solid reactors [7–9].

There are different cyclone configurations, but the main design for industrial gas cyclones is the tangential reverse-flow type [10,11]. Tangential cyclone design is traditionally based on geometries characterised by a fixed ratio of seven key-dimensions. Recent studies have proven that it is possible to

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Nomenclature

R/SR	ratio between the amount of slaked lime fed and that corresponding to the stoichiometric quantity
X_C	conversion of the solid captured in the cyclone reactor relative to the fresh feed
X_F	conversion of the solid captured in the filter relative to the fresh feed

Greek letter

η	HCl removal efficiency

design significantly improved reverse-flow gas cyclones by solving adequate numerical optimisation problems [12,13]. Based on this approach, a new cyclone geometry (RS_VHE) was identified and patented [14]. Cyclones with the RS_VHE geometry revealed a clear superiority over the Stairmand HE and other HE designs at laboratory, pilot and industrial scale [12,14,15].

One way to increase cyclone collection efficiency beyond improving its design is to promote partial recirculation with a straight-through cyclone concentrator. The proposed recirculation system (see Fig. 1) locates the collector (reverse-flow cyclone) upstream from the concentrator (straight-through cyclone). Partial recirculation of the gas and solids is promoted by some means, such as by a venturi for laboratory-scale or by a blower or ejector for industrial-scale and high temperature applications. These recirculating cyclones were already tested both at laboratory, pilot and industrial scales, achieving, under some circumstances, particle removal efficiencies comparable to those of a jet-pulse bag filter [14,16–18].

Previous studies have already proven the high capacity of reverse flow cyclones to remove HCl from a gaseous stream by the injection of solid particles of slaked lime [9,19]. The aim of the present work is to verify if there are significant differences between the behavior of recirculating and simple reverse-flow cyclones for dry cleaning of acid gases. These differences are anticipated to be: the reduction in fresh reactant's consumption due to the recirculation of some partially converted solid particles to the reaction chamber, and the removal efficiency of solid particles which does not require the use of a post reaction de-duster.

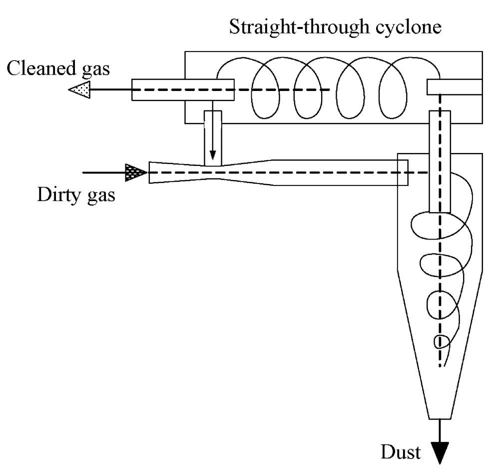

Fig. 1. Recirculating cyclones.

2. Experimental

Fig. 2 shows a schematic diagram of the experimental equipment, which basically promotes the continuous contact between slaked lime solid particles injected in a gaseous stream containing HCl in the cyclone reactor system. This equipment is placed in a high precision temperature controlled gas-chromatographic oven. The solid particles of slaked lime were injected in a carrier gas (N_2) using a Wright MK2 dust feeder [20]. To avoid the presence of large agglomerates at the test cyclone inlet, a 0.07 m diameter Stairmand HE de-agglomerating cyclone was placed after the dust feeder. The particles were joined together with a gaseous stream of N_2 with a controlled concentration of HCl and humidity level, immediately before the inlet of the cyclone. The effluent gas concentration is measured by bubbling a known fraction of this stream into distilled water by means of an appropriate external circulation airlift absorption vessel [19,21]. The HCl is completely absorbed in the water, and the pH of the resulting solution is monitored on-line with the aid of a computerized data acquisition system. The HCl removal efficiency is then calculated through material balances.

As a cross checking, at the end of the experiment the solids captured in the cyclone reactor system are dissolved in distilled water and chemically analyzed for chlorides by an appropriate potentiometric titration. The same procedure is followed for the solids captured in the back filter at the exit of the cyclone. The conversions of the solid reactant in the cyclone and in the filter are then obtained through material balances.

In this experimental study the temperature of reaction was 326 ± 0.4 K, the total gas flow rate at the inlet of the cyclone was $(2.9 \pm 0.1) \times 10^{-4}\,\mathrm{N\,m^3\,s^{-1}}$ and relative humidity of the gas was $8.5 \pm 0.2\%$. The experimental variables tested were the solids load (1.0–$9.2 \times 10^{-7}\,\mathrm{kg\,s^{-1}}$) and HCl concentration (0.4–$2.8 \times 10^{-2}\,\mathrm{mol\,m^{-3}}$). Two sets of experiments were performed: one using optimized recirculating cyclones and another with a modified Stairmand HE. The recirculating system is one RS_VHE cyclone with 0.02 m internal diameter, and one straight through cyclone also with 0.02 m internal diameter. The recirculation of the gas and particles is assured by a small ASME ventury (0.002 m throat diameter). The recirculation fraction was about 14% [17]. The Stairmand HE cyclone of 0.02 m internal diameter was modified by decreasing the vortex tube diameter by 60%, in order to increase its collection efficiency for the slaked lime test dust from 57% [12] to over 90%. All the cyclones are made in polytetrafluorethylene (PTFE) to avoid unwanted HCl consumption. Due to commercial sensitivity, the exact geometry of the RS_VHE cyclone cannot be disclosed, but the relative magnitudes of the dimensions that are thought to be most critical are as follows: the cyclone entry is essentially square; compared with a Stairmand HE design with an equivalent diameter, the cylindrical body of the RS_VHE is about 50% shorter, its vortex finder is narrower and its total length is about 6% longer. More details can be found elsewhere [12].

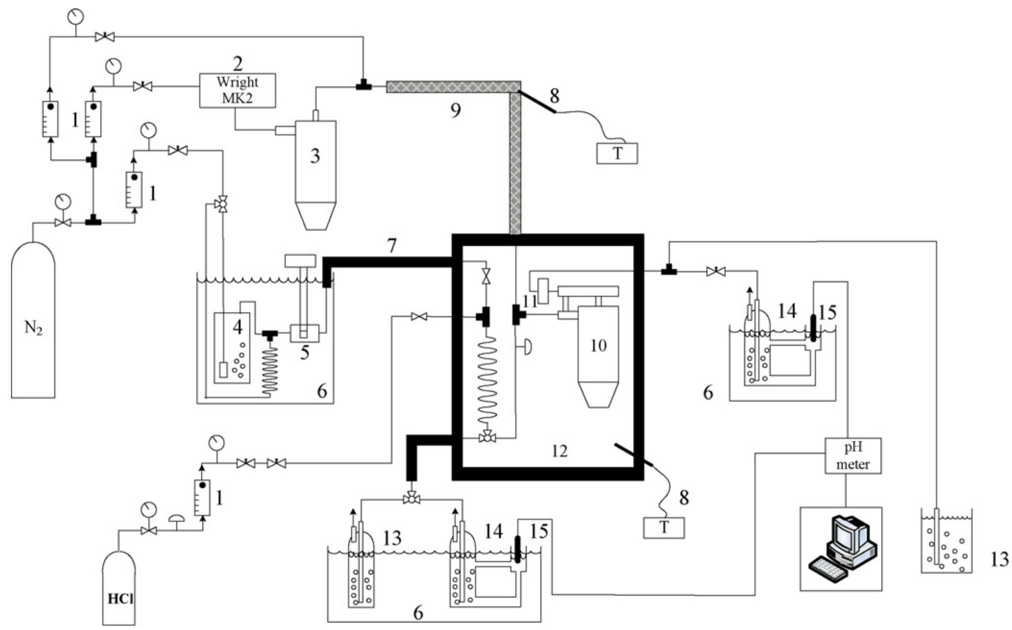

Fig. 2. Experimental set-up. (⚲) Manometer; (⚲) pressure regulator; (⚲) on/off valve; (⋈) needle valve; (⋈) three-way valve; (▷◁) check valve. 1: Rotameter; 2: solid particles feeder; 3: deagglomeratiing cyclone; 4: humidification column; 5: hygrometer; 6: temperature controlled bath; 7: thermal insulation; 8: thermocouple; 9: electrical heating tape; 10: cyclone reactor; 11: filter; 12: oven; 13: absorber; 14: absorber with lateral chamber; 15: pH electrode.

3. Experimental results and discussion

Preliminary experiments were carried out to evaluate the particles' collection efficiency of the cyclones in the absence of reaction. Slaked lime particles with a volume median diameter of 2.13 μm [12] were injected from the Wright dust feeder at an air flow rate from $(2.5–4.8) \times 10^{-4}$ N m^3 s^{-1} and inlet loads from 160 to 1545 mg (N^{-1} m^{-3}).

For the recirculating cyclones, the average particle collection efficiency in the absence of reaction was $98.0 \pm 0.7\%$, and for the Stairmand HE cyclone, $93.2 \pm 1.0\%$, both at 5% significance level. It is important to point out that, despite the modified Stairmand HE cyclone having a much higher pressure drop as compared with the RS_VHE design (typically three times more), its collection efficiency is much lower. This corroborates previous findings that show much higher efficiencies for the RS_VHE design as compared with the Stairmand HE and other HE designs [12].

It is well known that the cyclone efficiency increases with the decreasing diameter of the cyclone, so a cyclone of 0.02 m internal diameter is hardly representative of industrial scale. However experimental results at pilot and industrial scales with RS_VHE cyclones, with internal diameters ranging from 0.46 to 0.70 m, with recirculator diameters up to 1.3 m, showed much higher particle removal efficiencies as compared to smaller diameter multicyclone systems, viz. above 90% against 40–60% for biomass boiler applications [14,17,18].

In the experiments performed for the dry-scrubbing of HCl it was verified for both cyclone systems tested that when the feeding of the solid reactant starts, the HCl concentration in the effluent gas changes almost instantaneously to a constant value, corresponding to a steady state removal efficiency. This behavior had already been observed in previous studies [9]. The ratio between the amount of slaked lime fed and that corresponding to the stoichiometric quantity is represented by *R/*SR. This experimental variable can be changed either by altering the amount of solids fed as well as by changing the HCl concentration. The experimentally obtained values of HCl removal efficiency (η) varied from 10% for an HCl concentration of 1.3×10^{-2} mol m^{-3} and an *R/*SR of 0.6, to 96% for an HCl concentration of 0.67×10^{-2} mol m^{-3} and an *R/*SR of 6.5. Fig. 3a and b shows the experimental acid removal efficiencies for different *R/*SR values at two fixed HCl concentrations, and Fig. 4 shows the same information for a fixed load of slaked lime.

These results show that, under the present experimental conditions, the HCl removal efficiency increases with *R/*SR, which means that the dry-scrubbing process is more efficient for high solid's loadings and small HCl concentrations.

As can be observed in Figs. 3a and b and 4, in the experimental conditions tested, no significant differences occur for the acid gas removal between the performance of the recirculating cyclones compared with the single reverse flow cyclone. In an attempt to explain this behavior, the residence time of the gas in both systems was estimated to be 0.030 and 0.026 s, respectively, for the Stairmand HE and RS_VHE cyclones [22], and 0.091 s for the recirculation system [23]. This means that the reactor vessel is essentially the cyclone and not the recirculation loop, as further demonstrated below.

The results of the solid reactant's conversion obtained in the cyclone (X_C), relative to the fresh feed, are presented in Fig. 5a. The values obtained indicate that the usage of the solid reactant is low, with conversions on average lower than 25%, and are apparently not affected by *R/*SR. This behavior has been observed with all HCl concentrations studied. This indicates that the solid always reacts only to a certain extent, which means that the reaction is most probably controlled by product layer

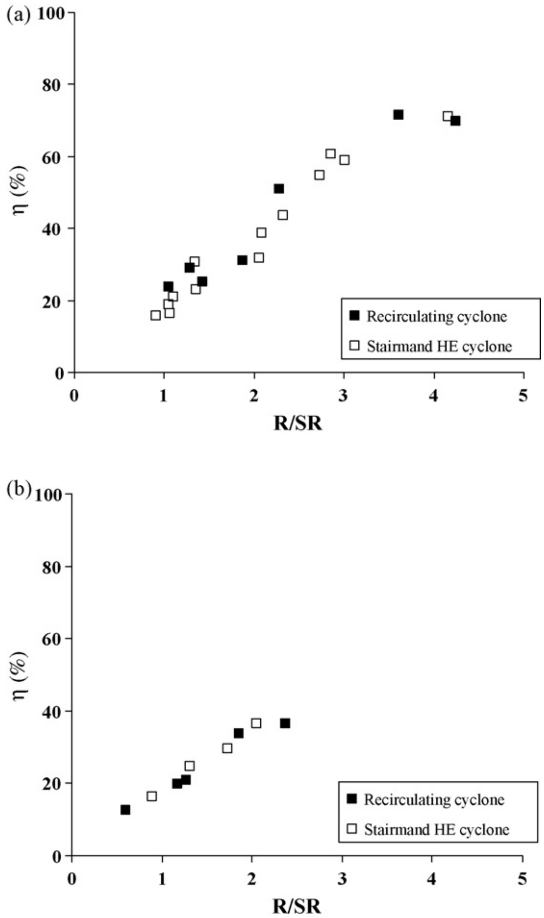

Fig. 3. (a) Experimental results for acid gas removal at HCl concentration 1.1×10^{-2} mol m^{-3} and (b) experimental results for acid gas removal at HCl concentration 1.8×10^{-2} mol m^{-3}.

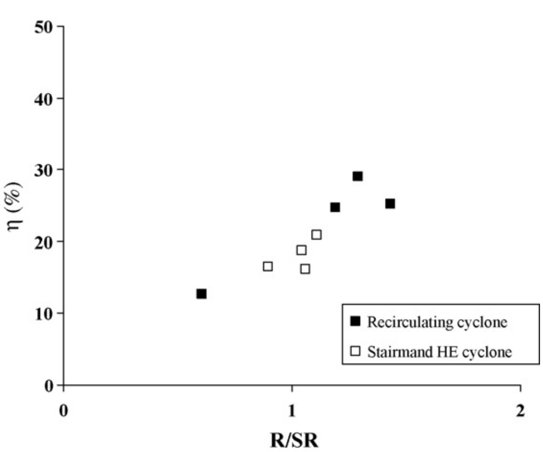

Fig. 4. Experimental results for acid gas removal at solids feed rate 1.8×10^{-7} kg s^{-1}.

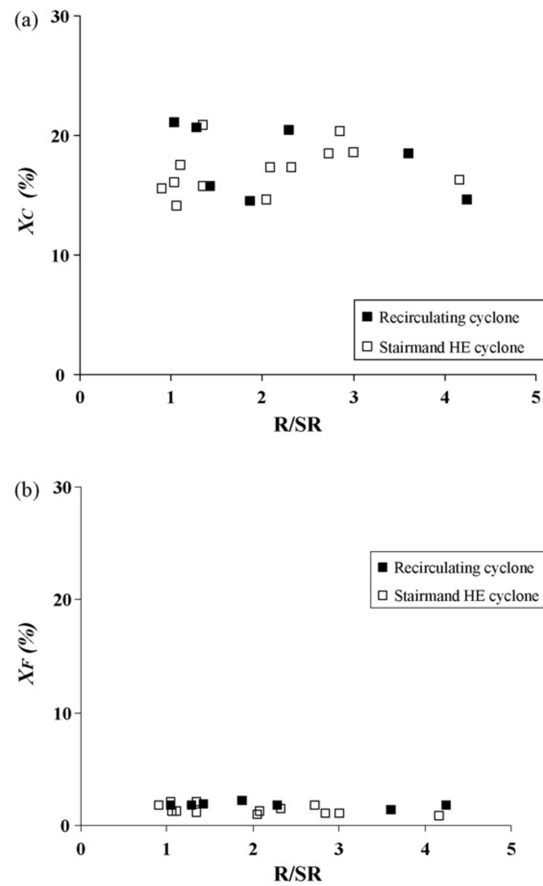

Fig. 5. (a) Experimental results for solids conversion in cyclone reactor (X_C) for HCl concentration 1.1×10^{-2} mol m^{-3}. (b) Experimental results for solids conversion in filter (X_F) for HCl concentration 1.1×10^{-2} mol m^{-3}.

diffusion limitations. Previous kinetic studies performed under similar operating conditions had already indicated that product layer diffusion would be the controlling step for this reaction [24].

For a fixed amount of solid reacted, the amount of gas removed is the same, but expressed in percentage of the total acid that enters the system, it will be lower for the higher values of acid concentration, viz. for the lower values of *R/*SR. Therefore the expectation that the recirculation of the partially reacted solid would increase the solids' conversion was not experimentally confirmed at the present conditions, but this is a direct consequence of the high removal efficiency (≈98%) of the tested recirculating cyclones.

The solid's conversion in the filter (X_F), also relative to the fresh feed to the system (see Fig. 5b), is extremely low, so one can conclude that HCl removal efficiency is mostly due to the cyclone system itself. Therefore it is confirmed, as in a previous work [9], that the cyclone is the main reaction chamber.

It should however be noted that these solid conversions are based on considering $CaCl_2$ as the sole reaction product, while recent experimental evidence [25–27] shows that Ca(OH)Cl is a likely candidate. This means that the solid conversions shown in Fig. 5a and b might be underestimated by as much as 100%.

4. Conclusions

The experimental results obtained at laboratory scale on the use of recirculating cyclones for the dry-scrubbing of HCl using fine slaked lime particles (2.13 μm median volume diameter) revealed very high efficiency (≈98%) for the solids' capture. As for the acid removal, the experimental results are highly dependent on the experimental conditions, but higher removal was obtained for high solids' loads and low HCl concentrations. The solids' reactant conversions obtained were on the average lower than 25%, revealing poor solids' usage. Therefore the expectation that the partial recirculation of the solid would increase the solids' conversion was not experimentally confirmed under the present conditions. These results indicate that the reaction is controlled by product-layer diffusion limitations, despite the short residence times in the cyclone.

The performance of a modified Stairmand HE reverse flow cyclone without recirculation was also tested for this process. The results show that for the particles' removal the recirculating cyclones have significant lower emissions with the additional advantage of having lower pressure drop values. As for the acid removal results, in the experimental conditions tested, no significant differences exist between the two systems. Since the residence time of the gas in the RS_VHE and in the Stairmand HE cyclones is very similar, these results indicate that the reactor vessel is essentially the cyclone and not the recirculation loop. The low solids' conversions found in the back filter at the exit of the cyclones corroborate this hypothesis.

As main conclusion of this work, it was proven at laboratory-scale that the use of recirculating cyclones for gas cleaning in a dry scrubbing process is efficient for the removal of acid gases and highly efficient for the capture of solid particles. This is a low cost technology which has the advantage of not requiring a post-reaction de-duster. The high consumption of solid reactant, necessary to achieve high acid removal efficiencies, may be offset by the simplicity and lower investment associated with the proposed cyclone reaction/collection chamber. As future works, it would be important to test if larger optimized RS_VHE reactors with correspondingly larger residence times and lower efficiency for solids' capture may increase the extent of the reaction, and therefore enable lower fresh solids' loads. Theoretical modeling of the experimental results is also important, which could open new possibilities to improve the solids' usage.

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