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Exergy destruction in ammonia scrubbers

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ABSTRACT

A theoretical ammonia scrubbing process by sulfuric acid solution is assessed with the concept of exergy. The exergy destruction of chemical neutralization is mainly (75–94%) due to changes in the chemical exergy of streams and thermal effects from the reaction while mixing effects have a limited contribution (6–25%). The minimum exergy consumption to remove one mole of ammonia chemically from an airstream could be two to eight times larger than the latent heat of evaporation of one mole of water depending on whether a concentrated (98% w/w) or a dilute (1% w/w) sulfuric acid solution is used. The exergy destruction per mole of ammonia scrubbed could be reduced by up to 75% when both the sulfuric acid solution and the ammonia at the inlet airstream are highly concentrated. The use of sulfuric acid concentration in the range of 10–50% w/w could lead to a very low exergy efficiency ($< 50\%$). The exergy efficiency could be improved up to ~87% by introducing an ammonia pre-concentration step right before the scrubbing process. The extension of system boundaries shows that the cumulative exergy loss rate for neutralizing a heavily loaded ammonia airstream with a flowrate of 1 $\mathrm{kg\,s^{-1}}$ ranges between 0.3–1.5 MW depending on the way sulfuric acid is produced. Consequently, an exergy-efficient scrubber design should balance between the minimization of the consumption of exergy-intensive resources, the minimization of the exergy destruction occurring in the separation process, and the maximization of output stream utilization.

1. Introduction

The levels of greenhouse gas (GHG) emissions worldwide increased by 35% between 1990 and 2010 reaching approximately 46 billion metric tons (United States Environmental Protecion Agency, 2014). This fact is pushing research agendas towards the design of sustainable emission abatement processes that focus on the efficient use of natural resources. A recent survey in Germany showed that energy efficiency improvements were the main reason for abating carbon dioxide emissions and with larger abatement volumes expected to occur by renewing existing production facilities between 2020–2030 (Heindl and Löschel, 2012).

An important GHG emission to consider is ammonia because its unconstraint release does not only lead to soil acidification, eutrophication, and air pollution but also to serious health problems (Melse and Ogink, 2005). Ammonia is a colourless, pungent, explosive, and corrosive gas noticeable above 50 ppm, which can irritate the eyes, nose, and throat, and it is toxic if inhaled in large quantities (Phillips, 1995). Moreover, ammonia can form small sized ($< 2.5\,\mu m$) particulate matter (known as PM2.5) together with other compounds, which penetrate the lungs and lead to serious respiration and cardiovascular problems (Gay, 2009). Therefore, human exposure to ammonia should not exceed 25 ppm for an eight hour shift and 35 ppm for 15 min (Health and Safety Executive (HSE), 2011). There has been a considerable reduction in ammonia emissions in Europe since 1990. However, since 2014 several EU Member States have been exceeding the set limits that were agreed in the Gothenburg protocol in 2010 (European Environment Agency, 2016). A number of industries (*e.g.* manufacturers of fertilizers and coke) and processes (*e.g.* fossil fuels combustion, livestock management, refrigeration) are ammonia emitters. Yet, the highest share of ammonia emissions in Europe (approximately 94%) is due to agricultural activities (*e.g.* manure storage and slurry spreading) (European Environment Agency, 2016). A recent study showed that ammonia emissions related to agriculture are currently so high that they even exceed the environmental impact of fossil fuel combustion emissions leading to the disruption of the natural nitrogen balance in Earth through wet deposition (nitrogen entering the cycle in the form of rain or snow) and dry deposition (direct deposition of ammonia molecules on Earth's surface) (Colorado State University, 2016). The concentration range of emitted ammonia can vary and is probably sector-dependent. For example, the average ammonia concentrations observed at three sites around Shanghai from March to June

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2014 varied between 0.0196 ppm ± 0.0082 ppm at the industrial site, 0.0104 ppm ± 0.005 ppm at the rural site, and 0.0054 ppm ± 0.0033 ppm at the urban site (Wang et al., 2015). In poultry production facilities the ammonia concentrations can be even higher, ranging between 1.8 and 13 ppm (Hayes et al., 2006), and which can be detected at an upwind distance of 2.8 km from the facilities (Jones et al., 2013), while in the composting process in industrial mushroom production a typical ammonia concentration range lies between 300 and 600 ppm (Den Ouden, 2012).

Technologies for the abatement of ammonia emissions have been described in literature (Busca and Pistarino, 2003) and a common way to reduce them efficiently is chemical neutralization by concentrated sulfuric acid in a process known as scrubbing (Melse and Ogink, 2005). Scrubber designs are quite diverse in their complexity and operation making it is difficult to capture all of them in one single categorization system. Usually, they are classified as wet or as dry scrubbers and their main differences as described in Joseph et al. (1984) are summarized below.

In wet scrubbers, the air is first forced either through a liquid pool (absorbers) or through a spray shower (packed or plate tower) of highly concentrated acid, and then it passes through a mist eliminator to collect any residual droplets. These systems are ideal for gases that contain both particles and gases. However, to achieve simultaneously high particle collection and gas separation efficiencies either the particles should be large and readily captured or the gases should be very soluble. The removal efficiency of wet scrubbers that are focused on particle collection is proportional to the power input. These scrubbers can be further categorized from the gas-side pressure drop of the system as low (< 12.7 cm), medium (12.7–38.1 cm), and high energy scrubbers (> 38.1 cm). Wet scrubbers have usually small space requirements and, therefore, capital costs. Consequently, they have large placement flexibility, they can handle high temperatures and very humid gases, and they eliminate the possibility of fire or explosion due to the use of water. Nevertheless, they also face a number of problems such as corrosion, high power (*i.e.* cost) requirements, the need for waste disposal systems to meet regulations, the difficulty to dewater and handle the produced sludge, and the potential impact on local meteorological conditions due to the saturated exhaust gases (Joseph et al., 1984).

Dry scrubbers are used to remove combustion acid gases by dry sorbent injection or by spray dryer absorption. In dry sorbent injection scrubbers an alkaline powder material reacts with the acid gases in the ductwork forming solid salts which are further removed in a particulate control device. The removal efficiency of these systems is limited but it can be improved by increasing the humidity of the combustion gas. Spray dryer absorbers operate in a similar manner but have higher removal efficiencies (~80%). In these systems hot acid gas comes in contact with a finely atomized alkaline slurry where the heat evaporates the water (Joseph et al., 1984).

All systems described above make use of large chemical driving forces to remove undesired gaseous components, which shows the importance of assessing the resource use efficiency of the scrubbing process. In this paper, the resource use efficiency of the scrubber is described with the concept of exergy. According to Dincer and Cengel (2001) exergy is the ability to produce work or, in other words, it is "*the maximum shaft work that could be done by the composite of the system and a specified reference environment that is assumed to be infinite, in equilibrium, and ultimately to enclose all other systems*". The operation of the scrubber requires the use (consumption) of natural resources which are expressed as the total exergy input and it constitutes of the chemical exergy (the sulfuric acid solution and ammonia loaded airstream) and the physical exergy (electricity and temperature of both inlet streams). Part of this input exergy is used, part is transiting, and part is destroyed to complete the process in finite time and thus compact equipment. Exergy destruction is the available work that is lost due to the irreversible operation of the system and it is related to the entropy generation proportionally through the absolute temperature of the environment (Bejan, 1982; Lucia, 2012). This proportionality relation is known as the Gouy-Stodola theorem (Bejan, 1982; Lucia, 2012) and it is important because it links the concept of exergy with the first and second law of thermodynamics providing, therefore, an objective method for quantifying the consumption of natural resource quality (Romero and Linares, 2014; Gaggioli and Reini, 2014). The quantification of exergy consumption and lost work can help in "unlocking" technological possibilities that are "hidden" or currently "unreachable" (Gaggioli, 2012).

The usefulness of exergy analysis in assessing the thermodynamic performance of systems has been demonstrated for gas separation processes (Ghorbani et al., 2012), for systems where the release of emissions is involved (Chitsaz et al., 2015; Flórez-Orrego and de Oliveira, 2016) and for the comparison of alternative environmentally-friendly hydrogen production routes (Koroneos and Rovas, 2012). In another example, the exergetic impact of producing harmless products from waste has been estimated for the production of ethanol from fossil fuels and from agricultural products (Dewulf et al., 2000). However, to the best of the authors knowledge exergy analysis has not been used so far to assess the thermodynamic performance of ammonia scrubbers.

In this paper, the conditions that would lead to the most exergy-efficient scrubbing operation are explored by assuming that ammonia is fully neutralized. The aim is to understand the theoretical influence of different concentrations of sulfuric acid solution and ammonia present in the inlet airstream on the thermodynamic performance of the scrubbing process.

2. Description of the scrubbing process

2.1. Reaction

A theoretical scrubbing process is studied at steady state and at atmospheric pressure where an ammonia laden airstream at 298 K with an absolute moisture of 8 g water per kg dry air (*i.e.* 41% relative humidity), and a flowrate of 1 kg/s is neutralized completely by a sulfuric acid solution leading to the formation of ammonium sulfate (Fig. 1). The variables studied are the concentration of ammonia (ppm) in the inlet airstream and the concentration of sulfuric acid solution (% w/w). The ammonia concentration at the inlet airstream is studied in a large range from too low to unrealistically high (*i.e.* reaching up to 10,000 ppm), and the sulfuric acid solution concentration are also varied from very low to ones that are typically used in the industry (*i.e.* 96% w/w) (Melse and Ogink, 2005).

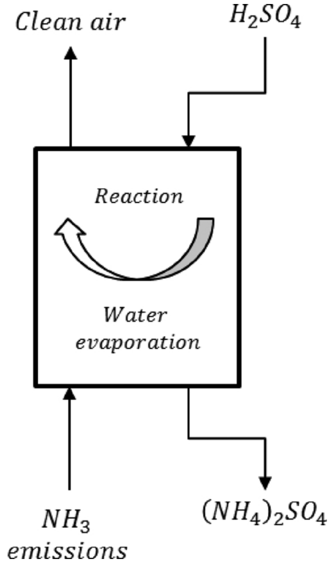

Fig. 1. Schematic representation of the ammonia scrubbing process.