



Extraction of tungsten and vanadium from spent selective catalytic reduction catalyst for stationary application by pressure leaching process

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ABSTRACT

This paper presents the direct extraction, which works by means of pressure leaching, of vanadium and tungsten from the spent SCR catalysts using a caustic soda. The experimental parameters were concentration of NaOH (*aq*), the particle size of the feedstock, the temperature, and the S/L ratio. The results of an assessment of the proposed method found that pressure leaching using a caustic soda can more effectively dissolve vanadium and tungsten from the spent SCR catalysts than atmospheric alkaline leaching. The dissolution efficiency of vanadium and tungsten tended to increase with an increase in the concentration of NaOH (*aq*), a higher temperature and a decrease in the particle size of the feedstock used. The S/L ratio, which affected the dissolution efficiency of vanadium and tungsten, differed depending on the concentration of NaOH (*aq*). The optimum conditions for the extraction of vanadium and tungsten by pressure leaching were as follows: concentration of NaOH (*aq*), 3 mol/L; temperature, 250 °C; particle size distribution; <150 μm; S/L ratio, 0.4. With these optimum conditions, the dissolution efficiency of vanadium and tungsten were found to be 91.5% and 87%, respectively. The established method for the direct extraction of vanadium and tungsten from spent SCR catalysts is effective and environmentally sound compared to atmospheric alkaline leaching.

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1. Introduction

Using a selective catalytic reduction (SCR) catalyst with ammonia has become the most effective means of reducing nitrogen oxide (NO_x) emissions from stationary segments and diesel vehicles around the world due to strengthened regulatory trends and emerging environmental issues (Forzatti, 2001; Spivey, 2002; Li et al., 2011). Currently, the most widely used SCR catalyst is the V₂O₅-WO₃/TiO₂ system due to its excellent catalytic properties, such as its wide operating temperature window, durability to

deactivation, and high conversion rate of NO_x (Li et al., 2011; Marshneva et al., 1995; Adrian et al., 2015). It is known that the composition of the SCR catalyst can differ depending on application (Adrian et al., 2015). In general, the composition of SCR catalysts used in stationary applications such as power plants and waste incineration is 0.5–1 wt% of V₂O₅ and 7 to 10 wt% of WO₃ (Adrian et al., 2015; Chen and Yang, 1992; Vuurman et al., 1991).

The typical chemical lifetime of a SCR catalyst is a few years, in general 3 years, depending on the composition and operation environment (Spivey, 2002). However, regeneration by a chemical and/or heat treatment can extend the lifetime, often doubling it (Yue et al., 2012). Despite prolonging the lifetime of the SCR catalyst via a regeneration process, the occurrence of completely abandoned spent SCR catalysts is unavoidable. Unfortunately, most spent SCR catalysts are treated as a specific type of waste and disposed of in landfills (Marafi and Stanislaus, 2008). This causes a considerable amount of waste of the secondary resources of vanadium and tungsten every year. Furthermore, it is known that spent SCR catalysts are environmentally unacceptable due to the

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toxicity of vanadium and tungsten to humans, plants, and animals (Imtiaz et al., 2015; Koutsospyros et al., 2006). Therefore, in terms of the industrial value of vanadium and tungsten (Gupta and Mukherjee, 1990; Moskalyk and Alfantazi, 2003) and reductions of pollutant emissions, their recovery for material recycling of spent SCR catalysts should not be disregarded. Nevertheless, recent studies on the recycling of SCR catalysts are typically oriented toward the regeneration by chemical washing to remove deposits on a catalyst. (Yuanjing et al., 2004; Khodayari and Ingemar Odenbrand, 2001; Xuesong et al., 2012). Although Wu et al. reported alkaline leaching of vanadium and tungsten, only classified conditions, means inadequate to practical operations, attain the high yield of vanadium and tungsten.

Dissolution of V_2O_5 and WO_3 as $NaVO_3$ (aq) and Na_2WO_4 (aq) in alkaline solution was known to be favor, thermodynamically (Gupta and Mukherjee, 1990; Huo et al., 2015). Following the known fact, direct leaching in alkaline solution was carried out in earlier study (Joung, 2012). According to the results for direct alkaline leaching, most results on the dissolution efficiency of vanadium and tungsten from spent SCR catalysts did not present the feasible outcomes (maximum dissolution efficiency: vanadium at 99% and tungsten at 60% under following conditions: NaOH, 5 mol/L; pulp density, 1.6%; particle size, <75 μm ; temperature, 50 °C). Although the dissolution of vanadium was completed, the dissolution efficiency of tungsten remains at a relatively low yield. Furthermore, the experimental condition for the maximum dissolution efficiency of vanadium and tungsten was not practical to apply for mass production due to the high concentration of aqueous NaOH, the low pulp density, and the difficulty in controlling the particle size. From this previous study, we concluded that the poor dissolution efficiency of vanadium and tungsten was caused by difficult diffusion control of the chemical reagent due to the nature of the SCR catalyst. According to the literature (Lee et al., 2007; Wachs, 2013; Anderson and Garcia, 2005), TiO_2 particles having tens of nanometers in size, is used as a supporting oxide to ensure the wide specific surface area (SSA) for catalytic performance. Therefore, the pulverized particles are not individual particles but are instead in an agglomerated form. Wachs (2013) reported that the formations of vanadium and tungsten were dispersed homogeneously and formed as an extremely thin layer termed the monolayer on the surfaces of ultra-fine TiO_2 particles.

Furthermore, it was reported that a superfluous amount of tungsten for a site at which a monolayer form usually exists with solvated state in the TiO_2 (Lee et al., 2007). Thus, pulverized spent SCR catalyst can be regarded as an agglomeration of ultra-fine TiO_2 particles supported on the surface with vanadium and tungsten. In particular, it was also assumed that diffusion control of tungsten remains due to its unique formation in SCR catalysts. Therefore, a chemically and physically favored leaching process such as pressure leaching is expected to dissolve vanadium and tungsten at high yield (>90%).

In the present study, we investigated the direct extraction of vanadium and tungsten from a spent SCR catalyst by pressure leaching for high yield using caustic soda. Based on this, it may possible to establish a metal recovery process and set the direction for the further separation and purification processing of tungsten, vanadium, and TiO_2 as new secondary sources.

2. Experimental

2.1. Materials

We obtained the honeycomb-type spent SCR catalyst module used in this study from the Samcheonpo thermoelectric power plant in South Korea as feedstock. All of the spent SCR catalysts

were physically cleaned by the removal of the attached and pore-plugged dust on the surface of the catalyst and then pulverized by a jaw crusher and grinding mill. The particle size distribution of the pulverized spent SCR catalyst was in the range of 1–200 μm mainly and the volume percent was 93.7% per total volume of particles as determined by a particle size distribution analysis (Mastersizer 2000; Malvern Instruments, USA). The pulverized spent SCR catalyst was dried at 80 °C in an oven during 24 h to remove residual moisture. The reduction in the weight by drying was 4.5% (± 0.5). An additional pre-treatment was not carried out before the experiment.

2.2. Pressure leaching and parameters

Pressure leaching was conducted using an autoclave reactor with a capacity of 1 L and with a copper seal to avoid vapor leaks. Figure 1 shows a schematic diagram of the autoclave reactor. It consists of SUS 316 to prevent alkali attacks and is designed to handle pressure levels as high as 100 bar. During the pressure leaching process, there were no leaks in any of the experiments. The reactor was equipped a cross-shaped four-blade impeller for agitation which was located at the bottom of the reactor.

The alkaline agent used for pressure leaching was sodium hydroxide (NaOH) (Junsei Chemicals, 97%, Japan). The parameters manipulated in this experiment were the particle size distribution (<44 μm to 1 mm), the NaOH (aq) concentration (1–4 mol/L), the

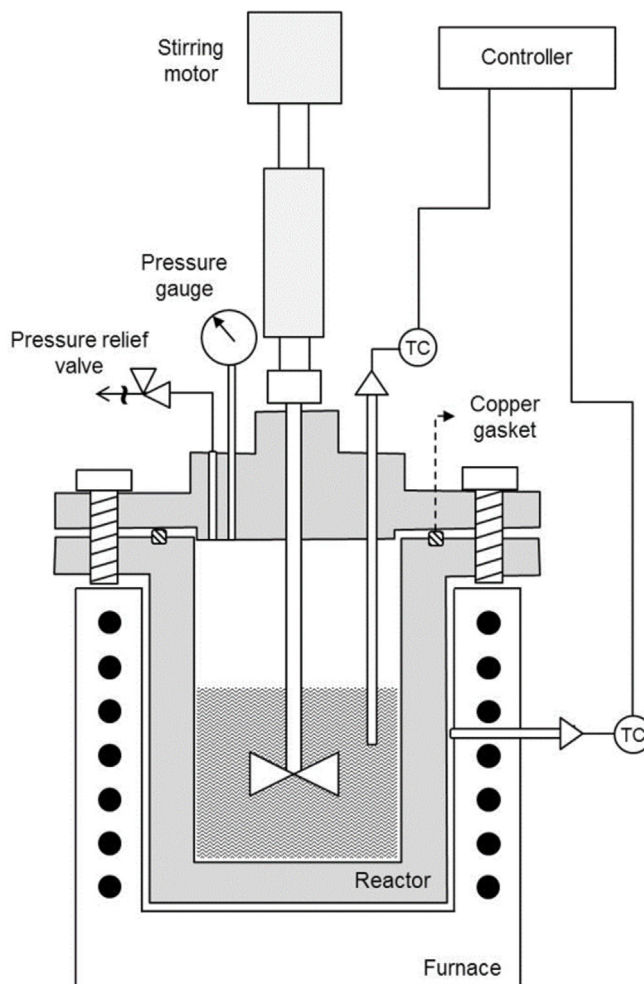


Fig. 1. Schematics of autoclave reactor for the pressure caustic soda leaching.

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