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Removing degradation products and reducing oxidation in aqueous piperazine with activated carbon

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Abstract

Piperazine (PZ) is a promising second generation amine scrubbing solvent for CO₂ capture due to its high reaction rate, capacity, and resistance to thermal and oxidative degradation. However, the finite rate of PZ oxidation adds the costs of reclaiming, solvent make-up, and waste treating as PZ oxidizes to piperazinol, piperazinone, ethylenediamine (EDA), NH₃, and heat stable salts such as oxalate and formate.

From February 2018 to June 2019, 5 m PZ was used for 4000 hours in a 0.5 MW pilot plant at the National Carbon Capture Center (NCCC) in Wilsonville, AL. A slip stream equivalent to 0.5 MW from a coal-fired power plant was used as the flue gas for treatment. The Piperazine Advanced Stripper (PZASTM) was used as the stripper configuration, and the pilot plant was mainly operated under a stripper temperature of 150 °C and a 90% capture rate. An FTIR was used to continuously monitor the NH₃ concentration in the water wash outlet, and sample solvents were collected daily to analyze for the concentration of the degradation products in the solvent. After 3600 hrs of operation, the solvent was fed through a bed of activated carbon to remove impurities and test its effect on amine degradation. As shown in Figure 1, The carbon treating had a significant effect on the color of the solvent, indicating that some degradation products had been removed. UV-Vis analysis has been performed to estimate the concentration of colored products.



Figure 1. Solvent Color Change after the Usage of Carbon Bed

Systematically changing peaks were observed 320 nm and 538 nm in the UV-Vis spectrum with NCCC samples. The peak intensity followed Beer's Law, and was used to quantify the accumulation of these degradation products. As shown in Figure 2, both peaks increased steadily before 3600 hrs, and decreased immediately after the activation of carbon bed. The peak at 320 nm is related to a complex formed from Fe^{3+} and PZ or the degradation products. More work is planned to identify the peak at 538 nm.

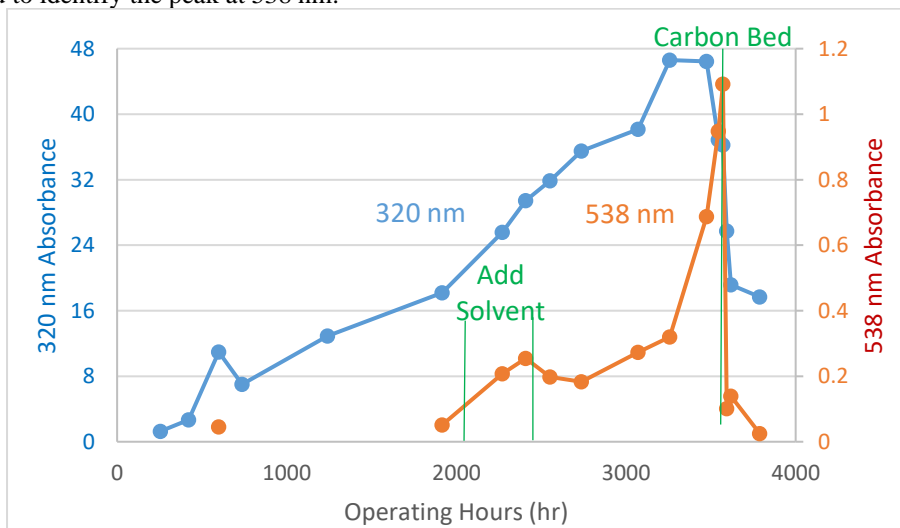


Figure 2. Absorbance of Two Characteristic UV-Vis Peaks of NCCC 2018-2019 Campaign Samples

Besides reducing the absorbance of the solvent, the carbon treating also reduced the NH_3 production rate, an indicator of the PZ oxidation rate. As shown in Figure 3, the NH_3 production rate increased steadily before 3600 hrs, which was consistent with previous observations that PZ oxidation increases with solvent degradation. After 3600 hrs, the NH_3 rate started to decrease, indicating that the solvent oxidation was mitigated, most probably due to the removal of degradation products that catalyze the oxidation.

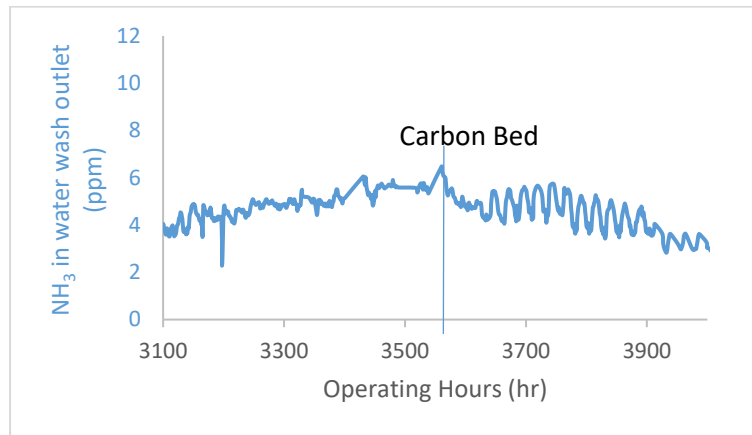


Figure 3. NH₃ Concentration Change in Water Wash Outlet in NCCC 2018-2019 Campaign

More work will be performed to identify the degradation products removed by the carbon. Bench-scale experiments will be performed to duplicate the effects. The capacity of the carbon bed will be tested, and multiple types of carbon will be used to find the carbon that can selectively remove the catalytic degradation products.

Keywords: Piperazine oxidation; Carbon adsorption; Pilot plant
