Process Simulation of Batch Thermochemical Treatment and Off-Gas Control System for Spent Activated Carbon

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1. Introduction

Spent activated carbon (AC) generated during the operation and decommissioning of pressurized heavy water reactors (PHWRs) may contain C-14, H-3, moisture, triethylenediamine (TEDA), and other adsorbed species [1, 2]. Thermochemical treatment can remove these species from activated carbon, but resulting off-gas requires additional treatment before discharge or immobilization.

A demonstration-scale treatment system with batch consists of high-temperature vacuum treatment, TEDA pyrolysis, catalytic oxidation, selective catalytic reduction (SCR) of NOx, and CO_2 capture followed by lime-based mineralization [3]. Process simulation was applied to establish material and energy balances and to estimate gas-treatment loads, oxidant and regent requirements, and operating conditions for each unit. Because the radionuclides are present at trace levels, C-14, and H-3 bearing species were represented by their stable chemical counterparts, such as CO_2, H_2, and H_2O, while their radionuclide fractions are considered separately.

For the present design study, the batch off-gas represented as a continuous stream under a conservative maximum-generation condition. The developed model will subsequently be refined by comparing predicted stream flow rates and compositions with off-gas data measured from the demonstration unit. This approach provides an engineering basis for evaluating the integrated treatment process and supporting further system design and scale-up.

2. Process Description and Modeling Methodology

2.1 Process Configuration and Design Basis

The process simulation was initiated from the off-gas generated during thermochemical treatment rather than by modeling the vacuum furnace itself. TEDA-impregnated AC was heated under controlled conditions, and the major gaseous species released at elevated temperatures were identified by FTIR Gas analysis. The resulting gas composition, together with the measured off-gas flow rate, was used to define the feed condition for the downstream process simulation.

The off-gas treatment process consists of a pyrolyzer, oxidizer, selective catalytic reduction (SCR) unit, and CO_2 capture system followed by lime-based mineralization. TEDA-derived vapor and other organic species are decomposed in the pyrolyzer, and combustible products are subsequently oxidized to CO_2 and H_2O. Nitrogen-containing species generated during thermal decomposition and oxidation are treated in the SCR unit, while CO_2 is captured and converted to $CaCO_3$ in the final treatment stage.

Although off-gas generation from the thermochemical treatment unit varies during batch operation, the maximum measured gas-generation condition was assumed to persist continuously in the process model. This conservative assumption was adopted to evaluate the maximum operating load of the downstream treatment units. Radioactive species were not explicitly modeled. Instead, stable chemical counterparts such as CO_2, H_2, and H_2O were used for the material and energy balances. Radionuclide fractions will be applied separately in subsequent evaluations.

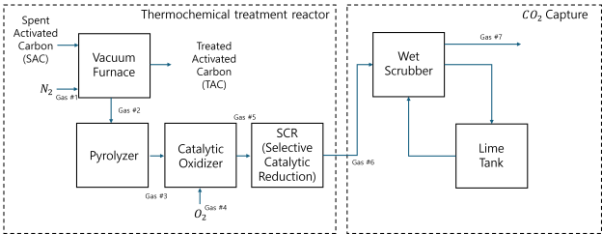


Fig. 1. Schematic diagram of the thermochemical treatment and off-gas control system.

2.2 Process Simulation Model

The downstream process shown in Fig. 1 was represented by a steady-state process model was developed using the design basis off-gas defined in Section 2.1. Major gas-phase components identified from thermochemical treatment were included as conventional species, while radioactive isotopes were represented by their stable chemical counterparts. The initial model was simplified to establish material and energy balances and to estimate the operating loads of the downstream treatment units.

The pyrolyzer was represented by a yield-based reactor using the estimated distribution of TEDA-derived decomposition products. Combustible species, including H_2, CO and hydrocarbons, were oxidized in a stoichiometric reactor with an external oxidant stream. The SCR unit was modeled using specified NOx conversion and reducing-agent consumption. CO_2

capture was initially represented by a separator with a prescribed capture efficiency, and the corresponding $Ca(OH)_2$ requirement and $CaCO_3$ production were calculated from the carbonation stoichiometry.

The IDEAL property method was applied to the initial gas-phase model because the process was primarily operated at low pressure. The calculated stream flow rates, compositions, heat duties, and reagent requirements will be compared with measurements from the demonstration unit to refine the model in subsequent work.

3. Results and Discussion

For the 50 kg (approximately 100 L) batch basis, the thermochemical treatment calculation gave a furnace off-gas flow of 2.724 Nm^3/h. The stream contained N_2, H_2O, O_2, CO_2, TEDA, and trace Ar, with TEDA accounting for approximately 0.242 Nm^3/h. This off-gas was used as the design-basis feed to the downstream treatment process. In the pyrolyzer, TEDA was converted into lower-molecular-weight species, including CO, CH_4, C_2-C_3 hydrocarbons, NH_3, CO_2, and H_2O. The resulting pyrolyzer off-gas was calculated to be 3.401 Nm_3/h.

The pyrolysis products were subsequently treated in the catalytic oxidizer. Under the assumed complete oxidation condition with excess O_2, combustible carbon species were converted mainly to CO_2 and H_2O. The calculated oxidizer outlet contained 1.451 Nm^3/h of CO_2 and 2.030 Nm^3/h of H_2O, together with excess O_2 and nitrogen-containing products. NO and NO_2 flows were estimated to be 0.459 and 0.024 Nm^3/h, respectively. These values represent the design basis for the downstream SCR unit and are dependent on the assumed conversion of TEDA-derived nitrogen during oxidation.

A 32.5 wt% urea solution was applied as the reducing agent for SCR. Under the specified NOx reduction condition, the required solution feed was approximately 1.94 kg/h, corresponding to 0.68 kg/h of urea. The calculated residual NO and NO_2 flows were approximately 0.00046 and 0.00100 Nm^3/h, indicating that the outlet condition was consistent with the specified NOx removal target.

After SCR, the gas was introduced to the wet CO_2 capture system. The CO_2 flow decreased from approximately 1.704 to 0.085 Nm^3/h, corresponding to an estimated capture of about 95% under the assumed operating condition. The captured CO_2 was converted to carbonate in the NaOH-based scrubber and subsequently treated with $Ca(OH)_2$ in the lime tank. The calculated $Ca(OH)_2$ requirement was 5.35 kg/h, and approximately 7.45 kg/h of $CaCO_3$ was produced. The final gas flow to the stack was estimated to be approximately 4.98 Nm^3/h.

The present results provide preliminary material-flow and reagent-load estimates for the integrated off-gas treatment system under a conservative continuous operating condition. Because the batch off-gas was represented by a maximum-generation steady-state stream, the calculated flow rates and reagent demands should be interpreted as design loads rather than batch-average consumption. In addition, the pyrolysis product distribution and subsequent NOx formation are currently based on assumed reaction behavior. These parameters will be refined by comparison with measured off-gas compositions and flow rates from the demonstration-scale treatment unit.

4. Conclusions

A preliminary steady-state process model was developed for the off-gas treatment system associated with a 50 kg (approximately 100 L) batch thermochemical treatment of spent activated carbon. Using the maximum off-gas generation condition as a conservative continuous design basis, the furnace off-gas flow was estimated to be 2.724 Nm^3/h and was subsequently treated through pyrolysis, catalytic oxidation, SCR, and CO_2 capture. Under the specified reaction and removal conditions, the model quantified the required SCR reagent and CO_2 treatment loads. With an assumed CO_2 capture efficiency of approximately 95%, the calculated $Ca(OH)_2$ requirement and $CaCO_3$ production were 5.35 and 7.45 kg/h, respectively. These results provide a preliminary engineering basis for evaluating downstream equipment loads and operating requirements. The model will be further refined by comparison with measured off-gas flow rates and compositions from the demonstration-scale treatment system.

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