



Optimization of the Catalytic Oxidation of Carbon monoxide by Response Surface Method

Mahnaz Pourkhalil^{1*}, Saeideh Tasharoffi², Nosrat Izadi¹ and Ensieh Ganji Babakhani³

1. Nanotechnology Research Center, Research Institute of Petroleum Industry (RIPI), Tehran, Iran

2. Biotechnology and Environment Department, Research and Development of Energy and Environment Research Center, Research Institute of Petroleum Industry (RIPI), Tehran, Iran

3. Gas Refining Technologies Division, Research Institute of Petroleum Industry, Tehran, Iran

pourkhalilm@ripi.ir

DOI: 10.22078/pr.2018.3439.2573

Received: August/25/2018

Accepted: November/19/2018

INTRODUCTION

Carbon monoxide gas from combustion of hydrocarbon fuels is one of the most important sources of environmental pollution [1,2]. One of the most effective and economical ways to remove carbon monoxide emissions from chimneys and vehicles is the catalytic oxidation process of carbon monoxide to carbon dioxide. It has been reported by Kim et al [3-5] reported that in the process of carbon monoxide oxidation, cerium oxide is a suitable enhancer for iron, cobalt and manganese catalysts due to the provision of active oxygenates and the switching between Ce³⁺ and Ce⁴⁺ is [6]. In the present study, a 15 wt.% cobalt-cerium oxide catalyst with a weight

ratio of 1.5 cobalt-cerium metal based on multi-walled carbon nanotubes was fabricated under the coherence method and investigated the parameters affecting the operating conditions on the conversion percentage. The carbon monoxide gas is converted to carbon dioxide, and the optimum amount of these parameters is calculated for maximum conversion of this pollutant. To investigate the effect of operating variables on reaction temperature, carbon monoxide pollutant concentration, oxygen gas concentration in the feed, and catalyst consumption rate on carbon monoxide gas conversion rate, response surface methodology (RSM) at five levels (+ 2, +1, 0, -1, -2) was used.

EXPERIMENTAL PROCEDURE

CATALYST PREPARATION

Cobalt-cerium oxide catalyst with a weight ratio of 1.5 to metal-cerium was fabricated based on multi-walled carbon nanotubes. The certain amounts of one-molar solutions of 6-cobalt nitrate and 6-cerium nitrate salts were mixed together under extreme stirring at 70 °C, then oxygenated carbon nanotubes (OMWNTs) were added to solution as desired, then resulting solution was sonicated for 30 minutes at 60 °C. The pH of the solution was adjusted to 10, and the solution was heated at 70 °C for one hour, and it was finally washed with distilled water until pH value reaches to 7. It dried in a thermal oven for 12 hours at 100 °C. The catalyst calcination performed at 550 °C in a horizontal furnace under argon for three hours.

CATALYTIC ACTIVITY TEST

In order to study and optimize the parameters affecting the catalytic performance of the carbon monoxide contaminant removal process from 15% wt.% cobalt-cerium oxide catalyst based on 1.5% metal to cobalt-cerium weight ratio, the process was evaluated in a reactor with quartz reactor. The reaction temperature which was controlled by a K-type thermocouple was inserted directly into the catalyst bed. The reactor temperature between 100-300 °C uses a thermal furnace equipped with a control system. In each experiment, different amounts of powder catalyst with 60-100 mesh were used depending on the amount of space speed considered as an operational variable.

RESULTS AND DISCUSSION PHASE COMPOSITION ANALYSIS

Response surface methodology (RSM) employed

for optimization of the important parameters in the catalytic oxidation carbon monoxide. The levels were employed for the different factors (temperature, oxygen concentration, carbon monoxide concentration in the feed mixture and the spatial velocity on the conversion rate of carbon monoxide gas on the catalyst), according to CCD. The basis for selecting the minimum and maximum values of the operational variables of carbon monoxide and oxygen was determined according to the conditions of the flue gas emissions of many refineries and power plants.

CATALYTIC PERFORMANCES

Increasing the temperature from 100 to 200 °C increased the conversion rate in the catalytic oxidation process, which is in line with the results of other researchers (Figure 1a) [7]. By increasing the temperature to 300 °C, the conversion rate decreased slightly, which can be attributed to the burning of the carbon base in the presence of oxygen (5.25% vol) at temperatures above 200 °C. The positive effect of increasing oxygen concentration from 0.5 to 5.25 has been shown to be due to an increase in the rate of carbon monoxide oxidation in an oxygen-rich environment (Fig. 1b) [8]. On the other hand, the inert concentration of carbon monoxide in the range of 15-60 ppm indicates that the presence of oxygen at a much higher level than required for carbon monoxide oxidation does not affect the catalyst surface (Figure 1c) [8].

The evaluation of Fig. 5d shows that with an increase in spatial velocity, the conversion was constant over the range 10000-30000 h⁻¹ but at higher values than 30000 h⁻¹ due to the reduction of the time required for carbon monoxide adsorption on the surface.

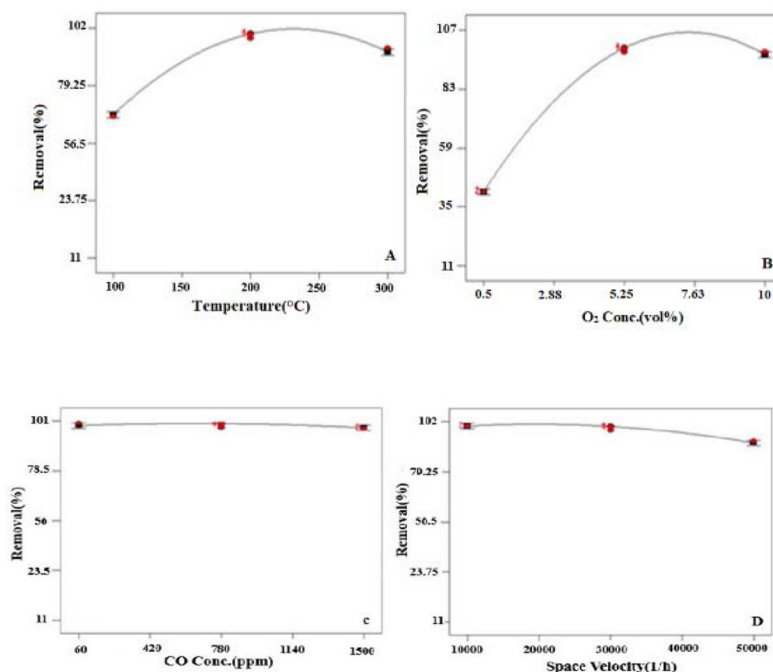


Figure 1: Two-dimensional curves of the effect of operating variables: (a) temperature, (b) oxygen concentration, (c) carbon monoxide concentration, (d) spatial velocity, on process conversion percentage.

The catalyst and related reactions have slightly decreased the conversion rate of the removal process. According to the two-dimensional curves of Fig. 1d, it is shown that oxygen concentration up to 5% vol and reaction temperature in the range of 100-100 °C are the most effective parameters in the catalytic process of CO₂ removal.

gas concentration variables in the feed on the conversion rate were investigated in Fig. 2a. As the oxygen concentration increased with increasing temperature, the higher conversion rate was, i.e. the interaction of these two independent variables on the response variable had a positive effect. At low oxygen concentrations, increasing temperature has less positive effect. The lowest and highest catalytic activity were at 0.5% vol at concentrations of 100 °C at 10% vol concentration in the temperature range of -200 °C respectively. The interaction of the two variables of temperature and spatial velocity on the conversion rate of carbon monoxide evaluated in Fig. 2b. In the operating range, spatial velocity

from 10000 to 130000 h observed with increasing temperature from 100 to 200 °C. As the rate of space velocity increases, the positive effect of temperature increase on the catalytic recovery process reduced, so that at the space velocity of h⁻¹ 10000 the temperature range of 200-200 °C has the highest conversion rate

The interaction of temperature and carbon monoxide concentration variables is ineffective in the catalytic activity process, as shown in Fig. 2c, 300 °C is the same.

OPTIMIZATION OF FACTORS

The optimal amount of factors affecting the conversion percentage response variable (R) can be calculated using the desirability Function. Based on the limits for the four independent variables and the maximum response variable R of 100%, the optimal operating conditions for the operating variables were obtained in accordance with Fig. 3.

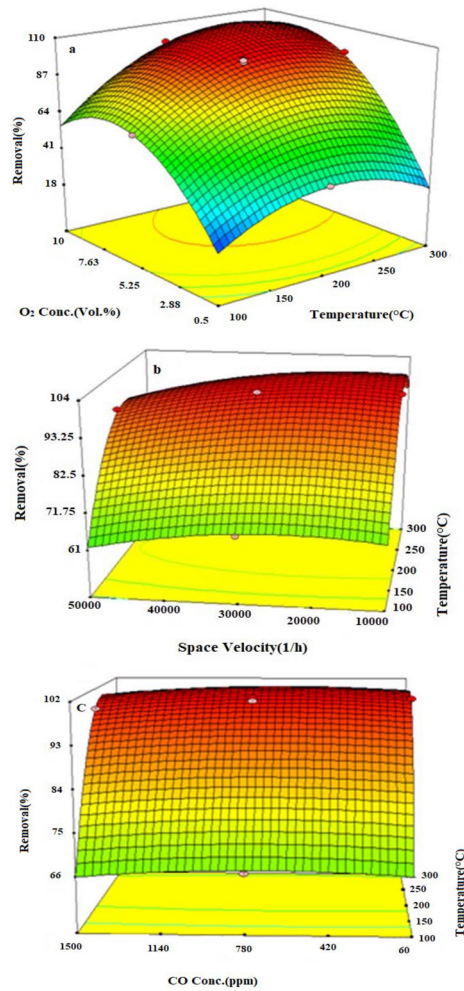


Figure 2: Three-dimensional curve of interaction of (a) temperature and oxygen concentration (b) space temperature and velocity (c) carbon monoxide concentration and temperature, on process conversion percentage.

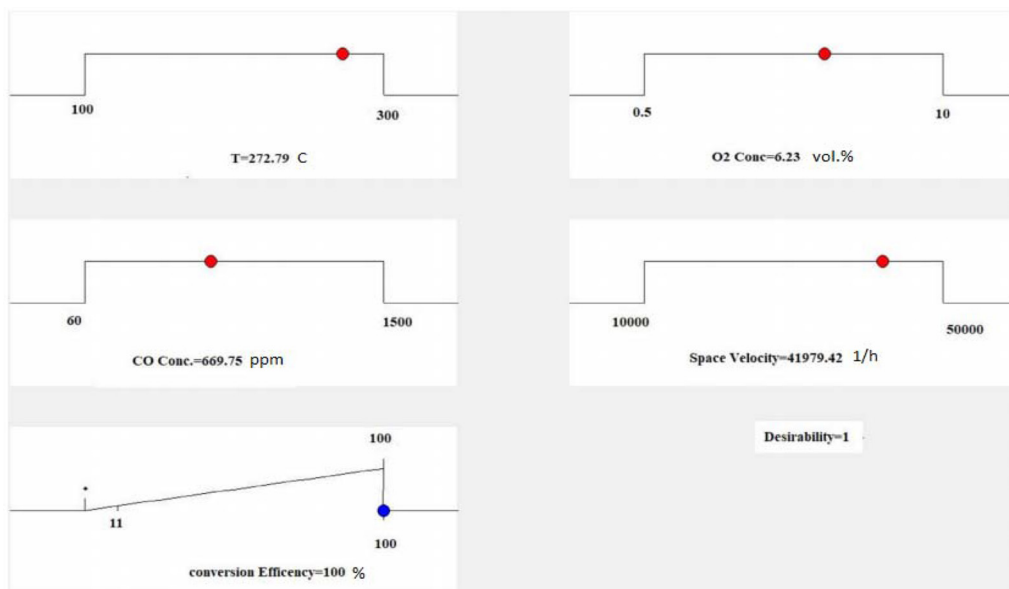


Figure 3: Optimal operating conditions based on desirability function.

CONCLUSIONS

In this study, the modeling and optimizing the effective operating parameters in the catalytic oxidation process of carbon monoxide gas (reaction temperature, spatial velocity and concentration of feed gases) were evaluated using surface response method. Temperature, oxygen concentration and spatial velocity as factors affecting the conversion rate and carbon monoxide concentration were not effective on catalytic conversion. In the interaction of independent factors, variables of oxygen concentration and temperature as well as temperature and spatial velocity were the factors affecting the conversion rate. It should be noted that temperature and oxygen concentration had the most influence on the conversion percentage in the variables under study.

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