



Enhancement of the efficiency of in situ combustion technique for heavy-oil recovery by application of nickel ions

Y. Hamed Shokrlu^a, Y. Maham^b, X. Tan^b, T. Babadagli^{a,*}, M. Gray^b

^a Department of Civil and Environmental Engineering, School of Petroleum Engineering, University of Alberta, Edmonton, Alberta, Canada

^b Department of Chemical and Materials Engineering, University of Alberta, Edmonton, Alberta, Canada

HIGHLIGHTS

- ▶ Nano nickel particles were tested as catalyst in high and low temperature oxidation.
- ▶ The effect of nickel ions on combustion increases by increasing temperature.
- ▶ A significant catalytic effect on low temperature reactions was observed.
- ▶ Findings are of significant importance for improvement of in situ combustion oil recovery.
- ▶ Low temperature oxidation increases oil viscosity. Nickel reduce this effect considerably.

ARTICLE INFO

Article history:

Received 18 February 2012

Received in revised form 3 July 2012

Accepted 5 July 2012

Available online 25 July 2012

Keywords:

Heavy-oil recovery

In situ combustion

Nano metal particles

Low temperature oxidation

ABSTRACT

Thermogravimetry coupled to Fourier Transform Infrared Spectroscopy (TGA–FTIR) was used to study the effect of nickel ions on heavy oil combustion. Mainly, the low temperature oxidation (LTO) process was studied by conducting isothermal α_0 experiments at three different temperatures: 200, 250 and 300 °C. The kinetics of the reactions was analyzed using the TG results. Nickel ions decreased the activation energy of the process from 16.9 kJ/mol to 10.9 kJ/mol. The FTIR spectra of the evolved gases were helpful in understanding the effect of nickel ions on the quality of LTO products. TGA–FTIR analysis revealed a production of different oxygenated compounds, such as phenols, carboxylic acids, sulfones and side products such as sulfur dioxide, during LTO. According to this analysis, using nickel ions decreased the concentration of oxygenated compounds in the evolved gases, and increased the concentration of carbon dioxide and water. It also decreased the concentration of sulfur dioxide. This effect was more promising at higher temperatures. This effect of nickel ions on LTO reactions can be of great interest in improving the efficiency of the in situ combustion technique used for heavy-oil recovery.

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1. Introduction

Heating is essential in in situ production of heavy-oil or bitumen. Steam-based thermal recovery techniques, such as steam assisted gravity drainage (SAGD), cyclic steam stimulation (CSS) and their modifications, are the most common ones. Despite its technical success, it is an energy intensive process due to the cost of steam generation. The in situ combustion technique is an alternative to heat generation at surface. This method is economically more viable compared to steam based techniques but it has its own technical challenges.

The main mechanism of the in situ combustion technique is the generation of heat via exothermic oxidation reactions which produce coke as the combustion fuel. Therefore, efficient oxidation

of the oil is required for successful combustion. Since there are different zones in the reservoir with different temperatures, different types of oxidation are expected. Dabbous and Fulton [1] classified the hydrocarbon oxidation reactions as: (1) high temperature oxidation (HTO) which occurs at temperatures above 300 °C and (2) low temperature oxidation (LTO) which corresponds to temperatures lower than 300 °C. HTO reactions mostly consist of carbon–hydrogen bond breakage and the production of water and carbon dioxide [2]. These exothermic reactions provide the energy required for continuous combustion. These reactions result in the partial upgrading of oil. On the other hand, LTO reactions produce oxygenated hydrocarbons such as carboxylic acids and sulfones [3]. Therefore, they adversely increase the viscosity of oil which can affect the displacement efficiency and recovery factor.

The catalytic effect of transition metal compounds on steam stimulation and in situ combustion processes have been investigated by several authors. In the case of steam stimulation, it is

* Corresponding author.

E-mail address: tayfun@ualberta.ca (T. Babadagli).

Nomenclature

| | | | |
|-------------|--|-------|---|
| α_0 | component fraction at $t = 0$ | k | reaction rate constant, min^{-1} for first-order reactions |
| $f(\alpha)$ | component fraction function | n | reaction order |
| α | component fraction (wt.%) | R | universal gas constant (J/mol K) |
| A | pre-exponential factor in Arrhenius equation (min^{-1}) | T | temperature ($^{\circ}\text{C}$) |
| E | activation energy (kJ/mol) | t | time (min) |
| $h(p)$ | pressure function | t_0 | initial time (min) |

shown experimentally that metal species catalyze the breakage of carbon–sulfur bonds existing in the organosulfur compounds. In other words, metal species catalyze the hydrodesulfurization and aquathermolysis reactions [4–7], which increase the upgrading effect of the steam-based techniques. He et al. [2] studied the catalytic effect of metallic salt additives on the efficiency of in situ combustion at the temperature range of HTO reactions. They observed that metallic salt additives (Fe^{3+}) enhanced fuel deposition, reduced the activation energy and produced more complete combustion. However, the exact mechanism of the catalytic reactions is still unknown. Also, the effect of metallic additives on LTO reactions was not studied. Later, Ramirez–Garnica et al. [8] investigated the effect of a nickel ionic solution on the recovery factor of the in situ recovery technique through combustion tube experiments. Again, the experiments were run at the temperature range of the HTO reactions. Ramirez–Garnica et al. observed that the nickel catalyst improved oil upgrading during in situ combustion and increased the recovery factor [8]. The catalytic effect of metal species on heavy oil upgrading was also studied by other researchers in different processes [9–12].

In this paper, the effect of a nickel ionic solution on the LTO reactions is studied using the TGA–FTIR system. LTO reactions can create low mobility zones which reduce the displacement efficiency and recovery. It was observed that metal species catalyze HTO reactions [2,8,13], but their effect on LTO reactions is unknown. The production of oxygenated compounds in heavy oil can drastically influence the oil viscosity and reduce the process efficiency. Metals may also coordinate to some big asphaltenic molecules and produce more complex structures, which adversely affect the oil viscosity. Therefore, the effect of metal species on oil properties at a low temperature needs to be investigated. Metals accelerate the HTO reactions by destroying the antioxidants which exist in oil [14,15] and we expect the same kind of catalytic reactions to occur in the temperature range of LTO reactions.

Thermogravimetric analysis (TGA) is a useful tool to study the kinetics of in situ combustion. The TGA was applied to study the kinetics of HTO reactions earlier [13,16,17]. Kok and Bagci [13] used simultaneous TG–DTA and reaction cell experiments to study the combustion of light crude oil in the presence of metallic additives. They observed that magnesium chloride and copper chloride lowered the reaction interval and peak temperature of the HTO reactions. The additives also decreased the activation energy of the crude oil. Ambalae et al. [17] applied TG experiments to study the pyrolysis and combustion behavior of heavy oil and its asphaltenes. Kok [16] investigated the effect of reservoir rock composition on combustion kinetics using TG/DTA. From the TGA data, they cal-

culated the activation energy of the LTO and HTO reactions. In the case of a limestone matrix, the activation energy of LTO (between 10–20 kJ mol^{-1}) reactions was about ten times smaller than the activation energy of HTO reactions (between 90–120 kJ mol^{-1}). In the case of the sandstone matrix, the activation energy of HTO reactions decreased to values between 50–70 kJ mol^{-1} while the activation energy of LTO reactions did not change significantly [16]. These observations confirm the importance of the catalyst type on combustion kinetics.

In the present paper, a simultaneous TGA–FTIR (Fourier Transform Infrared Spectroscopy) system was used to study the kinetics of heavy oil combustion at low temperatures (LTO region), as well as the effect of a nickel ionic solution on this process. FTIR was used to analyze the evolved gases. The amount and type of the evolved gases detected by the FTIR system are the important parameters, which were used as the analysis parameters for studying the mechanisms of the catalytic reactions.

2. Experimental

The equipment utilized for this study consisted of a Cahn TGA interface to a Nicolet 6700 Fourier Transform Infrared Spectrometer (Thermo Scientific). The evolved gases from thermogravimetric measurements were transported by a vacuum pump at constant flow rate, to a gas cell installed in the FTIR spectrometer through a transfer line. The transfer line was kept at 200 $^{\circ}\text{C}$ using a heating tape to prevent condensation of the released gases. The results obtained from the TGA–FTIR are presented as follows: (1) a Gram–Schmidt plot, which shows related information with the total IR absorbance of the evolved components in the whole spectral range; (2) a three-dimensional spectra (as a stack plot) of the evolved gases; and (3) the IR spectra obtained at the maximum evolution rate for each decomposition stage.

Heavy oil with 14.7 $^{\circ}$ API was used in all of the experiments. The physical and chemical properties of the oil sample are presented in Table 1. 0.1 M of nickel chloride solution was prepared and mixed with heavy oil with a 50–50 wt.% ratio. The suspension was mixed for 15 min at 6000 rpm using a Polytron PT6100 mixer. Around 20 mg of sample was put into a TG pan. The experiment started by heating the system at a 50 $^{\circ}\text{C}/\text{min}$ heating rate to reach the desired isothermal temperature. Then, the temperature was kept constant for a specific period of time. Air was used as the purge gas. Its flow rate was 40 cc/min in all of the experiments. The experiments were run at three different temperatures: 200, 250 and 300 $^{\circ}\text{C}$.

Table 1
Properties of the oil sample.

| Physical properties at 25 $^{\circ}\text{C}$ | | | SARA analysis (wt.%) | | | | Elemental analysis (wt.%) | | | |
|--|---------------|------|----------------------|-----------|--------|-------------|---------------------------|-------|------|------|
| Density (gr/cc) | Viscosity, cp | RI | Saturates | Aromatics | Resins | Asphaltenes | C | H | N | S |
| 0.965 (gr/cc) | 8500 cp | 1.56 | 36 | 37 | 13 | 14 | 80.47 | 10.63 | 0.43 | 4.09 |

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