Fuel Processing Technology 139 (2015) 108–116

Contents lists available at ScienceDirect

Fuel Processing Technology

journal homepage: www.elsevier.com/locate/fuproc

Research article

Modeling study of oil shale pyrolysis in rotary drum reactor by solid heat carrier

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ARTICLE INFO

Article history:
Received 30 April 2015
Received in revised form 20 July 2015
Accepted 1 August 2015
Available online 11 August 2015

Keywords:
Oil shale pyrolysis
Mathematic model
Galoter process
Rotary drum reactor
Heat and mass transfer
Decomposition kinetics

ABSTRACT

The mathematic model of oil shale thermal decomposition (pyrolysis) was constructed on the base of analysis of available experimental data. The model was applied to the engineering procedure that simulates the Galoter process, namely, pyrolysis of oil shale in a horizontal rotary drum reactor in contact with fine-grained solid heat carrier (hot ash). The model includes the kinetics of organic matter (kerogen) decomposition, the processes of heat and mass transfer inside of single oil shale particle, polydispersity of the particles, their fragmentation, and the secondary chemical reactions such as the carbonization of the emitted shale oil on the ash particles as well as shale oil decomposition the free volume of the reactor. Calculations show that the secondary reactions lead to reduce the shale oil yield from 0.27 to 0.13 kg/kg of dry shale, what is close to operating data of the various industrial units based on the Galoter technology.

1. Introduction

The increase of energy resources demand and the limited nature of the world reserves of conventional fossil fuels such as crude oil, natural gas, and coal raise a problem of alternative energy sources search. In this connection, oil shale has a great potential for future applications as energy source because the oil content in the world oil shale reserves (whether economically recoverable or not) exceeds conventional oil reserves [1]. Oil shale is characterized by a high content of mineral components that are closely connected with organic matter (kerogen) [2]. Therefore, the direct combustion of oil shale in boilers is confronted with some technical difficulties. Oil shale processing is a complex approach to solid fuel utilization, at which purely power processes (fuel combustion and heat release) are coupled with fuel conversion ones (gasification and pyrolysis) [3]. The main purpose of such approach is an increase of the efficiency of low grade fuel conversion with simultaneous minimization of the environmental impact [4]. The research efforts and industrial applications of oil shale processing are concentrated on the improvement of the existing technologies as well as on the development of new methods of oil shale use [5,6].

Currently, there are a number of technologies for thermal processing of oil shale [7]. However, only some of them have found commercial application. The most effective technology for oil shale thermal processing is the Galoter one [8,9]. This technology is based on the principle of a quick heating of fine grained oil shale by the solid heat carrier (hot ash) in the rotary drum reactor followed by its thermal decomposition. The Galoter technology was realized in two pilot units UTT-200 and UTT-500 with a capacity of 150–200 t and 500 t of shale per day, correspondingly. The industrial realization of the technology was implemented in two units UTT-3000 at the Estonian Power Station. The main operating parameters of the UTT units are given in Table 1 [10]. These units allow obtaining a high-calorific liquid fuel with a calorific value of 38–40 MJ/kg and gaseous fuel with a calorific value of 41–42 MJ/kg from primary oil shale (Baltic kukersite) with a calorific value of 7.8–8.4 MJ/kg. The characterization of the Baltic kukersite is given in Table 2. Its mineral part consists mainly of limestone and aluminum silicates. The Fisher assay yields are as follows (wt.%, dry basis): shale oil — 25.5; pyrogenic water — 4.6; coke residue — 65.8; gas and losses — 4.1.

The UTT units operate with oil shale fines with a particle size of 0–25 mm. The elimination of particle agglomeration in the pyrolysis reactor can be reached owing to continuous motion of the mixture of fuel and solid head carrier. The simple construction that can realize this requirement is horizontal rotating drum. The principle scheme of the UTT-3000 unit is presented in Fig. 1. The oil shale is previously dried in the aerated fountain dryer 1 and then thermally decomposes in the rotary drum reactor 4 at temperature of the order of 500 °C under mixing with hot shale ash in the mixer 3. The solid semicoke particles are separated in the dust separator 5 after pyrolysis reactor and then transported into aerated fountain furnace 6 with temperature 750–850 °C, where semicoke combustion occurs at the reducing conditions (excess air factor is less than unit) to ensure the required temperature of the ash. The volatile products of oil shale pyrolysis (shale oil and

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http://dx.doi.org/10.1016/j.fuproc.2015.08.001
0378-3820/

Table 1
Average operating data of the UTT units [10].

Parameter	Unit type		
	UTT-200	UTT-500	UTT-3000
Raw oil shale feed (t/d)	144	469	2385
Shale oil yield (kg/t, dry basis)	154	136	129
Gas yield (nm^3/t, dry basis)	52.0	49.0	36.4

gas) after rotary drum reactor are cooled in the condenser 12 with yield of heavy oil, light oil, and high calorific gas.

The rational hardware design plays an important role in research and development of new UTT units. In particular, this applies to the choice of optimal design parameters and the creation of the calculation method for the most responsible installation component, namely, rotary drum reactor.

Within the last decade, many studies have been carried out, which use multi-scale modeling of heat and mass transfer processes in various types of rotary reactors [11–14]. In this work, the comprehensive multi-scale numerical analysis of physical and chemical processes in the rotary reactor was performed as applied to oil shale thermal decomposition in the pyrolysis reactor of the UTT unit. The elaborated mathematical model includes the kinetics of kerogen decomposition, heat and mass transfer inside and outside of the single oil shale particle, the secondary chemical reactions, etc. The common description of these processes is impossible and requires some simplifications and model approaches that were received with the use of available experimental data.

2. Mathematical model of pyrolysis reactor

2.1. Motion of bulk solids in reactor

Despite the wide use of the rotary reactors in various industries (cement, silicate, chemical, metallurgical, and others), their investigations are not universal. Most of the works deal with inclined drums with open end that have a relatively large length and low coefficient of volume filling [15,16]. Experimental study of bulk solid motion along the axis of such reactors makes it possible to determine the dependence of the axial speed and the degree of reactor filling from design and operating parameters of the reactor (sizes, angle of axis inclination, speed of rotation, feed rate of the solids, and so on) [17]. Modeling study of such reactors is conducted with the use of one dimensional [18] and three dimensional [19] mathematical models. It must be pointed that the use of more complex models does not always give an adequate description of the processes occurring in the reactor.

The special property of the pyrolysis reactor in the UTT unit is that it belongs to short (the relation between the length L and the diameter D is equal to 2–3) horizontal drums that rotate with a low velocity (1–2 rpm) and operate with a high degree of the volume filling with moving material ($\varphi = 0.3$–0.6) at a relatively short residence time of the solid phase here ($\tau_r = 500$–2000 s) and a comparatively high temperature ($T = 400$–600 °C) [10]. The main technical characteristics of the pyrolysis reactor in the UTT unit that were used in calculations are given in Table 3. The particle size distribution of solids at the inlet to the pyrolysis reactor is given in Table 4.

The scheme of the reactor is shown in Fig. 2. The reactor loading by the solid phase (fuel and solid heat carrier particles) is realized by gravity without the use of feeding devices and mechanisms operating at relatively high temperatures. The simplest way to increase the reactor volume filling is by installing of supporting devices at the drum ends. The solid phase after mixer 3 (see Fig. 1) enters the reactor volume through the inlet supporting device 1 and then moves in the axial direction z by gravity due to the reactor rotation that results in the difference in the heights of the solid phase layer 2 at the inlet and outlet of the drum. The design of the outlet end of the reactor according to such scheme is determined by the supporting device 4 in the form of a cylindrical socket with a diameter d (relatively small as compared to D) and length l. The volatile products of fuel particle thermal decomposition are filtrated through the solid layer 2 in the free volume of the reactor 3 and then leave the reactor through the outlet supporting device 4.

Table 2
Proximate and ultimate analysis of Baltic kukersite.

Proximate analysis (wt.%)		Ultimate analysis (wt.%)	
Moisture	13.10	C	24.18
Volatile matter	29.41	H	3.10
Ash	54.22	O	3.69
Fixed carbon	3.27	N	0.11
		S	1.60

The comprehensive experimental study of the bulk solid motion in the reactor of this type was carried out in [20]. The analysis of the received experimental data makes it possible to determine the dependence of the main parameters of the solid dynamics from the design and operating parameters of the reactor. The velocity of solid phase motion u in the horizontal direction (or residence time of particles in the reactor $\tau = L/u$) and the degree of filling of the reactor with the solid phase φ depend on the flow rates of the fuel G_f and the solid head carrier (ash) G_a as well as the angular rotational velocity ω of the reactor and its diameter D and length L. They also depend on the dimension of the supporting device. It was determined on the base of analysis of experimental data [20] that these process parameters can be expressed as follow:

$$\phi = AR_s^p \,\mathrm{Fr}^{0.05}(d/D)^q(1 + l/D), \tag{1}$$

$$u = L(G_f + G_a)/(\rho_s \phi V), \tag{2}$$

where $R_s = 2\pi\,(G_f + G_a)\,/\,(\rho_s\omega V)$, $\mathrm{Fr} = (\omega/2\pi)^2 D/g$, ρ_s is the average density of the solid phase, $V = 0.25\pi D^2 L$ is the reactor volume, g is the acceleration of gravity. At $R_s = 0.01$–0.20, $\mathrm{Fr} = (3$–$110) \times 10^{-5}$, $L/D = 1.8$–5.0, $d/D = 0.24$–0.64, and $l/D = 0.005$–0.5 the parameters A, p, and q in Eqs. (1) and (2) are equal: $A = 0.066\exp(0.95\,L/D)$, $p = 0.18(L/D - 1)\,d/D$, and $q = 0.5\,(L/D - 3.1)$.

2.2. Heat transfer between heat carrier and fuel

One of the main stages in the construction of the mathematical model of the pyrolysis reactor is carrying out of the submodel of individual fuel particle behavior in the high temperature region. To ascertain the basic regularities of the heat transfer between particles of solid fuel and the heat transfer agent in the reactor, one can use the model of plug flow reactor, in which each volume element of solid medium is considered as an open chemical system moving along the reactor axis z (see Fig. 2) with the velocity u and exchanging by the matter and energy with environment. The running coordinate z of the volume element is determined by the current time t multiplied on the velocity u.

It is assumed that the size of head carrier particles is sufficiently small to be considered as isothermal and that their mixing with fuel particles occurs instantaneously and uniformly at the inlet of the reactor. The last assumption is not fully correct and needs further improvements. To take into account the polydispersity of the fuel, the fuel particles are divided into fractions, in which the particle size is the same. The equation of enthalpy balance between the particles of the solid head carrier and the fuel at constant heat capacities of the particles and in the absence of heat loss can be written as:

$$c_a G_a[T_{a0} - T_a(t)] = c_f G_f \sum_{k=1}^{N} (f_k/V_k) \int_0^{R_k} [T(r,t) - T_{f0}] 4\pi r^2 dr. \tag{3}$$

Here c_a and T_a are the heat capacity and temperature of the heat carrier; N is number of fractions; f_k is mass part of the particles of the