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Design of a moving bed reactor for the production of uranium tetrafluoride based on mathematical modeling

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ABSTRACT

A new model has been developed to analyze the performance of a moving bed reactor producing uranium tetrafluoride. The model includes the solid and gas flow rates, heat transfer by convection, conduction and radiation between gas and solid streams and by the external surrounding. A kinetics model has been also developed for the gas–solid reactions on the basis of the grain model in which, the different resistances are taken into account to determine the overall reaction rates. This study provides an initial and theoretical basis for optimum design of the moving bed on the basis of the kinetics models. The optimum design is carried out from the viewpoint of required length of the reactor and the maximum conversion. According to the primary expectations which are provided by the kinetics models, the feed temperature seems not to have any effective influence on the reactor performance. On the other hand, it is predicted that the inlet gas flow rate contributes an important role. The investigations show that although the kinetics models are rather useful in primary estimations, the strong interdependence between different zones of the reactor may prevent general expressions.

1. Introduction

Enriched uranium hexafluoride (UF_6) is mainly used in the nuclear industries to produce nuclear fuel. UF_6 is prepared via several stages from the uranium-bearing ores. In the French technology, UF_6 is produced from UF_4 in a flame reactor (Dussoubs et al., 2003). Several reduction reactions are carried out in a special type reactor to produce UF_4 that part of it resembles a moving-bed reactor in which granulated uranium trioxide (UO_3) is reduced to uranium dioxide (UO_2). The latter is then converted to uranium tetrafluoride (UF_4) by passing anhydrous HF over it in the next part of the reactor, which usually involves vertical and horizontal sections. The powder flows by gravity through the moving bed and is then propelled forward in a horizontal section by a built-in screw feeder. Fig. 1 shows illustration of the referred moving bed reactor (Dussoubs et al., 2003).

The moving bed reactor can be divided into four different zones including reduction, buffer, vertical hydrofluorination, and horizontal hydrofluorination zones. UO_3 pellets fed at the top of the reactor are exposed to a counter-flow partially cracked NH_3. In the presence of H_2, which is produced as the reducing gas through thermal cracking of NH_3, two consecutive reduction reactions take place. Through the first reaction UO_3 is converted into U_3O_8 and through the second UO_2 is produced as the main product of the reduction section from U_3O_8. In the vertical and horizontal hydrofluorination zones, UO_2 encountered a counter-flow of gaseous HF and through the hydrofluorination reaction it is converted into uranium tetrafluoride. UF_4 is extracted from the final side of the horizontal section by an Archimedes screw.

The required heat for NH_3 cracking is supplied by the heating muffles. Heating muffles partly surround the reduction and horizontal hydrofluorination zones. Since the reduction and hydrofluorination reactions are very exothermic, temperature control is one of the difficulties for proper operating of the process. Local hot spots also can soften the pellets and lead to stopping of the bed. The heat of very exothermic hydrofluorination reaction is removed by the air cooling of the outer walls of the reactor, and also by an inner, coaxial heat exchanger. A series of thermocouples diving in the moving bed allows the operators to continuously check the temperature at different locations (Dussoubs et al., 2003, 2002).

Mathematical modeling of an industrial type moving bed reactor producing UF_4 has been considered before by Dussoubs et al. (2003; 2002). By the application of the law of additive characteristic times, as kinetics model, they provided a mathematical model to analyze the behavior of different parts of the reactor. However, the validity of the law of additive characteristic times is verified only when there is no temperature gradient inside the pellet (Sohn, 2003). Furthermore, this law seems to be quite weak in predicting solid conversion in reversible reactions.

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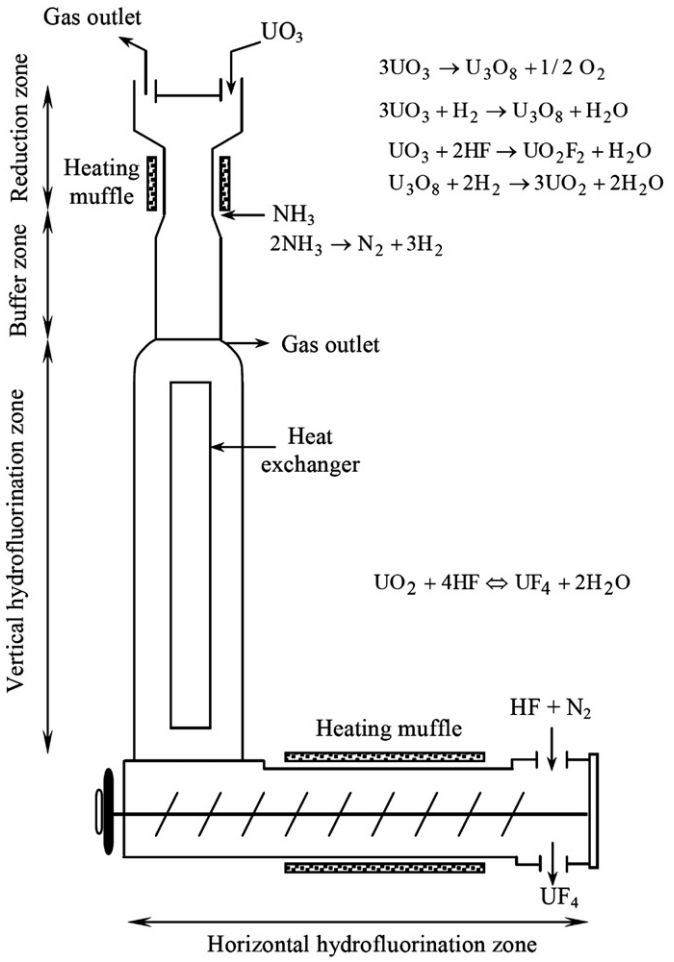

Fig. 1. Schematic configuration of a moving bed reactor (Dussoubs et al., 2003).

In fact, in the case of a reversible reaction, those relations commonly developed for the characteristic times are in no way capable to predict rates of the reverse reaction. Even though the reactor operational conditions are always controlled in a manner that the possibility of the reverse reaction (undesired one) is almost zero or negligible, however the presented models should be able to predict such a condition as well. To our knowledge, there is no other paper on this topic while the need for a more comprehensive model as well as further investigation is remarkable due to the importance of the moving bed performance in the nuclear industries. For this purpose, in this study a comprehensive mathematical model is developed in order to investigate the performance of a special type of moving bed reactors which is used to produce uranium tetraflouride. In this model the simultaneous effects of all types of transfer phenomena on existing gas–solid reactions are considered to reach a further realistic kinetics model. Besides using the proposed model, there is a theoretical basis offered for optimum designing of such reactors.

2. Mathematical model

2.1. Kinetics of chemical reactions

As mentioned before, the selected moving bed reactor for modeling in this study mainly involves the solid pellets which perform several noncatalytic gas–solid reactions. To provide a comprehensive model special attention must be paid to the chemical reactions.

In the reduction zone UO_3 is converted into desired U_3O_8 and undesired UO_2F_2 simultaneously by the first reduction and secondary hydrofluorination reactions, respectively. The former is carried out in the presence of H_2 coming up from the bottom of the reduction zone, and the latter is induced by the presence of a small amount of HF coming up from the lower hydrofluorination zone. Calcination of UO_3 to U_3O_8 is another chemical reaction that may be considered in the reduction zone. To our knowledge there is no explicit rate expression presented in the literature for this reaction but one presented by Dussoubs et al. (2003). In a primary attempt of Niksiar and Rahimi (2009a), this rate expression was applied for a single solid pellet and it is found that it over predicts the reaction rate. In this work we have neglected the calcination of UO_3. The second reduction reaction begins only when UO_3 is entirely converted to U_3O_8 (Page and Fane, 1974). Part of the required H_2 for performing all of the reduction reactions is attained by the homogeneous reaction of NH_3 cracking. Finally, the hydrofluorination reaction occurs in both vertical and horizontal hydrofluorination zones leading to the production of UF_4. The details of the reactions are presented together with their rate expressions in Table 1.

2.2. Modeling the vertical and horizontal zones

For the moving bed a one-dimensional model (in the z direction) is developed where the gas and solid streams are assumed to be in plug flow condition. Also, it is supposed that the pellets are not broken up during their precipitation. Each gas–solid reaction is modeled on the basis of the grain model (see Section 2.3). By this model the concentration distribution of the reactant or the product gas is determined inside each solid pellet. Therefore, the rate of disappearance/addition of different gas species from/to the gaseous bulk flow of the moving bed per solid pellet can be evaluated. For gas species i participating in reaction rxn it yields

$$r'_{rxn,i} = 4\pi R_p^2 D_{ei} \left.\frac{\partial C_i}{\partial r}\right|_{r=R_p} \tag{1}$$

Say $r'_{rxn,i}$ a virtual reaction rate of gas reactant i. If $\dot{n}_p$ is the number of all pellets per unit time entering the reactor, namely

$$\dot{n}_p = \frac{3F_{s,in}}{4\pi R_p^3 \rho_B (1-\varepsilon_p)} \tag{2}$$

the mass balance equation for gas species i in the gaseous bulk flow can be expressed as (Niksiar, 2009)

$$\frac{dF_{gi}}{dz} = \frac{\dot{n}_p}{u_s} \sum_{rxn=1}^{k} \alpha_i \, r'_{rxn,i} + r_{cr,i} A_b \varepsilon_b \tag{3}$$

where k represents the number of those gas–solid reactions that cause gas i to consume or produce. α_i is equal to 1 for gas reactants and equal to c_i for gas products. The last term on the right hand of Eq. (3) is introduced only for gas species H_2, NH_3 and N_2 in the reduction zone that incorporate the cracking reaction.

In addition, a total matter balance is written for the gas phase in each zone:

- in the reduction zone

$$\frac{dF_{gt}}{dz} = -2r_{cr}A_b\varepsilon_b + \frac{1}{2}\frac{\dot{n}_p}{u_s} r'_{hfs} \tag{4}$$

- in the vertical hydrofluorination zone

$$\frac{dF_{gt}}{dz} = \frac{1}{2}\frac{\dot{n}_p}{u_s} r'_f \tag{5}$$