

Modeling Operational Parameters for Uranium Dioxide Production Reactor through Uranium Trioxide Reaction Using Hydrogen

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Abstract

This article shows the modeling of a uranium dioxide production reactor using COMSOL Multiphysics software program in its 4.3b version. The model was made using 3 kinds of studies: momentum, heat and mass transport, in order to determine the influence of the most important operational parameters: UO₃ reaction rate, composition and flow of the reduction gas, the initial temperature reactor and the reducing gas. The operational parameters evaluated were the followings: constant gas flow of 2.5 L/min, initial hydrogen concentration of 0.25, 0.50 and 0.75 M, and initial temperature of 400°C. The obtained results allow to conclude that under these working conditions, uranium dioxide is obtained virtually instantaneous and, with concentrations close to 0.5 M H₂ in the reducing gas, the process can operate continuously and autogenously, without applying additional energy and temperatures around 600°C.

Keywords

Uranium Trioxide, Uranium Dioxide, Conversion, Modeling

1. Introduction

Nuclear energy is a source of major electricity production, due to the high-energy capacity of the uranium fuel element during nuclear fission. Uranium is used mainly in the form of UO₂ type compounds. For this purpose, it

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must be enriched in its U^{235} isotope to fulfill this purpose. The needed enrichment level will depend on the type of reactor used, and there are 2 types: Light Water Reactors (LWR), which require an enrichment of U^{235} minimum between 3 and 5% to operate, and Pressurized Heavy Water Reactor (PHWR), which uses natural uranium as fuel [1].

Figure 1 shows the uranium conversion for the production of UF_6 , raw material for the enrichment process.

This process reduces the uranium compound, either from UO_3 or from U_3O_8 , and obtains UO_2 powders, according to the overall reaction:



Subsequently, UO_2 powder should be treated by hydrofluorination to obtain UF_4 powders:



Finally, the UF_4 compound is taken to the fluorination process to obtain UF_6 :



The UF_6 compound, gaseous at $56^\circ C$ and atmospheric pressure, is carried to the enrichment process, in order to obtain a concentrated and a diluted fraction in U^{235} through centrifugal force reactors [3], using the different weights of this and the U^{238} isotope. This process is shown schematically in **Figure 2** [4].

The gaseous compound UF_6 , once enriched, must return to its UO_2 form. To do this, there are 3 methods to carry out this task [7] [8].

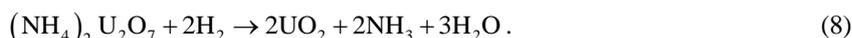
H₂ reduction: The $UF_{6(g)}$ compound is reduced using hydrogen according to the reactions:



Hydrolyzing: The $UF_{6(g)}$ compound is hydrolyzed using water:



Subsequently, the ammonium diuranate is reduced using H_2 at temperatures of $600^\circ C - 800^\circ C$:



Finally, the third method for enriched UO_2 production is through precipitation of ammonium uranyl carbonate instead of ammonium diuranate, according to reaction (10):

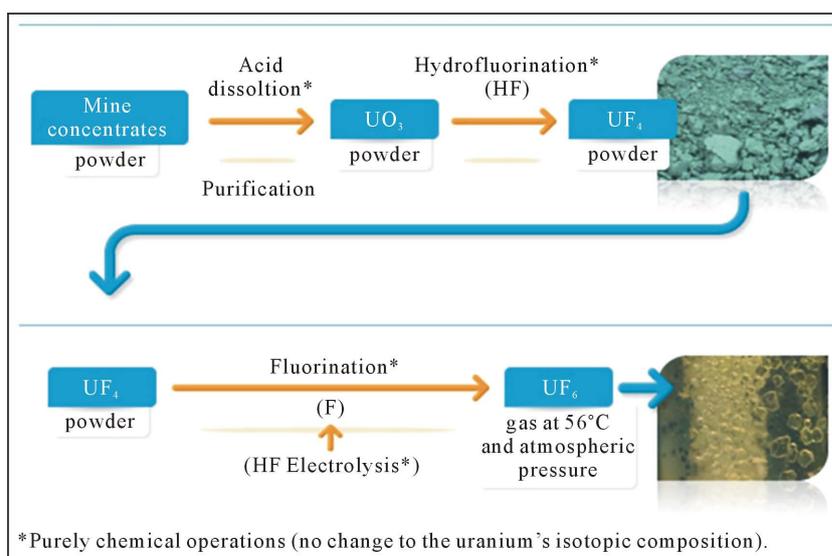


Figure 1. Uranium conversion process [2] [3].

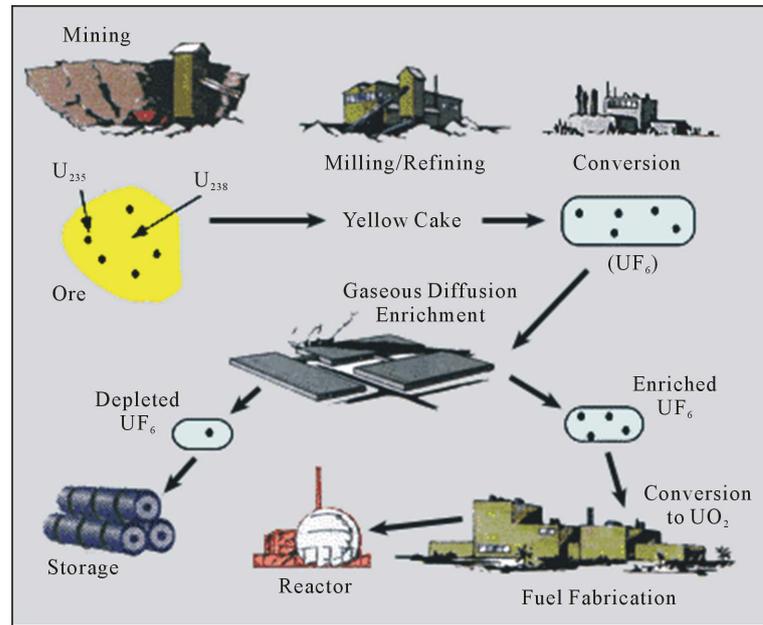


Figure 2. The process leading to obtain UO₂ fuel elements [5] [6].



All these processes require the use of a fluidized bed reactor for producing UO₂ for both UF₆ production, intended for further uranium enrichment, as for reproduction as UO₂ fuel pellets for nuclear power reactors.

It is for these reasons that this work will evaluate the main operating parameters of a fluidized bed reactor for UO₂ production from UO₃ concentrates using mathematical modeling techniques and the COMSOL Multiphysics software, in its 4.3b version. These parameters are the followings: feed flow, hydrogen concentration and initial temperature.

Conventional UO₃ to UO₂ reduction reactor is shown in Figure 3.

UO₂ production is conventionally performed in a reduction reactor as shown in Figure 3. Here, uranium trioxide and hydrogen are continuously fed, H₂ is normally diluted in an inert gas such as argon or nitrogen. It is also common to use ammonia, which is cracked [11] to obtain a nitrogen and hydrogen gas mixture. This UO₂ production reactor [12] [13] is controlled by the following operational parameters: composition and flow of the reducing gas, UO₃ feed and internal reactor temperature.

2. Theoretical Basis [14]

2.1. Momentum Transport

Momentum transport is given by the Navier-Stokes equation, for compressible fluids:

$$\rho(u \cdot \nabla)u = \nabla \cdot \left[-\rho I + \mu \left(\nabla u + (\nabla u)^T \right) \right] + F \quad (11)$$

$$\rho \nabla \cdot u = 0 \quad (12)$$

where: ρ : density, u : H₂ flow velocity, I : identity matrix, F : External forces.

Momentum transport will determine the gas behavior inside the reduction reactor, for laminar regime.

2.2. Heat Transport

Heat transport is determined by the following equations:

$$\rho C_p \frac{\partial T}{\partial x} + \rho C_p u \cdot \Delta T = \nabla \cdot (k \nabla T) + Q \quad (13)$$

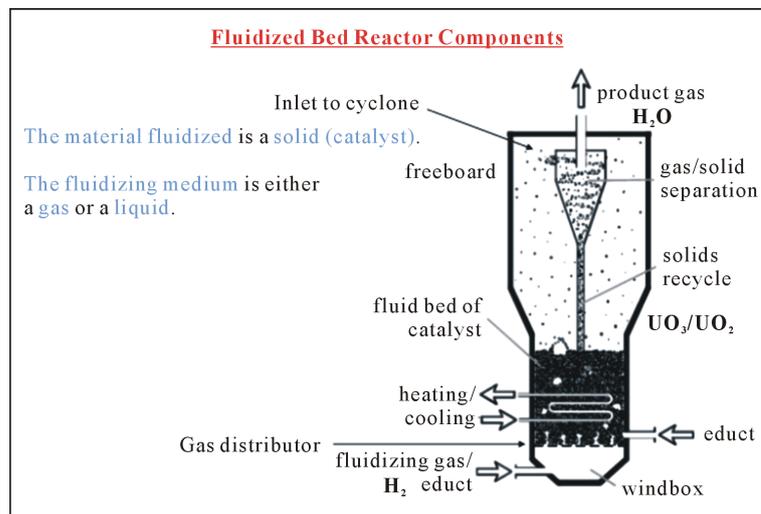


Figure 3. Conventional UO_2 production reactor [9] [10].

$$\frac{d\rho}{dt} + \rho \cdot (\nabla v) = 0 \quad (14)$$

where ρ : reduction gas density, C_p : specific heat, u : H_2 flow velocity, T : temperature, k : conduction coefficient, Q : source term, t : time, v : volume.

Solving this equation in the model will determine the temperature inside the UO_3 reduction reactor and the generated heat by the exothermic reaction between the UO_3 concentrate and the H_2 reducing gas.

2.3. Transport of Diluted Species

Mass transport for dilute species is determined by the equation:

$$\nabla \cdot (-D_i \nabla C_i) + u \cdot \nabla C_i = R_i \quad (15)$$

where D : Hydrogen diffusion coefficient, C : Hydrogen concentration in the reduction gas, u : Gas velocity flow, R : Hydrogen consumption rate in the reaction zone.

This equation will determine the concentration profile of hydrogen and/or water displayed by the system in continuous operation type.

3. Reactor Modeling [15]

The reactor used in the development of experiences is shown in **Figure 4**.

This reactor is a tubular type, with input and output in conical shape. Its length is 1.66 m. and it has an internal diameter of 4.5 cm. The development of the experiences was performed by setting as parameters the initial temperature of both the gas and the reactor, which was between 400°C - 500°C and initial hydrogen concentrations in the reducing gas: 0.25, 0.5 and 0.75 M. The flow feeding the reduction reactor was kept constant at 2.5 L/min. The input speed of this flow was 0.1 m/s, and the cross section of 1.5 cm, so the feed flow regime was laminar.

To develop this model, the 3 transport phenomena mentioned before were occupied: momentum, heat and mass for dilute species. Working conditions are shown in **Figure 5**:

Figure 5 shows the considered parameters for the modeling: In the case of momentum transport, the reducing gas has inlet and outlet inside the reactor, and there is no slip flow on the walls. For heat transport, it is considered that the initial temperature for uranium conversion processes is 700 K. The walls symbolize heat losses by natural convection, where this system was covered by a thermal jacket at 450 K. The remaining heat will be generated by the exothermic reaction inside the vessel between UO_3 and H_2 . Finally, for the transport of diluted species, reducing gas flow has the aforementioned initial hydrogen concentrations. A fraction of this gas is consumed by the UO_3 vessel. The remaining hydrogen at the reactor outlet was burned to prevent gas leakage. In



Figure 4. UO₂ conversion reactor.

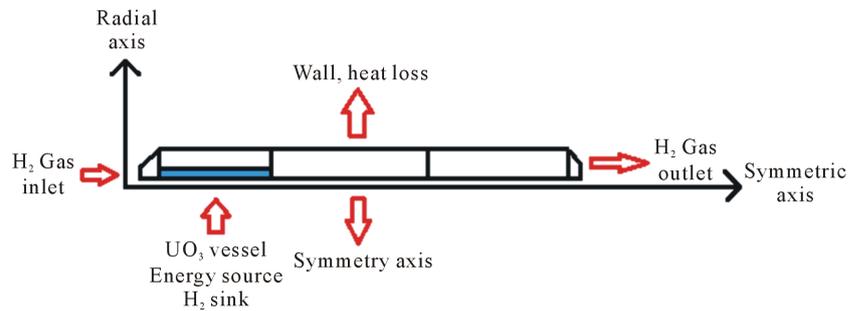


Figure 5. Working conditions for UO₂ production.

order to simplify the development of the calculations, the model was proposed using as axial symmetry, in the bottom of Figure 5.

The equations used to the model development were obtained from previous studies [15]. In this work, the reduction kinetics of uranium trioxide was developed based on the formation of an intermediate compound, U₃O₈, according to the following reactions:



The reduction rates for both equations were determined by using Arrhenius's Law, according to Equation (18):

$$\frac{\partial y}{\partial t} = A * \exp\left(\frac{-E_a}{R * T}\right) * cH_2^n \quad (18)$$

where $\frac{\partial y}{\partial t}$: UO₃ reaction rate, cH_2 : hydrogen concentration in the reducing gas, n : reaction order for hydrogen concentration, R : universal gas constant, T : absolute temperature, E_a : activation energy.

The corresponding reaction rates for Equations (17) and (18) are, respectively:

$$\frac{\partial y}{\partial t} = 6.66 * 10^5 * pH_2^{1.17} * \exp\left(\frac{-96799}{R * T}\right) \quad (19)$$

$$\frac{\partial y}{\partial t} = 6.7 * 10^{13} * pH_2^{1.07} * \exp\left(\frac{-203693}{R * T}\right). \quad (20)$$

According to other authors [16], the conversion process is usually made from U₃O₈ compounds, as the uranium oxide with greater chemical stability. This implies that reaction (19) occurs uneventfully. Moreover, reac-

tion rate (20) involves the formation of other intermediate oxides of uranium, such as U_3O_7 or U_4O_9 , which, as already discussed in other studies [15] [16], decrease the overall speed of the process at temperatures above 700°C . For this reason, reaction kinetics (21), which is the controlling step, will be used for modeling the reduction reactor.

4. Results

The obtained results were as follows:

According to Equations (12) and (13), it can be said that the progress of these reactions is quantified through water production. **Figure 6** shows the UO_3 reduction kinetics is so fast at these temperatures, almost no hydrogen exists in the vicinity of the vessel, where the reaction occurs. The following figure shows the linear profiles generating water into the reactor, along the axial symmetry.

Figure 7 shows that, for all hydrogen concentrations in the reducing gas, they are consumed almost entirely in the UO_3 vessel.

However, the most important parameter for the development of experiences is the temperature profile along the reactor, and the exothermic peak reached by the system due to the release of reaction heat, as this defines the capacity of the system to work autonomously, without the need of additional energy. This profile is determined by the release of energy from the exothermic reaction between uranium oxides and hydrogen. The profiles obtained for the hydrogen concentrations in the reducing gas, are shown in **Figure 8**.

According to bibliographic data [15], the ideal temperature for the development of these experiences is 500°C - 600°C . The objective of this result is to check whether the process can operate autonomously, keeping the temperature within this range. For this reason, the process must be performed at 0.5 M H_2 concentrations to ensure this stationarity condition.

5. Validation of Results

The results of the experiments carried out in this reactor are shown in the following figure:

Figure 9 shows that UO_3 reaction kinetics is virtually instantaneous in contact with the reducing H_2 gas, since for temperatures of 500°C and 600°C , the transformed fraction of UO_3 to UO_2 it is above 90%.

In the case of temperature, the experimental exothermic peaks obtained are compared with the modeling system.

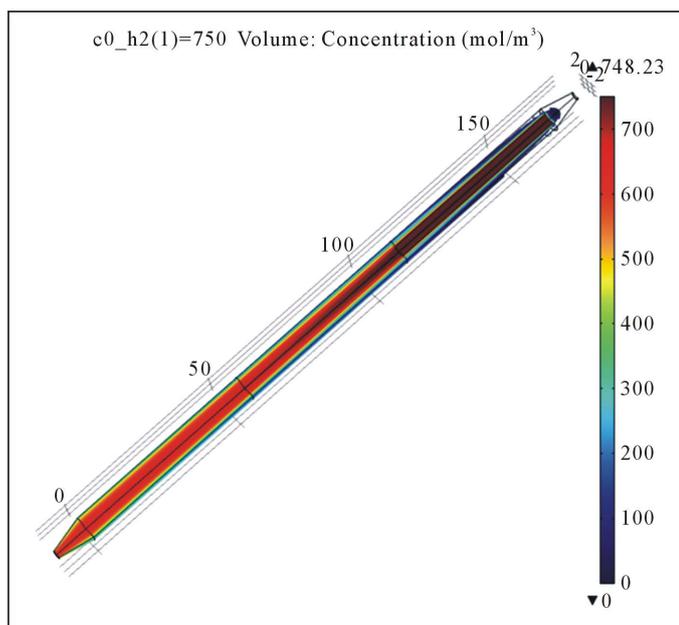


Figure 6. Volumetric concentration profile of water inside the reduction reactor.

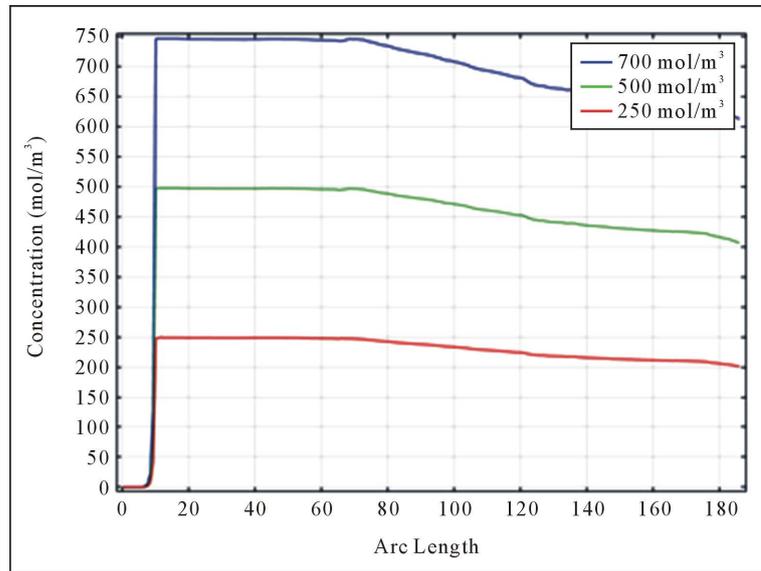


Figure 7. Water profile concentrations inside the reduction reactor.

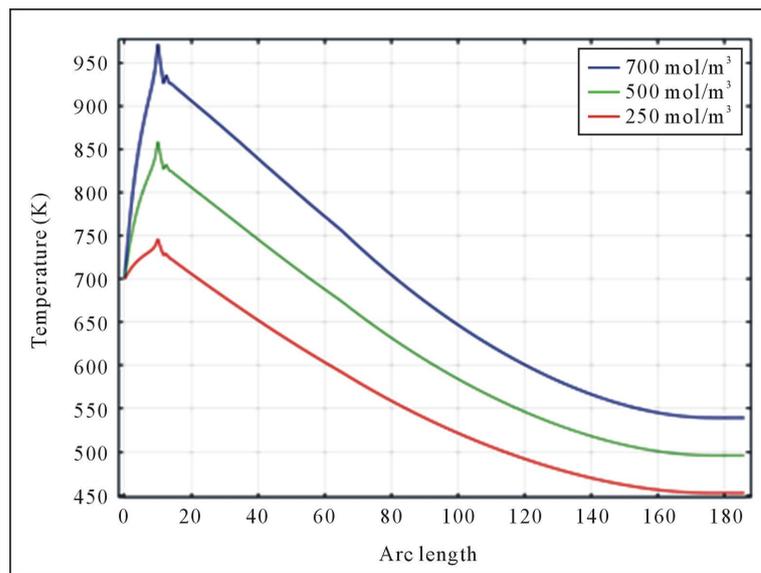


Figure 8. Temperature profile for the UO₃ reduction reactor.

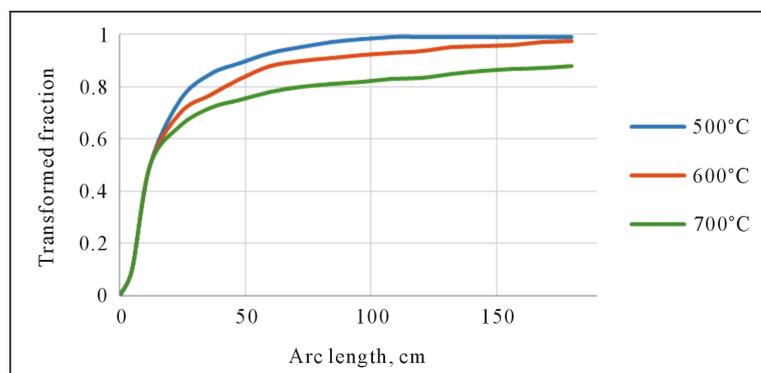


Figure 9. Reaction kinetics for uranium concentrates, at constant temperature.

Table 1. Comparison between exothermic peaks.

Initial T° (K)	[H ₂] (Mol/m ³)	Exothermal peak (K) (experimental)	Exothermal peak (K) (model)	Relative error (%)
700	250	725	750	3.44
700	500	805	860	6.83
700	750	890	980	10

Table 1 shows that the temperature system, operating continuously, can be predicted in an acceptable way using mathematical modeling techniques, since the data obtained for the proposed working conditions show relative errors of less than 10%.

6. Conclusions

- From the previous data of UO₂ production, it is possible to predict the reduction of UO₃ compounds with hydrogen using mathematical modeling techniques.
- The fluidized bed reactors can be monitored effectively with the gaseous products, which means that it is not necessary to manipulate the UO₃ vessel. This fact allows the safety during the data collection.
- For the studied kinetic parameters, the UO₃ reduction reactions occur almost instantly, as the results show a hydrogen conversion to water almost completely.
- The condition that allows continuous operation for the reduction reactor is feeding with reducing gas at 0.5M H₂.
- The temperature profiles and relative errors allowed to conclude that the reduction of U₃O₈ compounds to UO₂ provided the required energy to maintain the working temperature in the required ranges.

Acknowledgements

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