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Mathematical model of the oxidation of a uranium carbide fuel pellet including an adherent product layer

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ABSTRACT

Uranium carbide is a candidate fuel for Generation IV nuclear reactors. However, like any candidate fuel, a reprocessing route should be established before implementation. One proposed method involves a pre-oxidation step, where the carbide fuel is oxidized to an oxide and then reprocessed as normal. A mathematical model has been developed to simulate such an oxidation using finite difference approximations of the heat and mass transfer processes occurring. Available literature was consulted to provide coefficients for the reaction rates and importantly the diffusion of oxygen through the adherent oxide layer that forms on the carbide: the rate limiting step. The transient temperature, oxygen and carbon monoxide distributions through the system are modeled in order to predict oxidation completion times and the temperatures reached. It was found that for a spherical pellet of radius 0.935 cm, the oxidation can take from 1 to 19 h depending on the oxidation conditions and reach temperatures of up to 1556 °C. A robust model results that offers increased understanding of a process crucial to the sustainable use of carbide fuels in energy generation.

1. Introduction

Uranium monocarbide (UC) is under consideration for use in Generation IV reactors due to its higher conductivity and metal atom density making it easier to control in a reactor [1–4]. An important consideration when implementing a novel fuel type into the nuclear fuel cycle is that it must be able to be efficiently reprocessed. Ideally, carbide fuel would be reprocessed in the same manner as the current most widely used fuel, uranium oxide, in order to save on infrastructure and having to develop new expertize.

A complication arises upon dissolution of carbides in nitric acid, a step in the Purex process currently employed to reprocess oxide fuel. A significant quantity of the carbon displaced by the oxidation remains in the solution as soluble organics, which then reduce the extractability of the fissile material, uranium and plutonium, from the dissolution liquor. This leads to unacceptable losses of fissile material that could otherwise be reused, increasing the volume of nuclear waste generated by carbide fuels.

One proposed solution is pre-oxidizing the UC fuel pellet in air to its oxide form allowing reprocessing as normal. The oxidation, however, is highly exothermic making the process potentially dangerous [1,5]. The motivation of this work, therefore, is to provide a model simulating the oxidation in order to predict the temperature, reaction rate and the occurrence of

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Symbols

A	Surface area of the uranium carbide pellet m
a	Pore size m
C	Concentration mol m^{-3}
C^B	Concentration in the bulk gas mol m^{-3}
c_p	Specific heat capacity J kg^{-1} K^{-1}
D	Diffusivity m^2s^{-1}
$\mathcal{D}_K$	Knudsen diffusivity m^2s^{-1}
d_e	Spherical pellet diameter m
E_A	Activation energy of uranium carbide oxidation J mol^{-1}
E_A^+	Activation energy of oxygen diffusion through the oxide layer J mol^{-1}
E_{CO}	Activation energy of carbon monoxide oxidation J mol^{-1}
h	Heat transfer coefficient W m^{-2} K^{-1}
i	An integer representing the radial node
k	Thermal conductivity, or an integer representing the radial node at the solid surface W $m^{-1}K^{-1}$
k_1	Rate coefficient for UC oxidation m s^{-1}
k_g	External diffusion coefficient m s^{-1}
L	Cylindrical pellet length m
$L_{(U)(C,O)}$	Solubility of oxygen in uranium carbide J mol^{-1}
M	Molar mass kg mol^{-1}
n	An integer designating the current time step
Nu	Nusselt number
n_{UC}	Number of moles of uranium carbide mol
$n_{U_3O_8}$	Number of moles of triuranium octoxide mol
Pr	Prandtl number
p	An integer representing the radial node at the reaction interface
r	Radius within solid m
R	Gas constant J $mol^{-1}K^{-1}$
R_C^*	Rate of oxygen transfer across gas film layer mol s^{-1}
r_{ox}	Radial thickness of the initial layer m
R_C	Rate of oxygen consumed by UC oxidation mol s^{-1}
R_{CO}	Rate of carbon monoxide oxidation mol s^{-1}
Re	Reynolds number
r_1	Radius of the carbide pellet m
r_2	Radius of the solid m
Sc	Schmidt number
Sh	Sherwood number
t	Time passed since reaction started s
T	Solid temperature K
T_{Amb}	Ambient temperature K
T^B	Bulk gas temperature K
$\bar{T}$	Average temperature K
u_1, u_2, u_3	Solutions at increment sizes of h_1, h_2 and h_3 respectively
u	Estimated solution using an infinitely small increment size
V	Volume of oxidizing gas m^3
v	Coefficient representing $1 - 1/i$
w	Coefficient representing $1 + 1/i$

Greek symbols

α	Thermal diffusivity m^2s^{-1}
ΔH_R	Enthalpy of oxidation at carbide surface J mol^{-1}
ΔH_{CO}	Enthalpy of carbon monoxide J mol^{-1}
Δr	Radial increment size m
ε	Emissivity
μ	Fluid dynamic viscosity kg m^{-3}
μ_{O_2}	Oxygen potential J mol^{-1}
ρ	Density kg m^{-3}