Unsteady State Heat Transfer in Externally Heated Magnesio Thermic Reduction Reactor: An overview

Harekrushna Sutar[1] and Abanti Sahoo[2+]

[1]Uranium extraction division, BARC, Mumbai, INDIA

[2] Chemical Engg. Dept., *National Institute of Technology*, Rourkela ,INDIA

Abstract. The magnesio thermic reduction process for the production of uranium which involves reduction of uranium tetra fluoride with magnesium in a sealed reactor has been studied in the present work. The process is highly exothermic and generates very high temperature in the core region. Safe operations of the process require accurate temperature control and heating. Simulations are carried out using Anupravha a computational fluid dynamics (CFD) and heat transfer solver to study the temperature profiles inside the reactor including its lining. The results are studied for both preheating and reaction stage which gives an idea about the reaction temperature and molten mass inside the reactor. Thus the present study can be of great help for correct design of reactor thereby preventing nuclear radiation to the surroundings.

Keywords: Magnesio Thermic Reduction, Uranium Tetrafluoride and CFD Simulations

1. Introduction

Uranium metal can be produced by a number of routes; the choice is strongly influenced by the thermodynamics of the system and the physical properties of the reactants and products. Almost all the nuclear processes employ a uranium halide as starting material. Massive uranium is produced by the thermite type reaction between uranium tetra fluoride (green salt) and magnesium. The reduction of uranium tetrafluoride (UF_4) by magnesium (Mg) is one of the main industrial methods for producing commercial pure uranium ingot.

2. Literature

Magnesium is used as a reactant for converting uranium fluoride to uranium in bomb reductions. Such reductions involve preheating the mixtures of magnesium-uranium fluoride reactants prior to initiation of the reduction reaction. If thus preheated, the hot mixtures will fuse to molten products when supplied the additional heat of the reduction reaction. Even though such fusions produce high pressures of magnesium vapour, with corollary need for expensive high-pressure vessels, still the fusions are essential for separation of uranium (the product) for magnesium fluoride (the by-product).

Uranium tetra fluoride is reduced with magnesium at very high temperature in a closed reactor under an inert gas atmosphere [1]. The main reaction is

$$UF_4 + 2Mg \rightarrow 2MgF_2 + U, \quad \Delta H_{298} = -\ 83.5 \text{ kcal/g atom of uranium.}$$

The hot metal product is also very reactive chemically and therefore, must be contained in an inert reaction vessel. The lining material is used to protect the freshly reduced molten uranium metal from coming in contact with the material of construction of the reaction vessel. Furthermore, the peak temperature attained in the reactor is well above the boiling temperature of magnesium and therefore, the reaction must be carried out in a closed system. The performance of magnesio thermic reduction reactor depends upon all the input materials and the firing time. Green salt (UF_4) composition is a well recognised variable in the reduction of UF_4 and has a significant effect on firing time, bomb yields and uranium metal quality.

+ Corresponding author. Tel.; + 91-6612462258(O), 6612463258(R), Fax: +91-6612472926
E-mail address: asahu@nitrkl.ac.in, abantisahoo@gmail.com

Compactness of the reaction mixture depends on the tap density of UF_4. More the compactness faster is the heat transfer to reactant mixture and lesser is the firing time. A good quality lining should act as a good heat conductor in the preheating stage, despite itself being a bad conductor of heat. Faster the heat transfer rate, lower the firing time. However, the lining should act as a heat insulator immediately after the reaction, i.e. it should contain the heat generated during the reduction, thereby, holding the molten mass for sufficiently long time to effect a good separation of metal and slag. But moisture present in the lining material affects the firing time like the moisture present in UF_4. Presence of iron and uranium increase the conductivity of the lining material. Since Magnesio Thermic Reduction reaction is very probably initiated by the reaction of Mg vapour and UF_4, size of the Mg particle and MgO content in Mg are very important parameters for initiation of the reaction and, hence, in firing time [2]. Dissolution of Mg in the molten salt solutions lowers the thermodynamic activity of the Mg, thereby allowing Mg to be retained in solution at temperatures above the normal boiling point of the Mg (1363^0K). Thus, even if the Mg does not immediately react with UO_2, its chance of vaporization will be reduced by the said dissolution [3].

The Magnesium and uranium oxide reactants, especially the magnesium are somewhat, soluble in the molten salt solution. Such solubility serves to spread out the magnesium-uranium oxide reaction zone and even more important, the solubility of Magnesium serves to hold magnesium in solution at temperatures above normal boiling of magnesium (1363^0K). However some of the unreacted magnesium will be expected to rise to the surface of the molten salt solution and escape as vapor. In this case a temperature gradient in the molten salt solution is developed so that un-reacted, floating magnesium can exist (temperature below 1363^0K, the magnesium boiling point) at the top of the molten salt solution while molten uranium is present (temperature above 1406^0K, the uranium melting point approximately) at the bottom of the molten salt solution [3].

3. Process Description

The reaction vessel commonly used, called *bomb reactor*, is a flanged steel bomb lined with either electrically fused dolomitic lime or recycled MgF2 slag liner. Dimensions of the reactor shell (Fig.1) used for a production scale reactor in this process are chosen [1] as 0.38 m in outside bottom diameter by 1.143m in height. The shell is tapered, with the large end at the top and tapered towards the bottom. The top is flanged, so that a cover or lid can be bolted in place, the bottom consists of 0.0254 m thick plate reinforced with a high rib for extra strength. Core-ten steel or 0.0127 m thick mild steel is used for the tapered wall of the vessel. The lid is made of 0.0127m thick mild steel. Since the temperature reached during the reaction is about 1900^0 K [1], the steel bombshell must be lined with refractory material. Recycled magnesium fluoride is used as a refractory material for lining of the reactor and thus prevents the exposure of the vessel wall to high temperature generated due to exothermic reduction reaction. The very different density of molten uranium is about 17.9gm/cc and solid magnesium oxide is about 3.4gm/cc at 1423^0K.

The other possible refractory materials and reducing metals investigated so far are available in [6] and [1]. The charge consists of 86 % green salt and 14% of magnesium [1]. The average yield can be expected to be of 98.3% [6]. The vessel loaded with the charge is then placed in an electrically heated furnace and the temperature is raised to 1023^0K [5]. The bottom plate is also heated. The reduction reaction then commences raising the temperature to a level at which the uranium melts. The duration of the heating varies with the amount of the charge and dimensions of the bomb shell [6], [1].

4. Numerical Simulations

4.1. Governing equation

As convection is absent and only conduction is present in the solid domain, the temporal terms are retained in the energy equation. Considering now the unsteady state diffusion in the context of heat transfer, in which the temperature, T, is the scalar. The corresponding partial differential equation which governs the conservation of energy is [4]

$$\frac{\partial(\rho CpT)}{\partial t} = \frac{\partial}{\partial xj}\left(K\frac{\partial T}{\partial xj}\right) + Q_0 \qquad (2)$$

Where, ρ, Cp, k, $Q_{0,}$ are the density, specific heat and thermal conductivity of the solid, and the volumetric heat generation respectively. Cp and k varies with temperature T. The term on the left hand side of the above equation is the storage term, arising out of accumulation/depletion of heat in the domain under consideration. Above equation-(2) is a partial differential equation as a result of an extra independent variable (t). The corresponding grid system is shown in Fig-3.

4.2. Range of parameters

The value of time step is taken to be 0.01. The amount of heat supplied at the bottom of shell in the reduction of UF_4 by 2 moles of magnesium at 298^0K to produce 1 mole of uranium and 2 moles of liquid MgF_2 at its melting point (1536^0 K) is 7.2cal/gm mole [1]. Density, thermal conductivity and specific heat used for different materials are shown in Table-1.

Table-1: Thermal diffusivity and conductivity of components used in process. [5].

Material	Packed Density(Kg/m^3)	Diffusivity(α) (10^{-5} m^2/s)	Conductivity(K) W/m°c
UF_4	3,300	0.01382(1+0.000200T)	0.167(1+0.000542T)
UF_4	3,500	0.01382(1+0.002380T)	0.190(1+0.000387T)
UF_4 + Mg	2,900	0.02523(1+0.000320T)	0.308(1+0.00106T)
UF_4 + Mg	3,100	0.02502(1+0.000387T)	0.415(1+0.00121T)
MgF_2 recycle slag	2,120	0.01847(1+0.000063T)	0.379(1+0.00104T)

4.3. Initial and boundary conditions

The temperature values are initialized at a value of 308^0 K, the boundary conditions for temperature on the axis-symmetric wall is homogeneous Neumann for the base, where there is constant heat source the inward gradient of the temperature is specified and temperature values varying with time are specified for the right outer boundaries, from initial to 18.31 hours. For top insulated boundary homogeneous Neumann boundary condition is specified.

5. Results and Discussion

The simulations are performed using Anupravha, a general purpose CFD and Heat transfer solver. The solver is a multi-block finite volume solver for non-orthogonal hexahedral structured grids. The grid used for computations is shown in Figure no.2. Temperature contours at some time intervals are also studied.

Heat transfer in the reaction vessel occurs in stages. In the pre-ignition heat transfer, the vessel is heated in a furnace as per predetermined schedule to the ignition temperature. During this period, the heat transfer within the reactor takes place by conduction (the outside surface of the reactor is heated by radiation and convection by electrical resistance heaters).This stage is followed by ignition stage during which reduction takes place. A large amount of heat is liberated in a very short period, which causes a sudden increase of temperature .Post ignition heat transfer occurs after this.

Temperature profiles inside the reactor including lining are obtained from initial stage to the time at which reaction occurs (time = 18.3 hours).Pre-ignition heat transfer is first analyzed, during this the vessel is heated in a furnace as per a pre determined schedule to ignition temperature (T=908^0K), heat transfer within the reactor takes place by conduction. This stage is followed by ignition stage, during which reduction takes place. The heat liberated is seen by sudden shoot up of core temperature .The temperature contours follows the pattern as indicated by Harrington and Reuhle [1]. It shows that the maximum temperature varies from 600^0K to 900^0K before the reaction and then increases to 1600^0K as the reaction proceeds.

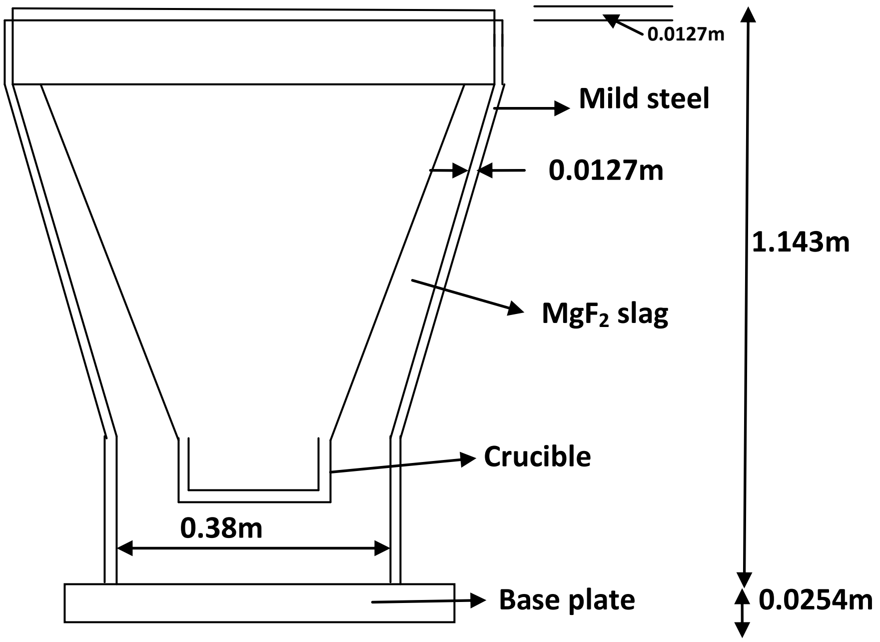

Fig.1: Geometry of MTR reactor (not to scale)

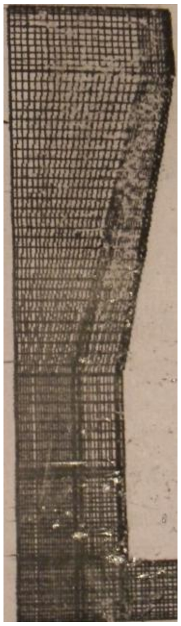

Fig.2: Grid used for computations

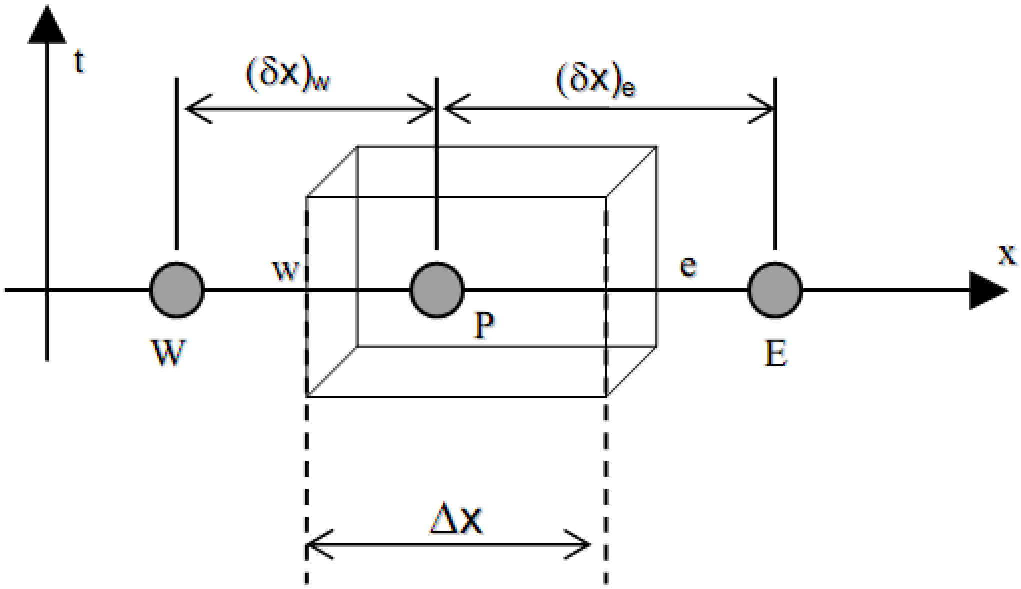

Fig.3: Grid system of an unsteady one-dimensional computational domain

6. Conclusion

The simulations are carried out for pre ignition and reaction stage, the core centre temperature is always at lower temperature than the interface. The temperature profile inside the reactor shows that after the reaction, the burning front moves through the charge at a slightly faster rate at the liner charge interface than it does in colder central portion of the bomb. The temperature values suddenly shoot up, within a very shot time interval, between 18.3 & 18.31 hours, indicating the occurrence of reaction that is highly exothermic. The maximum temperature shoots up from 900^0K to 1600^0K during the reaction. The reaction is assumed to take place instantaneously at a temperature of 908^0K. The maximum values attained are above the melting points of uranium and magnesium fluoride which shows that the product mixture will be molten.This flow of molten mass needs to be modeled. Thus the temperature profile can be anlysed further for a correct design of a Magnesio Thermic Reactor thereby preventing any leakage to the environment.

7. References

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