CHAPTER 8. Conclusion and scope for future work

8.1. Summary of the findings of the current work
Various experimental studies have been carried out as a part of the present work for evaluating the dissolution kinetics of plutonium rich FBR driver fuel in nitric acid. Due to the complexities and safety requirements involved in handling plutonium based nuclear fuel, a systematic approach was undertaken by first studying the dissolution kinetics of urania followed by urania ceria MOX fuel with cerium acting as a non-radioactive surrogate for plutonium. The results obtained for these two types of fuel pellets paved way for better designing of experimental techniques, equipments and modeling procedures in thorough understanding of the dissolution behaviour of urania plutonia MOX pellets.

8.1.1 Major findings of UO\textsubscript{2} fuel pellet dissolution studies in nitric acid
The following are the major findings with respect to urania fuel dissolution behaviour under typical PUREX process conditions based on the current work.

1) The dissolution of UO\textsubscript{2} in nitric acid medium under typical PUREX process condition is chemical reaction controlled with the activation energy typically of the order of 90 kJ/mol

2) Sodium hydroxide scrubbers coupled with ion chromatography is a simple and efficient technique to determine the composition of the NO\textsubscript{X} gases evolved during the dissolution of UO\textsubscript{2} fuel in nitric acid.

3) From the concentration profiles of the NO\textsubscript{X} gases in the off-gas stream and nitrous acid in the bulk liquid evolved during dissolution, the mechanism for the dissolution of UO\textsubscript{2} fuel under typical PUREX process condition was determined.

4) Nitrous acid plays an autocatalytic role during the dissolution of UO\textsubscript{2} fuel in nitric acid. Thus due to the dispersion of nitrous acid away from the pellet-nitric acid interface under agitated condition, retards the rate of dissolution of UO\textsubscript{2} pellets proportionately.

5) Shrinking particle model explains the dissolution behaviour of UO\textsubscript{2} fuel in nitric acid satisfactorily. The model equations developed based on this method with diffusion effects coupled with chemical reaction predicts the concentration profiles of all the important reactant and product species accurately.
6) Since agitation of the dissolving mixture is essential to improve the effective surface area of the pellets, addition of NO$_2$ gas into the reaction mixture is a simple and easy way to improve the dissolution kinetics of UO$_2$ spent fuel in nitric acid. This would also lead to better production rates in thermal reactor fuel reprocessing plants.

7) The order of the reaction with respect to nitric acid, rate constant and the validated model rate equations developed in the present work would be useful tools in designing a continuous dissolution system for processing urania based nuclear fuels.

**8.1.2 Major findings of simulated MOX fuel pellet dissolution studies in nitric acid**

The following are the major findings with respect to urania ceria MOX fuel dissolution behaviour under typical PUREX process conditions based on the current work.

1) Method of fabrication of the MOX pellets influences their dissolution behaviour in nitric acid to a great extent.

2) Addition of cerium to urania during the fabrication of urania ceria MOX pellets renders the pellet more difficult to dissolve under typical Purex process conditions.

3) The composition of cerium in the pellets is inversely related to the rate of dissolution of the MOX pellets in nitric acid.

4) Both the penetration and surface area dissolution model equations based on shrinking particle theory explains the dissolution behaviour of urania ceria MOX pellets satisfactorily.

**8.1.3 Major findings of U, Pu MOX fuel pellet dissolution studies in nitric acid**

The following are the major findings with respect to urania plutonia MOX fuel dissolution behaviour under typical PUREX process conditions based on the current work.

1) The qualitative dissolution behaviour of urania plutonia MOX pellets in nitric acid is comparable to that of its urania ceria counterpart of same composition.

2) Thus the choice of cerium as a non-radioactive surrogate of plutonium for studying the dissolution behaviour of MOX pellets in nitric acid is justified.

3) Though the presence of nitrous acid does not chemically alter the dissolution rate of plutonia directly, it increases the rate indirectly by catalytically
dissolving urania faster and thereby making the pellet more porous. This leads to better contact between the dissolving pellet and nitric acid enabling faster dissolution of the rest of the pellet.

4) The penetration based dissolution model equations coupled with shrinking particle theory predicts the concentration profiles of the various species involved in the reaction more accurately than the surface area based method.

5) The lack of a suitable analytical system presently for the surface area measurement equipment of radioactive samples is the reason for the reduced accuracy of the surface area based model equations in predicting the dissolution behaviour of urania plutonia MOX fuel dissolution in nitric acid medium.

6) The order of the reaction with respect to nitric acid, rate constant and the validated model rate equations developed for the dissolution of urania plutonia MOX fuel would be useful tools in designing a continuous dissolution system for FBR fuel reprocessing plants.

8.2. Scope for future work

8.2.1 Surface area measurement for radioactive samples
The surface area of the dissolving pellets is an important parameter which influences the rate of its dissolution as explained in this thesis. For the studies on the MOX fuel dissolution, due to lack of a suitable analytical facility to measure the surface area of plutonium based pellets, only its initial surface area calculated from its dimensions were used. Thus there is a plenty of scope in setting up a facility for measuring the surface area of radioactive solids which will facilitate the measurement of surface area of any radioactive solid during the course of its dissolution accurately. This data would improve the accuracy of the model equation which would auger well for designing a continuous dissolver more efficiently.

The results of the MOX pellet dissolution clearly brings out the preferential leaching of UO₂ over PuO₂ in spite of the fact that the MOX pellets are complete solid solutions of urania and plutonia. This is a classic example for the sluggish behaviour of plutonia towards dissolution in nitric acid. Measure of surface area of the dissolving pellet without any ambiguity would go a long way in understanding the influence of lattice collapse mechanism during dissolution of urania and plutonia which could help in fine tuning the
understanding and prediction of the MOX pellet dissolution in nitric acid.

8.2.2 Dissolution experiments on radioactive spent fuel samples
All the experiments carried out as a part of the present work employed only unirradiated pellets. Since the irradiated pellets are expected to dissolve rapidly due to its higher porosity caused by the diffusion of the gaseous fission products produced inside its matrix during irradiation, the prediction based on the current model equations would be more conservative than the actual spent fuel dissolution. Hence for improving the model equations developed through the current work, dissolution experiments are required to be carried out using irradiated fuel samples under typical PUREX process conditions. As this requires dedicated shielded facility with provisions for sampling and remote analysis, this could be an important work to be carried out in the future.

8.2.3 Dissolution of MOX fuel with varying plutonium composition
The effect of plutonium composition on the dissolution behaviour of MOX pellets in nitric acid could only be qualitatively predicted based on the dissolution behaviour of simulated urania ceria MOX pellets. Hence for studying the quantitative effect of plutonium composition on the MOX fuel dissolution kinetics, MOX pellets with varying plutonium compositions are required. Such pellets are currently unavailable as it requires special facilities for their fabrication. Thus it becomes an important and necessary work for the future to determine the existence of any unusual dissolution behaviour of MOX pellets with varied plutonium compositions before and after irradiation. These studies would lead to the development of the continuous dissolution process system which is the need of the hour for designing energy efficient, high throughput and safer FBR fuel reprocessing plants.