

SUMMARY AND CONCLUSION

"Management of the Waste Produced from Yellow Cake Purification"

The main objective of the work presented in this thesis is to develop selected process for removal of uranium from the raffinate solution produced during purification of the yellow cake process, to reach the lowest possible content of uranium in the waste solution. Within these objectives, two approaches were investigated based on chemical precipitation and solvent extraction. The different parameters affecting the removal of uranium from this raffinate solution by precipitation or solvent extraction were studied. Further the best conditions for uranium removal were obtained and assessed. The present work in this thesis, is classified into three chapters. These are presented as follows.

Chapter-1, Introduction:

This chapter includes a brief account on; chemicals and radioactive contaminants as a general statement, nuclear fuel cycle, yellow cake production and purification, some aspects of the nuclear properties and aqueous chemistry of uranium. It contains an outline of radioactive waste and its management, physical classification of radioactive wastes, sources and quantities of radioactive waste from nuclear activities. The main separation methods for decontamination of radioactive waste with more focusing on chemical precipitation and solvent extraction were addressed. The aim of the present work and literature survey which is related to the present work is also given.

Chapter-2, Experimental:

This chapter includes all chemicals used in the course of this work with their chemical purity and reagents used in the experimental work. It includes detailed description of the different instruments used, which are: Atomic Absorption Spectrometer, Environmental Scanning Electron Microscope, UV-Visible

Spectrophotometer. This chapter also contains methods of determination of solid content, nitric acid concentration, density, iron concentration, and uranium concentration. Details of experimental procedures such as precipitation of the hydroxides, carbonate leaching, precipitation cycles, carbonate precipitation / UO_2^{2+} complexation, extraction process, stripping process, and equations used for determination of the uranium precipitation percent, uranium recovery percent, uranium extraction percent, and uranium stripping percent.

Chapter-3, Results and Discussion:

This chapter includes the experimental results obtained and their interpretation. This chapter is classified into three main parts; namely, characterization of waste solution, separation of uranium from waste solution, and recommended procedure for UO_2^{2+} recovery.

Part One (Characterization of Waste Solution):

Main physical and chemical characteristics of raffinate solution was investigated such as color, density, solid content, nitric acid concentration, uranium concentration, iron concentration, and calcium concentration. Also main and minor elements present in raffinate solution were evaluated using Environmental Scanning Electron Microscope (ESEM).

Part Two (Separation of Uranium from Waste Solution):

This part contains the procedures used to separate uranium from raffinate waste solution and the optimum conditions from results obtained from three applied procedures. It includes three sub-section, as follows:

Section One (Precipitation / leaching investigations):

This section includes hydroxide precipitation and the effect of pH during UO_2^{2+} precipitation on its recovery. It includes carbonate leaching of UO_2^{2+} from precipitate and the parameters affecting uranium recovery such as effect of Na_2CO_3 concentration, effect of mixing period, effect of Na_2CO_3 volume added, effect of temperature of Na_2CO_3 , effect of number of precipitation cycles, effect of other leaching reagents, and repeated leaching cycles.

Finally, the specification of recovered uranium was carried out which illustrated that the main and minor elements accompanied the recovered uranium are K, Na, Ca, Ra, Bi, Si, Mg, Al, S, and P. The best conditions obtained from this section to recover uranium from 50ml raffinate are:

80 ml of 1M Na_2CO_3 at 90°C , 3 minutes as mixing time, pH of precipitation = 5, and two precipitation cycles which gives total recovery of 97.6 % after the third leaching cycle with uranium recovered purity of around 78 %.

Section two (Carbonate precipitation and UO_2^{2+} complexation):

Several parameters were studied such as effect of Na_2CO_3 concentration, effect of mixing period, effect of Na_2CO_3 volume added, effect of temperature of Na_2CO_3 , and effect of number of precipitation cycles. Some trials were investigated to improve UO_2^{2+} recovery such as number of washing times and volume reduction. Specification of recovered uranium was carried out which illustrated that the main and minor impurities accompanied the recovered uranium are K, Na, Ca, Si, Mg, Al, Ni, Mn, and Fe.

The best conditions obtained from this section to recover uranium from 50 ml raffinate are: 50 ml of 2M Na_2CO_3 at 90°C , 1 minute as a mixing time, and two precipitation cycles which gives total recovery of 98.54 % after the third washing time and uranium purity of 82 %.

Finally, the comparative recovery of UO_2^{2+} between the two procedures was carried out.

The results obtained in the aforementioned sections were discussed in terms of the different uranium hydroxyl and carbonate species present in solutions.

Section three (Solvent extraction investigation):

This section include the studies of extraction of uranium from acidic raffinate waste solution, stripping of uranium from the loaded organic phase by sodium carbonate, and water.

Extraction of uranium from acidic waste solution by TBP was studied. Many parameters were investigated such as effect of TBP concentration, effect of contact time, effect of temperature, and effect of aqueous/organic phase ratio. The best conditions obtained from this section for uranium recovery are: 20% TBP in kerosene, contact time of 1 minute, and phase ratio equal unity which gives total extraction of 95 %. EDX analysis of the precipitated raffinate after extraction of uranium indicated that uranium is present as minor amount by 3.31 % which represents the 5 % remained unextracted ,75ppm. Uranium extraction percent of A/O phase ratio from 1 to 10 and 1 to 5 extraction stages were calculated theoretically, and found to be similar to the experimental results. The experimental results indicated that the required extraction can be obtained from theoretical calculations with a good accuracy.

At study of stripping of uranium from loaded organic phase by sodium carbonate was carried out. Different parameters were studied such as effect of sodium carbonate concentration, effect of phase ratio O/A, effect of contact time, and effect of temperature. The optimum conditions obtained from these results are the following;

0.3 M Na_2CO_3 at the ambient room temperature ($25\pm 1^\circ\text{C}$), 1 minute as a contact time, and phase ratio equals 3, which gives total stripping of $> 99.9\%$. The specification of uranium stripped by sodium carbonate was investigated using EDX analysis of precipitated uranium stripped indicated that uranium is present as a major amount constituent with a purity of 90.77 %.

A study of stripping of uranium from loaded organic by water was carried out. The parameters studied were effect of phase ratio O/A, effect of contact time, and effect of temperature. The optimum conditions obtained from this section are:

at 70°C, 1 minute as a contact time, and phase ratio equals unity, which gives total stripping of more than 99.9 %. Specification of uranium stripped by water was investigated using EDX analysis of precipitated stripped uranium which indicated that uranium is present as a major amount with purity of more than 92 %.

Part Three (Conclusion of General Recommendation):

The present work indicated that recovery of uranium from waste produced from yellow cake purification can be successfully achieved by two main routes. The first is based on hydroxide precipitation-carbonate leaching and the second is based on liquid-liquid extraction.

The first method is characterized by simple precipitation and leaching of uranium by sodium carbonate. The best conditions for this process recovered more than 98% of uranium in the raffinate solution with a purity reached more than 80%. The main advantage of this method is that it is a simple and economic technique. However, this process is most applicable on batch wise treatment.

The second method is based on liquid-liquid extraction, where uranium recovery can reach 99% with a purity of more than 92%. The main advantage of this process is that it can be operated continuously. This is of high benefits if increased amounts of raffinate are produced and can be connected to the circuit of yellow cake production process. However, the process needs more chemicals and it is more expensive than the precipitation process.