COMPARATIVE STUDY OF TURBO AND SPRAY DRYING TECHNIQUES IN THE PRODUCTION OF NUCLEAR GRADE URANIUM DIOXIDE

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ABSTRACT

Natural Uranium Di-Oxide (UO_2) powder for the manufacturing of fuel for Pressurised Heavy Water Reactors (PHWR) is produced through the Ammonium Di-Uranate (ADU) route. The characteristics of the virgin UO_2 powder influence the sintered density of UO_2 pellets, which in turn is decided by the physical state of the starting material ADU. The physical characteristics such as the morphology, particle size, particle size distribution of ADU depend a lot on the precipitation and drying conditions. Different modes of drying are utilised on industrial scale to obtain sinterable grade UO_2 powder. Nuclear Fuel Complex, Hyderabad has successfully developed both the spray and turbo drying techniques for the production of dry ADU for further thermal treatment.

Turbo drier is basically a bulk drier where the mechanism of drying is 'cross and through circulation'. The spray drier is a micro drier in which the slurry is atomised in a hot gas stream inside a chamber where drying takes place within a few seconds, because of high specific surface area. Design and operational parameters for the spray drier were optimised based upon the comparative study of product quality. Scanning Electron Microscope(SEM) images of ADU and UO_2 produced by turbo and spray dried routes were analysed.

This paper deals in detail with the comparative studies carried out on both drying techniques, along with their behavior on further processing steps such as calcination, reduction, stabilization and sinterability of UO_2 powder.

1.0 INTRODUCTION

The ADU route is followed at Nuclear Fuel Complex(NFC), Hyderabad for the production of UO_2 powder for the PHWRs [1]. The first solid phase intermediate, ADU, is formed at the precipitation step of the production process. The ADU precipitate cake is dried and then subjected to further thermal treatment process of calcination, reduction and stabilization for the production of nuclear grade UO_2 powder meeting all the required physical, chemical and sinterable characteristics.

The most important process step in the above mentioned flow sheet is found to be the precipitation, where the final UO_2 powder particle characteristics as well as the bulk solids behavior is decided. The individual particle characteristics decide the sinterability, where as the bulk solids and powder behaviour, including agglomerate quality is crucial to the compaction process.

The ADU cake obtained from the precipitation and subsequent filtration has to be dried before further thermal treatment. The type of drying decides the type of agglomerate formed, the presence and fraction of hard agglomerates if any, residual moisture & nitrate contents, the bulk density of the ADU powder and flowability.

2.0 PRINCIPLE OF DRYING

Thermal drying of wet solids is the process of removal of free moisture present in the solids by evaporation [2, 3].

The nature of the solids and accompanying moisture determines the drying characteristics. Thus, when a wet solid is subjected to thermal drying, the following two processes occur simultaneously.

A) Transfer of energy from the surrounding environment to evaporate the surface moisture.
B) Transfer of internal moisture to the surface of solid (diffusional and capillary flow) and its subsequent evaporation due to process A.

The rate at which drying is accomplished is governed by the rate at which the above two processes proceed.

3.0 EQUIPMENT

At NFC, during the development and initial setting-up of the ADU process, its standardization and subsequent scale-up, various types of driers have been used, starting with the band drier. The turbo drier, which was introduced to replace the band drier was chosen due to the various advantages like superior quality of dried ADU with respect to consistency, absence of hard lumps/agglomerates, improved flowability, etc., and also because of the added advantage of 'closed' operation which has eliminated the air-borne activity in the shop floor.

In order to further improve the homogeneity and flowability of the ADU powder so as to make it amenable to pneumatic conveying for further processing step, spray drying presented attractive alternative. Thus, in the new UO_2 powder production plant, spray drier was installed instead of turbo-drier, after carrying out laboratory trials .[4].

Turbo Drier : Turbo drier is a high temperature (250 - 300° C)stainless-steel heat transfer unit, designed with a stack of circular trays rotating slowly inside the drying chamber [3]. Turbo-fan assembly is mounted on a central shaft, co-axial to the tray drive and is driven by another motor. ADU cake containing 20-30% moisture is fed to the top tray. Each tray consists of a leveller and a scrapper. After one complete rotation of the tray, a stationary scrapper scrapes off the powder, which falls through the radial slot, on to the next tier of tray in a plug flow manner. The pile formed here is then levelled evenly by a leveller. Thus, in the turbo-drier, the material travels both horizontally as well as vertically. During its horizontal travel, the dry air flows over it in a direction opposite to its own movement. In the vertical motion, the product falls down to the tray below and encounters dry air flowing in the countercurrent direction. The dry powder is collected at the bottom of the unit into a hopper which is later used as the feed hopper for the calcination furnace. The exhaust from the Turbo drier is passed through cyclone separator &wet scrubber before it is let out through a stack.

Spray Drier : The spray drier consists of a drying chamber into which the ADU slurry containing about 40-50% solids is admitted. The slurry is transformed into fine droplets by the atomizer rotating at high speed under the action of the centrifugal force. The droplets come in contact with the hot flue gases from the combustion chamber where Liquid Petroleum Gas (LPG) fuel is burnt. The increased surface area of the atomised droplets allows rapid heat transfer and quick evaporation of moisture. The dry ADU powder is separated from the exhaust gases in a cyclone separator and the fines (<5 microns) are collected in a bag filter installed before exhaust blower. The dry ADU is collected into hoppers placed at the bottom of the cyclone. These are later used as the feed hoppers of the calcination furnace. The entire system runs under negative pressure with the help of the exhaust blower. The exhaust gases pass through pre-filter and High Efficiency Particulate Arrestor (HEPA) filter before being let out through a tall stack.

4.0 COMPARATIVE STUDIES

The first solid intermediate produced in the production process is ADU, at the precipitation stage. 'Drying' follows the precipitation and filtration. The ADU after subsequent thermal treatment will yield UO_2 of required characteristics.

The turbo drying process was well established at NFC to produce dry ADU, which yielded consistent sinterable grade UO_2 powder. The newly installed spray drier was, however, expected to yield an ADU with different physical characteristics from that of turbo-dried ADU. Hence suitable process optimization in the subsequent thermal treatment cycles of calcination, reduction and stabilization would be necessary, so as to produce UO_2 powder of the same sinterable quality. This need prompted a comparative study of the properties of spray and turbo dried ADU at production scale so that the process parameters of the down-stream operations could be optimised for spray dried ADU.

Table-1 gives the comparison of physical, chemical and morphological properties of turbo and spray dried ADU produced at NFC. SEM pictures of ADU and UO_2 powder produced from two different drying techniques are shown in Figure 1 and 2.

5.0 OBSERVATIONS AND DISCUSSIONS

The drying temperature and residence time in the turbo drier is optimized so as to meet the specification with respect to moisture and nitrate content. The conditions are such that both the following processes occur :

a) Evaporation of the unbound and also bound moisture.
b) Heating of the dry solid, resulting in conversion of the ADU to UO_3 partially.

The output from the turbo drier is thus, (ADU+UO_3) and not dry ADU alone [5] The SEM picture of turbo-dried ADU in Fig.1 (a, b) is that of ADU + UO_3. The figure depicts, agglomerates of needle like structures with a lot of inter-particle porosity. Each agglomerate is, rounded in shape. The agglomerate nature and the needle like structures are retained even after the three subsequent thermal treatment steps of calcination, reduction and stabilization, as seen in Figure 2 (a, b), which is an SEM picture of UO_2 produced with turbo dried ADU.

In the spray drier, the ADU slurry droplets are dried at a temperature of 120° - 140° C within a few seconds. The ADU particle does not see higher temperature since the conditions are suited only for evaporation of free moisture and thus, there is no scope for conversion of ADU to UO_3. Figure 1 (c, d) gives the SEM picture of spray dried ADU particles. The agglomerates in this case are made up of spherical particles held together loosely with a lot of inter-particle porosity. However, the SEM picture in Fig.2 (c, d) showing agglomerates of UO_2 produced from Spray dried ADU have similar characteristics as that of UO_2 produced from Turbo dried ADU. Thus, subsequent thermal treatments seem to change the shape of particles within the agglomerate.

Comparison of properties of Turbo and Spray Dried ADU [6] :

Flowability : Good flowability of ADU powder is important for the next step of calcination to ensure controlled feeding. Good flowability of UO_2 powder resulting from such an ADU improves compactability. Spray dried powder is more flowable than turbo dried ADU and thus more amenable to vacuum / pneumatic conveying in closed conduits and controlled feeding during Calcination.

Agglomerates quality : Soft agglomerates in the ADU powder retain their softness up to final stage, provided down-stream thermal processes are carried-out under controlled conditions. Soft UO_2 powder is very advantageous for compaction. UO_2 powder from both the drying processes are found to be equally compactible, implying that ADU agglomerates from both the drying processes have similar softness.

Porosity : Inter-particle porosity in the ADU agglomerate aids smooth escape of dissociation products during the conversion to UO_3 / U_3O_8 in calcination and formation of UO_2 during reduction stage. Porosity of ADU was not measured in these studies. However, similar characteristics of UO_2 powder from these ADU and consistently high UO_2 pellet densities in the range of 10.55 - 10.65 g/cc achieved with UO_2 powder from both the drying processes, indirectly indicates that porosity in both the ADU powders is similar in nature.

Moisture content : The residual moisture content in the dry ADU plays a role only in the flowability of the powder being fed into the calcination furnace. It does not have much effect on calcination process. Though spray dried powder has relatively higher moisture content, its higher flowability was not affected, except when aged in the feed hopper.

Nitrate Content : The nitrate content in the dry ADU is an important quality control parameter due to its suspected role in the formation of hard particles during the calcination process, which eventually results in pits formation in the UO_2 pellets. In spray dried ADU, nitrate was found to be relatively higher than in turbo dried ADU. This is mainly due to higher temperature in Turbo drier as compared to Spray drier. However, hard agglomerates could be completely avoided in the U_3O_8 and subsequently in the final UO_2 powder, via spray dried ADU, because of inherent homogeneity of spray dried powder and by suitably modifying the calcination process parameters.

Bulk density_: Bulk density of the spray dried ADU is slightly lower than that of turbo dried ADU, resulting in lower bulk density of UO_2 powder. However, this has marginally affected only the pre-compaction step. Bulk density increase of ADU, if necessary, can be achieved by various means in the spray drying operation. It can also be changed by effecting changes in the precipitation process, which is more preferable. This aspect is being studied at present.

6.0 CONCLUSIONS

Precipitation of ADU is the most critical step in the production of sinterable UO_2 powder, as the basic characteristics of the particles are formed during precipitation itself. Both, turbo and spray drier, have yielded dry ADU of required quality, which, with suitable adjustments of parameters in the down-stream thermal processes have produced sinterable grade UO_2 powder.

The contribution of the drying process towards achieving final quality of UO_2 powder is limited to the formation of suitable agglomerates. The actual method of drying could be turbo or spray drying as long as homogeneous ADU powder with soft agglomerates, having sufficient inter-particle porosity are produced at precipitation stage. Uniform heat transfer should be ensured during drying in order to produce homogeneous product without hard particles. These characteristics can be retained through the calcination, reduction and stabilization steps by modifying the process parameters slightly, so that flowable, compactable and sinterable grade UO_2 powder results.

Both the driers have merits and demerits. The turbo drier is a sturdy equipment and can operate for very long periods without major maintenance (the present unit at NFC has operated continuously for more than 4 years). Turbo drier has added advantage of handling relatively very low quantity of air, which is a significant factor in case of drying of radioactive materials such as ADU. However, in case of need for maintenance, the high hold-up of radioactive material within the drier poses a major disadvantage.

On the other hand, in the Spray drier, the atomiser, a critical component, operates at very high speed and is prone to frequent maintenance. However, since the Spray drier does not have much hold-up of radioactive material, maintenance is easier.

Considering the above advantages, its low capital cost and higher operational convenience, turbo dryer seems to be a better drying equipment for ADU. At the same time, since our operating experience with Spray dryer is limited, for detailed comparison and final conclusion, Spray drier operations need to be studied for a longer period.

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TABLE 1 : COMPARATIVE STUDY OF SPRAY AND TURBO DRIED ADU

	Turbo Dried - ADU	Spray Dried - ADU
I. PHYSICAL PROPERTIES		
• PSD	5 - 75 microns With max. % between 10 - 20 microns	5 - 45 microns With max. % between 10 - 17 microns
• FLOWABILITY (50 gm through 10 mm Hall's flow meter)	20 secs.	5 secs.
• BULK DENSITY	1.3 - 1.6 g/cc	1.0 - 1.2 g/cc
II. CHEMICAL PROPERTIES		
• NO_3	1.0 - 2.0%	4.5 - 5.0%
• MOISTURE	< 1.0%	1.0 - 2.0%
III. MORPHOLOGICAL PROPERTIES		
• Agglomerate Shape	Predominantly rounded, with a small percentage of flaky platelets	Homogeneous and uniform agglomerates, with rounded off edges
• Porosity	Large percentage of open pores	Lot of inter-particle pores
• Particles in the agglomerate	Needle like particles	Spherical particles

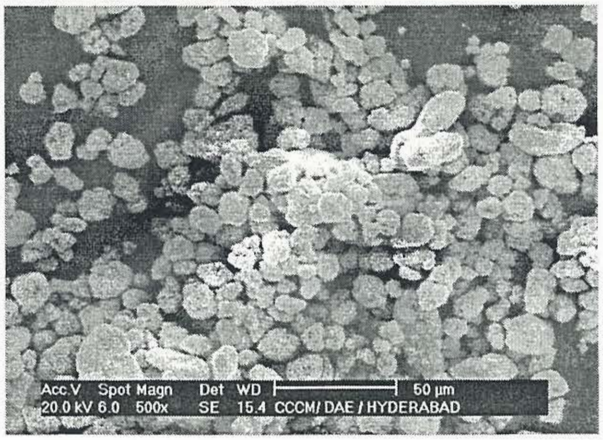

Fig.1 a. Agglomerates shape and size distribution of Turbo dried ADU

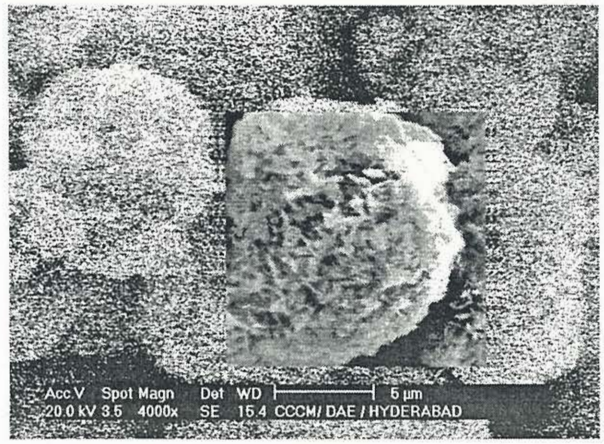

Fig.1 b. Enlarged view of single agglomerate showing needle like particles with large porosity

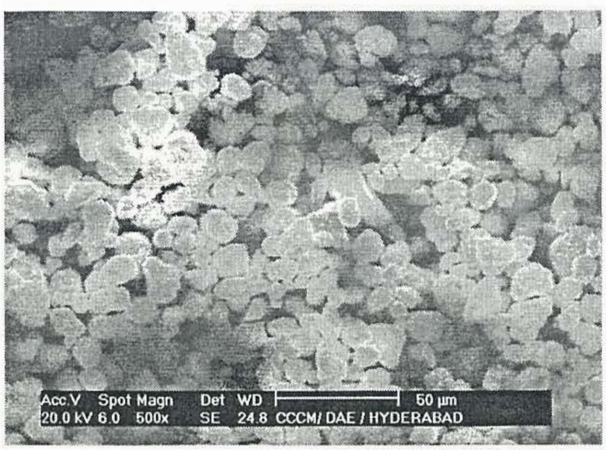

Fig.1 c. Agglomerates shape and size distribution of Spray dried ADU

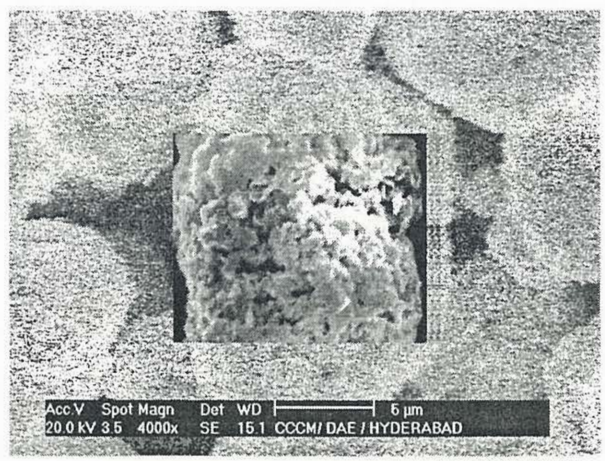

Fig.1 d. Enlarged view of single agglomerate showing spherical like particles with large porosity

FIG 1: SEM PICTURES OF ADU PRODUCED THROUGH TURBO AND SPRAY DRIERS

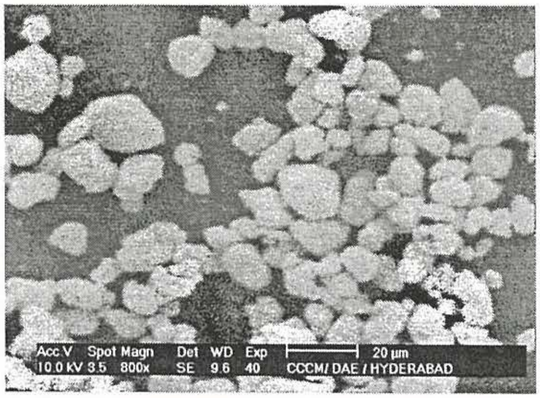

Fig.2 a. Agglomerates shape and size distribution of UO2 produced via Turbo dried ADU

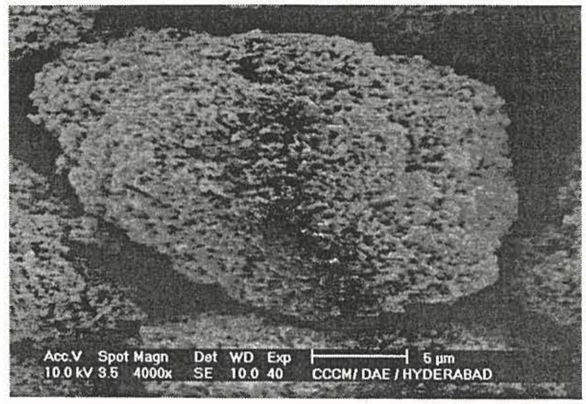

Fig.2 b. Enlarged view of single Agglomerate of UO2 produced via Turbo dried ADU

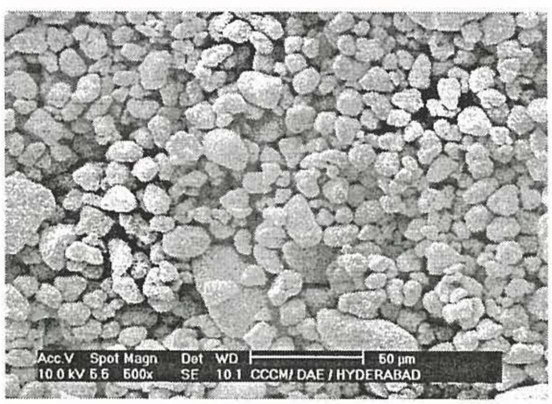

Fig.2 c. Agglomerates shape and size distribution of UO_2 produced via Spray dried ADU

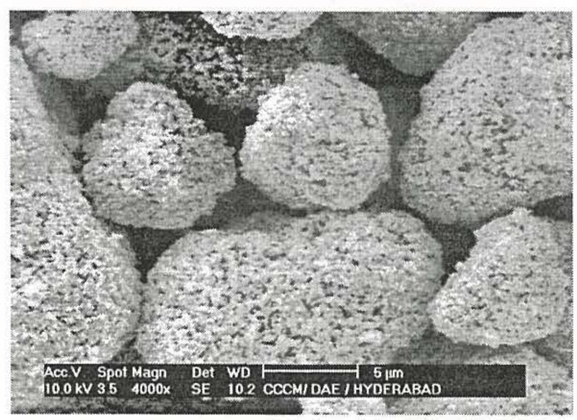

Fig.2 d. Enlarged view of single Agglomerate of UO2 produced via Spray dried ADU

FIG 2: SEM PICTURES OF UO2 PRODUCED THROUGH TURBO & SPRAY DRIED ADU