EFFECT OF LOADING PATTERN ON HIGH TEMPERATURE SINTERING OF UO_2 PELLETS

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ABSTRACT

For high temperature sintering of UO_2 pellets different methods of arrangement of cold-compacted pellets are followed all over the world. The paper discusses the different loading patterns of pellets in molybdenum charge carrier. Experiments were conducted by charging green UO_2 pellets horizontally, vertically and randomly in the charge carriers, after sintering the same under identical conditions of temperature and pressure in a pusher type continuos sintering furnace , the sintered density and the defects observed in UO_2 pellets were studied.

1.0 INTRODUCTION :

The sintered density of UO_2 fuel pellets specified for PHWR application is maintained in the range of 95 to 98% of theoretical density. The variation in density not only depends on the powder characteristics like specific surface area, particle size distribution and oxygen to uranium ratio but also on the temperature profile of the pusher type furnace, the furnace atmosphere and on the arrangement of green pellets in the charge carrier.

A study has been made to understand the density behavior as an offshoot of the experience obtained during the increase in charge in the boat for one of the operating furnaces. This charge increase was done to increase the throughput .The furnace parameters remained unchanged and only charge was increased from 12.5 kg to 25 kg per boat of green compacts. After sintering the boat with the increased charge exhibited bloating of pellets these defective pellets were located in the bottom central portion of the carrier. This generated a good amount of interest to conduct certain experiments to analyse and solve the problem.

2.0 SINTERING EXPERIENCE :

Though the temperature profile of the sintering furnace is a dominant parameter other important factors include the powder particle size distribution, applied pressure in compaction, sintering time, heating rate and furnace atmosphere.

Various attempts were made to overcome the variation in sintered density for the same powder lot.

* Increase the main flow rate of cracked ammonia gas counter-current to the movement the Charge carriers.
* Purge the gas through the shell of the furnace.
* Charge pellets longitudinally instead of transverse to the direction of travel in the boat.
* Maintain a higher incoming pressure of NH_3 to the dissociator.

With the above changes a marginal improvement was noticed in the reduction of bloating of pellets.

Bloating of the pellets is normally due to the following reasons.

2.1 High surface area of the UO_2 powder.
2.2 Oil mix in powder.
2.3 Faster rate of heating.
2.4 Solarization.
2.5 Large pore size in green compacts.
2.6 Inhomogenity in admixed powder.

2.1 High surface area in the powder causes the shell of the compact to get sintered first resulting in closure of porosity on the surface of the sintered compact and thereby preventing the gases inside the pellet from coming out. At higher temperatures this leads to bloating of the pellets due to increasing gas pressure within them[1].

2.2 The mix up of oil that leaked from hydraulic presses with the powder may take place sometimes during pre-compaction stage. The green compact produced has small pockets of heterogeneously distributed hydraulic oil. During sintering, it is essential that the soaking time at 400-800° C be maintained for sufficiently long period for the oil to volatilize. Shorter residence time at the above mentioned temperature will result in bloating of pellets.

2.3 The faster rate of sintering of the green compact is the same as described for high surface area UO_2 powders.

2.4 The effect of Solarization takes place when the pellet is subjected to repeated sintering, leading to a bloating of the pellets due to the expansion of occluded furnace gases.

2.5 The size of the porosity in the green compact has a large bearing on the bloating of the pellets. The size of the porosity depends on the size and strength of the granules/particles, which do not break on application of pressure during final compaction and result in large sized pores[2].

2.6 Addition of solid lubricant can also cause bloating, if the solid lubricant is not uniformly blended with UO_2 powder[3].

The bloating observed in pellets passed in a six-zone furnace was a local effect, which repeated, in a similar fashion for a large number of charge carriers, narrowing down to two reasons.

- Dependence in the furnace parameter like flow rate /temperature profile/ preheat temperature.

- Type of loading green compacts in change carriers.

Pattern of charging green compacts is horizontal as shown in the FIG.3. The charge was increased to twice the quantity from 12.5 Kg. to 25 Kg. Bloating of pellets was in the bottom central portion region of the charge.

Since the similar defect was not observed in the 12.5-Kg charge it could be correlated to the increase in charge to 25Kg.

This analysis narrowed down to the sintering behavior for different charging patterns dictating the gas flow behavior. Three different patterns of charging were attempted.

- Vertical charging.(FIG.1 & 2)
- Hexagonal closed packed structure charging. (FIG.3 & 4)
- Random charging.(FIG.5 & 6)

Single lot produced through the batch precipitation route was taken having the following characteristics.

SSA = 2.82 m^2/g ; O/U = 2.04

The lot was first pre-compacted and the parameters maintained are as follows.

Pressure -- 125 MPa.; Dwell time -- 7 seconds
Bulk density of granules -- 2.58 g/cc

The granulated powder size -- -10# & +60 # fraction collected was taken for final Compaction at the following parameters.

Pressure -- 325 MPa.; Dwell time -- 7 seconds

The green pellets produced under the above similar conditions were subjected to the above three different types of loading patterns. The three charge carriers were passed in the same sintering furnace with the following temperature profile 400\800\1200\1400\1700\1700\1700 °C Each zone was having a length of 0.9 meter and the pushing interval was maintained at 1 hr.

For density measurement samples from different location were taken. More stress was put for sampling for the bottom layer in the central portion.

3.0 EXPERIMENT ON LOADING PATTERN

The natural UO_2 powder produced through the ADU route with the acceptable powder characteristics was taken for experimentation.

The Powder was precompacted in a 200 ton Hydraulic press applying a pressure of 125 MPa. The cakes produced were granulated in an oscillatory granulator and sieved for the separation of fines. The graded fraction i.e. -10 # and +60# fraction was the input to produce green compacts with density range of 52 to 55% of TD.

The UO_2 green compacts arranged in three different patterns in molybdenum charge carrier were passed in seven zone sintering furnace with a reducing atmosphere of cracked NH_3. The temperature profile maintained in the furnace gives a heating rate of 150°C/ hr. The highest temperature maintained in the furnace is 1700°C with a residence time of 9 hours at the highest temperature.

All the charge carriers are sintered at the above temperature profile and subjected to same gas flow rate of cracked NH_3.

The boats formed a continuous chain and were passed at 1 hr pushing interval into the furnace. Boats discharged from sintering furnace after densification for vertical, horizontal, random are as per FIG.2, FIG.4, and FIG.6 respectively.

The sintered pellets collected at specific location in the charge carriers were centreless ground and the geometrical density was measured. The dimensions were taken in laser measuring device and the weights were taken on electronic balance with 0.001gram accuracy.

The computation of the density was done on line with the help of electronic balance.

4.0 DISCUSSIONS

4.1 Density Distribution

The density measured for three different types of charging is given in the table 1. It can be seen that the density values achieved in random and vertical type of charging showed acceptable range of values above 95% TD whereas horizontal charging has shown values as low as 91% TD. The deviation from the mean was also very high and the sigma calculated was three times as high compared to the vertical and horizontally charged.

The quantity of pellets in horizontal charging for the same dimension of charge carrier is higher by 25% when compared to vertical & random charging.

The gap between the pellets loaded horizontally decreases progressively as the sintering takes place whereas an opposite affect will be experienced in vertical charging wherein the gap between the pellets increases on sintering.

The decrease in gap increases the resistance for the flow of cracked ammonia, which retards the sintering rate for horizontally, charged pellets.

The parallel gap between the pellets in horizontal charging is dependent on the diameter of the pellet and is given by the equation - 1

$$\text{Inter pellet gap in horizontal charging} = (2\sqrt{3} - \Pi)\, d^2/8$$

Where d = diameter of the pellet.

During sintering 92% of the theoretical density is obtained at 1400°C temperature zone, further densification takes places in the 1700°C zones. The shrinkage at the above theoretical density of 92% is the order of 18% on diameter.

The decrease in diameter reduces the inter pellet gap by a factor of 0.67 to the flow of gas . The reduced area in the parallel path resistance for the flow of gas thereby reducing the heat transfer by convection to the pellets.

This gap progressively decreases as the pellets further densify in the 1700°C zones.

In the case of vertically charged pellets the gap increases as the theoretical density approaches 95 % of theoretical density by a value equivalent to 0.4 d. This helps in improving the heat transfer by convection enormously.

For vertically charged pellets the increase in gap helps in the free flow of cracked ammonia.

Heat transfer to the pellets from the molybdenum-heating element is partially due to conduction and convection and predominantly due to radiation.

The transfer of heat by conduction mode for the three different types of charging patterns is approximately the same.

But heat transfer by the remaining two modes i.e. convection and radiation will be different for three different types of charging patterns.

Heat transfer by convection is obtained from the calculation of heat transfer coefficient or rate of heat transfer obtained from dimensional analysis as given in equation - 2 [4].

$Nu = a (Re)^b (Pr)^c$ ---- 2
Nu = Nusselts Number.
Re = Reynolds Number.
Pr = Prandtl Number.

Where a, b and c must be evaluated from a minimum of three sets of experimental data.

The conductivity of all the common gases and vapors increases with increasing temperature. Sutherland deduced an equation -3 from the kinetic theory, which is applicable to the variation of conductivity k.

$K = K32 * [(492 + Ck)/(T + Ck)] * (T/492)$ ---- 3

Ck = Sutherland constant.

T = absolute temperature.

K32 = conductivity of the gas at 32 deg F.

From the value of conductivity obtained from equation -3 and the flow rate of cracked gas, dimensions of the hearth size viscosity, density etc the heat transfer coefficient can be arrived.

The heat transfer by convection dQc can be calculated from equation - 4

$dQc = h * dA * dt$ ---- 4

The heat imparted to the pellet depends on the geometric surface area dA of the pellet exposed to the flow of hot cracked gas. The increase in area of the gap between the pellets exposed at higher temperatures is higher in vertical charged pellets compared to horizontally charged pellets in charge carriers.

Transfer of heat by radiation is predominant in the case of high temperature furnaces. The total incident energy on the pellets is unity and is given by the equation -5.

$\alpha + \Upsilon + \tau = 1$ ---- 5

Where the absorptivity α is the fraction absorbed and the reflectivity Υ the fraction reflected and transmissivity τ the fraction transmitted.

The substances having nearly complete or unit absorptivity are lampblack, platinum black and bismuth black absorbing 0.98 to 0.99 of all incident radiation. In the list of substances given above we can include the sintered uranium dioxide pellet.

By considering the above, the radiant energy transfer from the molybdenum heating element maintained at an absolute temperature T1 to the pellets maintained at T2 is given by equation --6.

$$Q = \sigma * A * [(T1/100)^4 - (T2/100)^4] \quad \text{---- 6}$$

σ = Stefan-Boltzman constant.

A = exposed area of the pellet to radiant energy.

The exposed area forms an important component for absorptivity / transmission of radiant energy. The exposed area is higher in vertically charged pellets thus making the transfer of heat by radiation more effective.

4.2 Defect Distribution

The visual defects calculated for these lots are given as per the Table 2. The chipping defect is higher in the case of random charging compared to the horizontal charging .

The chipping defect in the random charging is mostly attributed to the transmitting of shock load to the edge of the pellet where as the shock transfer for vertical charging is to the total circular area of pellet in the area of the flat end.

5.0 SUMMARY

1. Gas flow path plays an important role for achieving sintered density.
2. For homogenous powders with low activity horizontal charging can be used for LWR pellets.
3. Moisture content has to be controlled in the powder stage for horizontal charging.
4. Inventory of molybdenum increases with vertical charging of pellets.
5. Consumption of molybdenum items like shrouds and sheets also increases in vertical charging of pellets
6. Defects due to chipping is higher in random charging compared to vertical charging.

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TABLE - 1

Type of Charging	Density Range	Mean x	Standard Deviation	x-3sigma
1.Random	10.64 to 10.71	10.678	0.0208	10.615
2. Vertical	10.66 to 10.74	10.706	0.0219	10.64
3.Horizontal	10.17 to 10.46	10.346	0.068	10.139

TABLE - 2

Type of Charging	Accepted %	Chips	Endcaps	Pits	Cracks
1.Random	63	21	7	-	9
2. Vertical	89	9	1	1	-

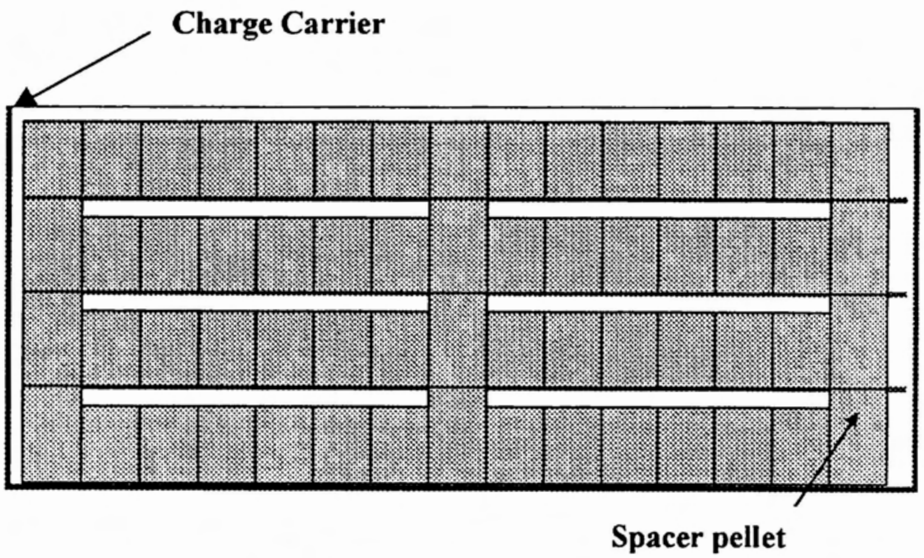

FIG.1 :VIEW OF THE GREEN COMPACTS CHARGED VERTICALLY BEFORE SINTERING

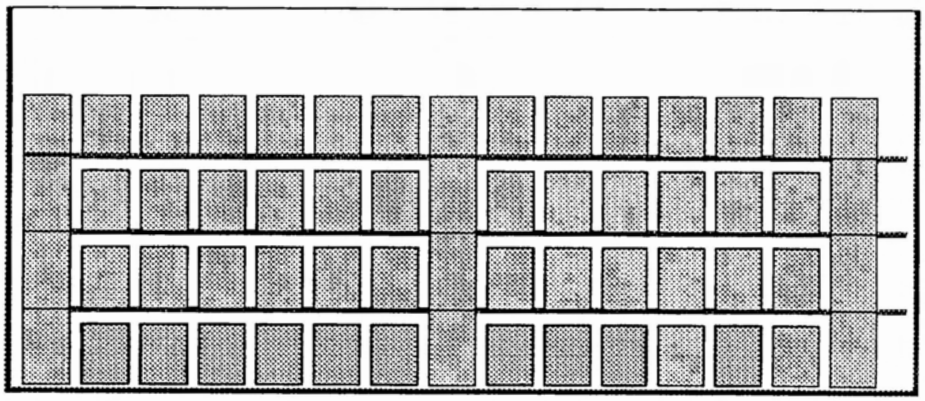

FIG.2: VIEW OF THE VERTICALLY CHARGED PELLETS AFTER SINTERING

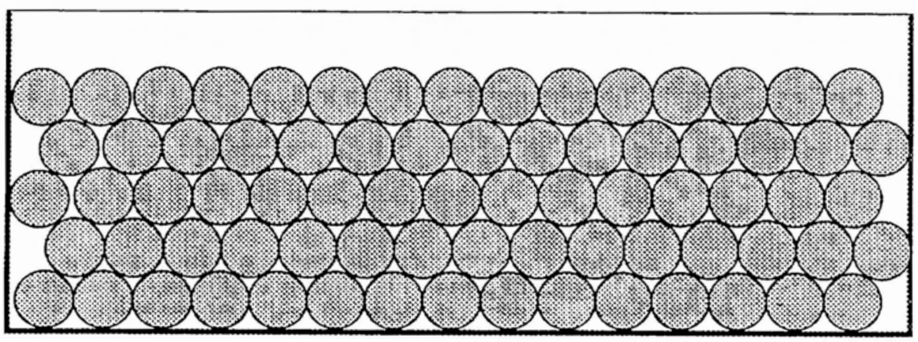

FIG.3 : VIEW OF THE HORIZONTALLY CHARGED GREEN COMPACTS BEFORE SINTERING.

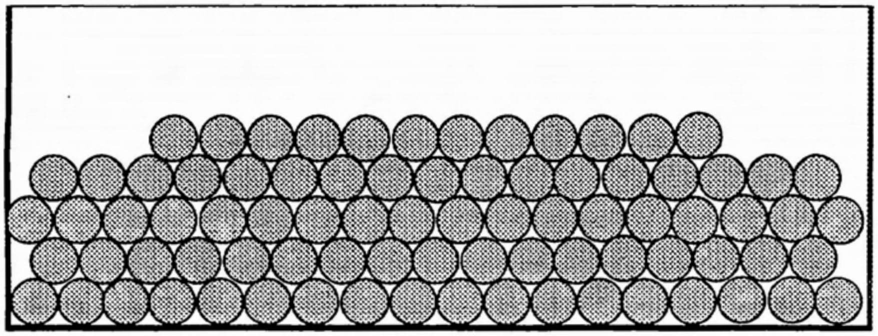

FIG.4: VIEW OF THE COMPACTS CHARGED HORIZONTALLY AFTER SINTERING

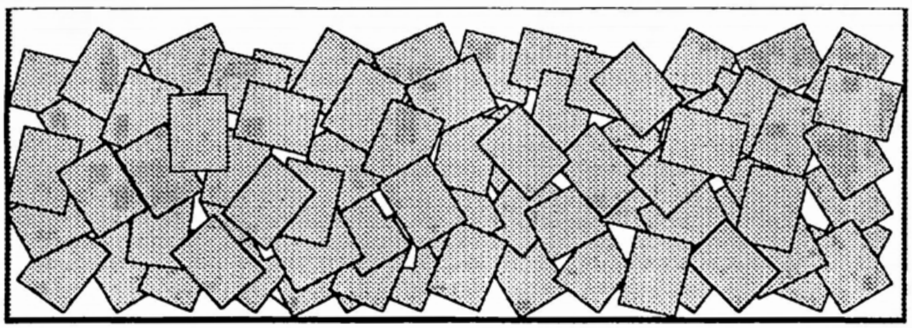

FIG.5 : VIEW OF THE RANDOMLY CHARGED GREEN COMPACTS.

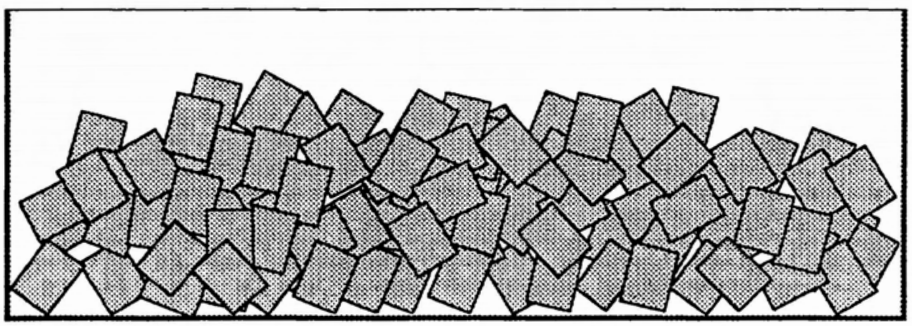

FIG.6: VIEW OF PELLETS RANDOMLY CHARGED AFTER SINTERING.