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REVIEW ON QUALITY CONTROL TECHNIQUES OF UO_2 PELLETS UNDER PILOT-PLANT CONDITIONS, AT INSTITUTO DE ENERGIA ATOMICA, SÃO PAULO, BRASIL

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ABSTRACT

The main objective of the Instituto de Energia Atomica's Metallurgy Division Pilot Plant is to adapt known procedures for fabrication and testing UO_2 pellets starting from uranium salts, and, when necessary, to develop new methods that would better suit local conditions, in order to build up progressively a national know how in this field. Its present capacity is 5 tons per year and its flexible layout and equipment affords easy changes in operating steps when necessary.

The paper describes the quality control techniques that have been adopted, both in the powders and in pilot plant pellets fabrication lines. Much of the work done dealt on natural UO_2, but limited production was also achieved in low-enrichment pellets.

Results which were recently obtained in short test rods irradiation in foreign reactor centers have shown that fabrication procedures and quality evaluation are satisfactory, demonstrating that an appreciable know how has been gained from development work done entirely within the Pilot Plant and other Division's laboratories.

1. INTRODUCTION

The Instituto de Energia Atomica's Metallurgy Division through its Pilot Plant aims to fabricate experimental quantities of UO_2 pellets for fuel elements, to develop related fabrication and control techniques, to train personnel, and to acquire experience through implanting quality control programs for fuel elements. Its design, construction and operation resulted wholly from the experience gained in research and development within other Division's facilities, since its start back in 1962.

Efforts have always been directed towards keeping pace with recent foreign specifications and developments, and therefore to fabricate UO_2 pellets which consistently would meet such standards, in view to both performance and safety during their utilization in power reactors.

This trend reflects the adapting of known procedures to suit the particular set of conditions of scale and availability of materials, in order to build up, progressively, a national know how in this field. Most of the work done dealt with natural UO_2 powders, but several experimental lots of low-enrichment UO_2 pellets have also been varried out.

The work done has always been closely associated with development work in the Chemical Engineering Division's Pilot Plant, which, among other activities, deals with uranium and thorium salt purification. Such association provides a straight knowledge on the influence of chemical processing variables on resulting powder properties and their behavior in sintering and on the properties of the resulting UO_2 pellets.

The Pilot Plant layout design and equipment selection were set to afford ample flexibility. It comprises several segregated units for each particular operation, to assure the changes that may be required in operating procedures to suit the specified UO_2 pellet properties, starting from a wide variety of uranium salts and oxides, including imported ones.

Usual fabricating procedures comprise the following main steps: calcining (ammonium diuranate or ammonium-uranyl tricarbonate) to U_3O_8; reduction under hydrogen to non-stoichiometric UO_2; blending the product of individual batches; conditioning the powders; pressing in automatic presses, with pre-pressing and granulating steps when necessary; sintering in a continuous high-temperature sintering furnace under hydrogen; centerless grinding of the pellets, and storage until shipment. Fig. 1 reproduces the plant flowsheet with indication of tests and controls exerted.

Present installed capacity is 5 tons per year of UO_2 pellets. Production can be increased through installation of a second sintering furnace.

2. QUALITY CONTROL IN THE UO_2 POWDERS LINE

The Pilot Plant has been designed and built |1| with due consideration to the experimental character of the unit, that is, aiming to establish data and parameters of each fabrication step with precision and reproducibility (Fig. 2).

Accordingly, each main operation is carried out in closed compartments, segregated self-contained plant areas, conveniently disposed within the Pilot Plant building (Fig. 3). Each of them has its own operating staff, simplifying quality and accountability controls; adequate working conditions are assured, and in case of eventual contamination, such segregated area can effectively be dealt with. In each of those areas one technician that belongs to the quality control group is also a member of the operating group. His responsabilities include to follow strictly the operations and to take all the required samples for the operation control.

Controls exerted on powders comprise routine and special tests.

The routine tests performed depend upon the operation. For the uranium salts reception the following tests are carried out: humidity; bulk apparent density; BET surface area; chemical analysis of impurities; and net U_3O_8 content. Sometimes a special test is performed through scanning electron microscopy, to study morphology, agglomeration and open porosity, as well as the crystallites shape.

The next fabrication step is calcining to U_3O_8, done in a large batch muffle furnace, which assures uniform temperature on the several rows of shallow alloy steel trays which are loaded with uranium salt, previously classified at minus 9 mesh. This fabrication step was described in previous papers |2,3,4|.

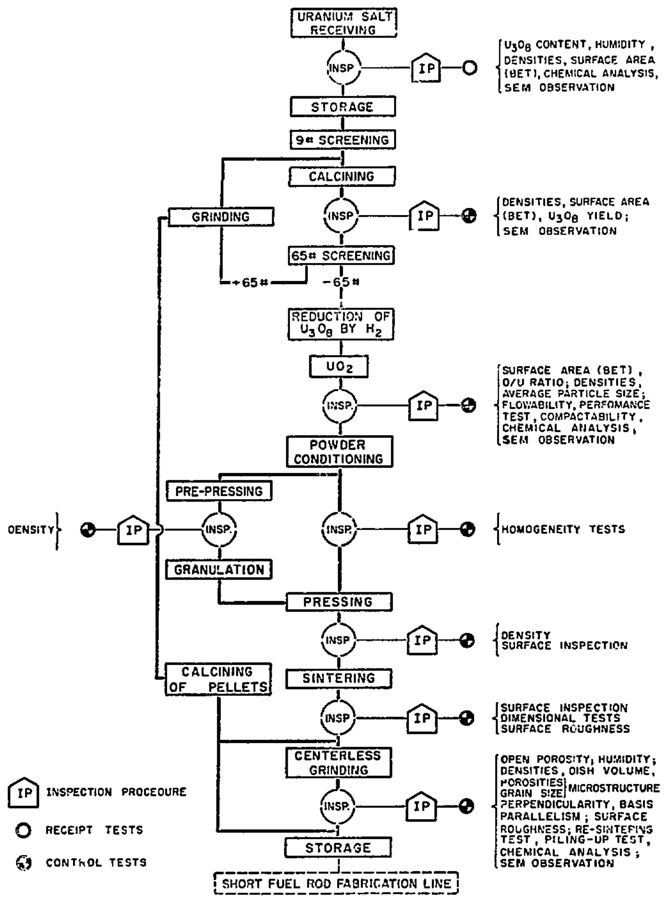
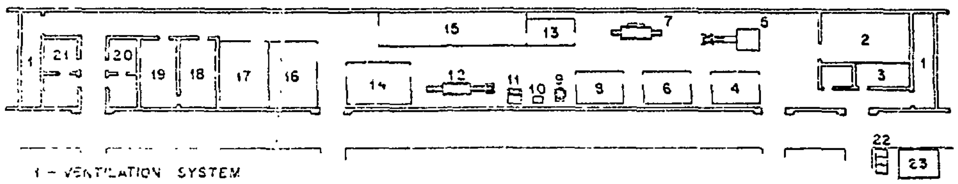


Fig. 1 - Simplified flowsheet of pilot plant operations, with indications of tests controls exerted.

Quality control of U_3O_8 powder comprises the following routine tests: apparent bulk density; BET surface area, and yield of U_3O_8 thus produced. The accurate weighing of charges and products in each individual tray assures good accountability control in this step. This particulary important in dealing with enriched U_3O_8 powders.

Before reduction, U_3O_8 is sieved through 65 mesh screen; the +65 fraction is further ground and recirculated into the calcining step, and the -65 product after blending and homogenizing is subjected to hydrogen reduction to non-stoichiometric UO_2. The electric continuous reduction furnace has been designed and locally built with experience gained in two other similar, but smaller, units that are in operation since 1964; it has three independently controlled heated zones and the trays are pushed mechanically



1 – VENTILATION SYSTEM
2 – URANIUM SALT STORAGE
3 – ENRICHED UO_2 STORAGE
4 – CALCINING CHARGE PREPARATION
5 – CALCINING FURNACE
6 – SCREENING AND REDUCTION CHARGE PREPARATION
7 – REDUCTION FURNACE
8 – BLENDING OPERATIONS AND MUFFLES
9 – PRE-PRESSING UNIT
10 – GRANULATOR
11 – PELLET PRESSING UNIT
12 – SINTERING FURNACE
13 – DIMENSION CONTROL ROOM
14 – CENTERLESS GRINDING
15 – ROD LOADING UNITS
16 – END PLUGS WELDING
17 – PELLETS STORAGE
18 – PERFOMANCE TEST LABORATORY
19 – SEM AND MICRO PROBE
20 – MEN'S LOCKER ROOM
21 – WOMEN'S LOCKER ROOM
22 – AUXILIARY POWER SUPPLY
23 – BOTTLED GASES STORAGE

Fig. 2 - Pilot plant layout (area 1300 sq. m)

through the furnace to the water-cooled final section before opening to atmosphere.

Extensive routine testing |5,6,7,8| is performed on UO_2 powders produced, as the properties of the final pellets do depend upon powder characteristics. The tests are: U-235 content; BET surface area; O/U ratio; apparent densities (bulk, tap and limit); average particle size analysis; flowability; ability to be pressed; granules and crystallites apparent density, chemical analysis of impurities, and behavior in sintering (performance test).

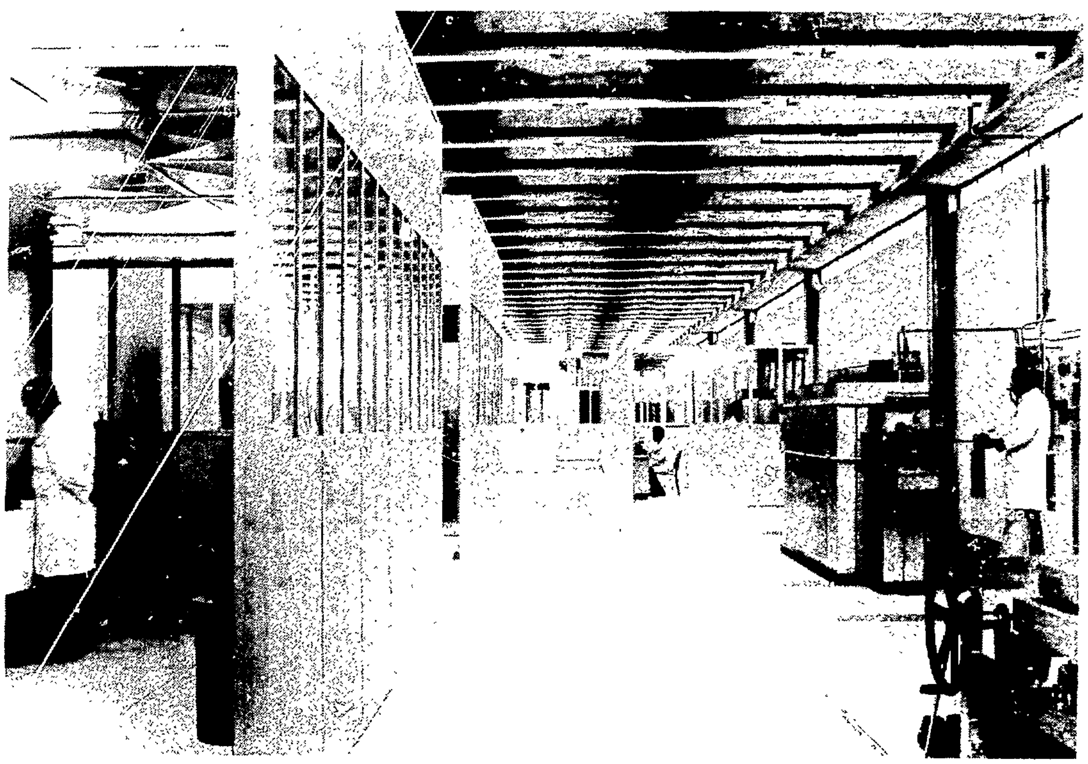


Fig. 3 - Internal view of the pilot plant showing some of the installed equipment and the compartments, where specific segregated operations are carried out.

In addition to surch routine tests, some special ones are also carried out, specially when changes of physical parameters are found to exist. Those tests are: electron scanning microscopy |9,10|, aiming to ascertain the pore shape and size; and sedimentation test (done in a large sedimentation balance) to measure the relative fractions of minus 35 mesh powder, which can be correlated with particular microestructures of sintered pellets.

At this point, the UO_2 can receive several different additions, aiming both to improve the green pellets properties and to meet the required sintered pellet densities. Some special tests were developed to ascertain the batch homogeneity. A special dilatometric test |11| is also used to obtain the kinetic parameters of UO_2 powders sintering, and therefore to determine the particular sintering cycle that will be used in actual furnace operation.

The development and implementation of such uranium oxide powders control aim through well established procedures to define how adequate can a powder be, from a given batch, to assure a final pellet which complies with the specifications, or otherwise, to indicate the changes that would be required to achieve such results. Much experience has been gained in actual Pilot Plant operation and some definite correlations of test results have been established.

Duplicate test samples are kept in order to be eventually further used in other tests, should any disagreement arise in the course of work.

3. QUALITY CONTROL IN UO_2 PELLET LINE

Depending upon the powders test results, operating procedures are established to guide actual pellet fabrication. Occasionaly new steps, including the so-called "conditioning" and pre-pressing, are introduced, to improve powder compactability.

Pressing is done is floating-type dies in hydraulic or mechanical presses. The controls exerted are green pellet density and surface inspection. Pellets that are coming from the production line are sampled at random time periods and tested for green density, but all for the pellets are examined for eventual surface defects |12,13,14|.

Pellets are placed in molybdenum sheet boats and further stored in drying ovens until loaded into the continuous sintering furnace, under hydrogen. The complete thermal cycle is previously established, based on the powder test results and on correlations with final sintered properties.

Sintered pellets are then submitted to extensive series of tests, aiming not only to ascertain their quality level but also to supply additional parameters in feed-back, which could be useful in further developments |15,16,17,18|.

Current practice at the Pilot Plant is to direct all the UO_2 sintered pellets to stepwise centerless grinding; therefore 100 pct of the pellets go through such final diameter control. This procedure has been adopted as result of the very strict specification diameter tolerances, that requires reproducibility within $\pm$ 0,005 mm. After grinding the pellets are submitted to visual inspection, under proper lighting to check for chips, cracks and surface pits |19,20|. As this is a subjective control, acceptance standards were established, in a pictorial way.

The tests performed in sintered and surface ground pellets |20,21| are: geometric and hydrostatic densities; measurement of dish volume; open and closed porosities; deviation of parallelism of pellet basis, and of perpendicularity of surface and basis; microestructure examination for grain- and pore size distribution, both in longitudinal and transverse sections; surface roughness; chemical analysis of relevant impurities, and humidity content.

A re-sintering test is another routine control regularly used and to which great practical importance is attached |22|. Samples of finished pellets are submitted to a re-sintering operation, at 1700 or 1750 ºC during 24 hours at temperature, and their dimensional and density changes are determined and evaluated under structural changes found, both in grain-and pore size distribution. Such changes are further to be correlated with actual behavior in irradiation tests.

Other special tests are sometimes carried out, to enlarge the knowledge thus gained in the many steps involved in actual pellet fabrication. This provides a feed-back of information, easing or modifying the current operating procedures. Among others, microestructure examination under scanning electron microscope, and fractographic studies on compression test fracture surfaces, supply important addicional information on volumetric homogeneity of grain-size and pore-size shape distribution |23|. Duplicate pellet samples are kept for repeating similar tests, should any doubt arise.

For each of the tests mentioned in this paper, the corresponding testing methods were established, as a result of the experience gained within the Division's laboratories or of adapting methods available in the international specialized litterature.

Aiming future action enlargement of Pilot Plant in fabrication of short irradiation test rods, it is presently under development a pellet piling-up test. Its purpose is to determine the change in height of the pellets piled-up one over another and the algebraic sum of their individual heights. Such parameter is important, as it defines the linear UO_2 mass concentration within the fuel element can.

4. TESTS UNDER IRRADIATION

As a consequence of the establishment of the described fabrication control program, need was felt for further evaluation of pellets behavior under irradiation, to ascertain their intrinsic quality levels for reactor operation.

Accordingly, efforths have been pooled together with other Brazilian institutions and agencies through testing contracts to be carried out in foreign nuclear centers. Such testing programs were envisaged to gather data on actual pellet behavior under irradiation at neutron fluxes and linear power rates which would correspond to the expected ones is power reactor operation.

The good results which were recently obtained in such tests have shown that the adopted fabrication procedures and quality evaluation are fully satisfactory, demonstrating that an appreciable know how has been built up from actual development work which has been carried out entirely within the Pilot Plant and other Division's laboratories.

5. CONCLUSIONS

1. In order to build up progressively experience in fuel elements fabrication and to suit local conditions, fabrication procedures and testing methods were adapted or developed; they resulted from previous work done at the Instituto de Energia Atomica's Metallurgy Division facilities and many of them have been reported in litterature |1 to 23|.

2. The Pilot Plant and adjoining laboratories, locally designed and equipped, are sufficiently flexible to allow changes in operating procedures both in the UO_2 powders production and in the sintered UO_2 pellets fabrication.

3. The adopted quality control techniques were described. So far, such procedures and methods have been adequate for the fabrication programs tackled, which have been evaluated through some limited short test rod irradiation testing in foreign reactor centers.

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