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STUDY OF PRODUCTION OF FUEL PELLETS FOR A REACTOR

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ABSTRACT

Nowadays the electrical energy was been used much on society. A method for getting electricity is through nuclear power plants, this power plant uses fission that occurs inside the UO2 pellets to generate thermal energy that will be transform into electric. The pellets production was made from enriched UF6 uses some techniques of reprocessing UF6 gas to UO2 powder. This reprocessing process done by wet route (Ammonium Diuranate ADU or Ammonium Uranium Carbonate AUC) or by dry route (Fluidized bed or GECO). With getting of UO2 powder is forwarded to metallurgy where this powder is compacted in cylindrical matrix so that powder take the desired shape, this green pellets are full of the empty spaces (porosity) for this it is sent to the sintering. The sintering consists of a joint of these particles of powders by means of the heating of this green pellets, coming arrive the melting temperature, the UO2 molecules melting each other so decrease the porosity and increase the density. For the production of fuel pellets the process all most used is wed route by means the AUC ,this process arrive created for replace the ADU because the AUC is a process where less rework for the pore geometry is required compared to DUA. The fluidized bed process is more used in small samples however, for a large amount it becomes unfeasible, moreover the dry route process require more robust materials because of the generation of HF that is highly corrosive and cannot used the UNH (uranyl nitrate hexahydrate) used for recycle materials discarded in manufacturing.

1. INTRODUCTION

Currently in Brazil there is a great consumption of electric energy. The electricity consumption of all regions in December 2014 reached a total of 473,396 GWh.

The electric energy in Brazil is separated into several energy matrixes, there are several models of Power plants for the production of electric energy in large scale. Represented by 1.2% of the entire Brazilian energy matrix is the Nuclear Plants. In Brazil are two nuclear power reactors that are installed in the state of Rio de Janeiro.

Angra reactors are of the Pressurized Water Reactor type, they are the largest type of reactors manufactured in the world. This type of reactor has two kinds of types of circuits for its operation. The primary circuit is where the reactor core is located which works by fissioning the uranium atoms inside the rods of the fuel element, thereby generating heat and heating the water that functions as a coolant that passes through the core at a temperature of 320 degrees Celsius . The water inside this circuit has to be pressurized there are 157 atmospheres. With the heated water being sent to a heat exchanger, this heat exchanger or steam generator is formed by a series of U-pipes where the water of the primary passes, which transmits this temperature by means of thermal induction to the water of the secondary which is on the outside of the tubes. The pressure inside the secondary is much lower than that of the primary then the vaporization of the water that is routed to the turbine is coupled to an electric generator. This steam, after moving the turbine, passes through a condenser, where it is cooled.

2. OBJECTIVE

Study on the methods for the production of fuel pellets for a PWR reactor, starting from the conversion of UF6 to the production of pellets.

3. PRODUCTION OF FUEL PELLETS

The figure 1 describes the fuel cycle from the prospecting to the UO2 fabrication and its treatment after use in the reactor.

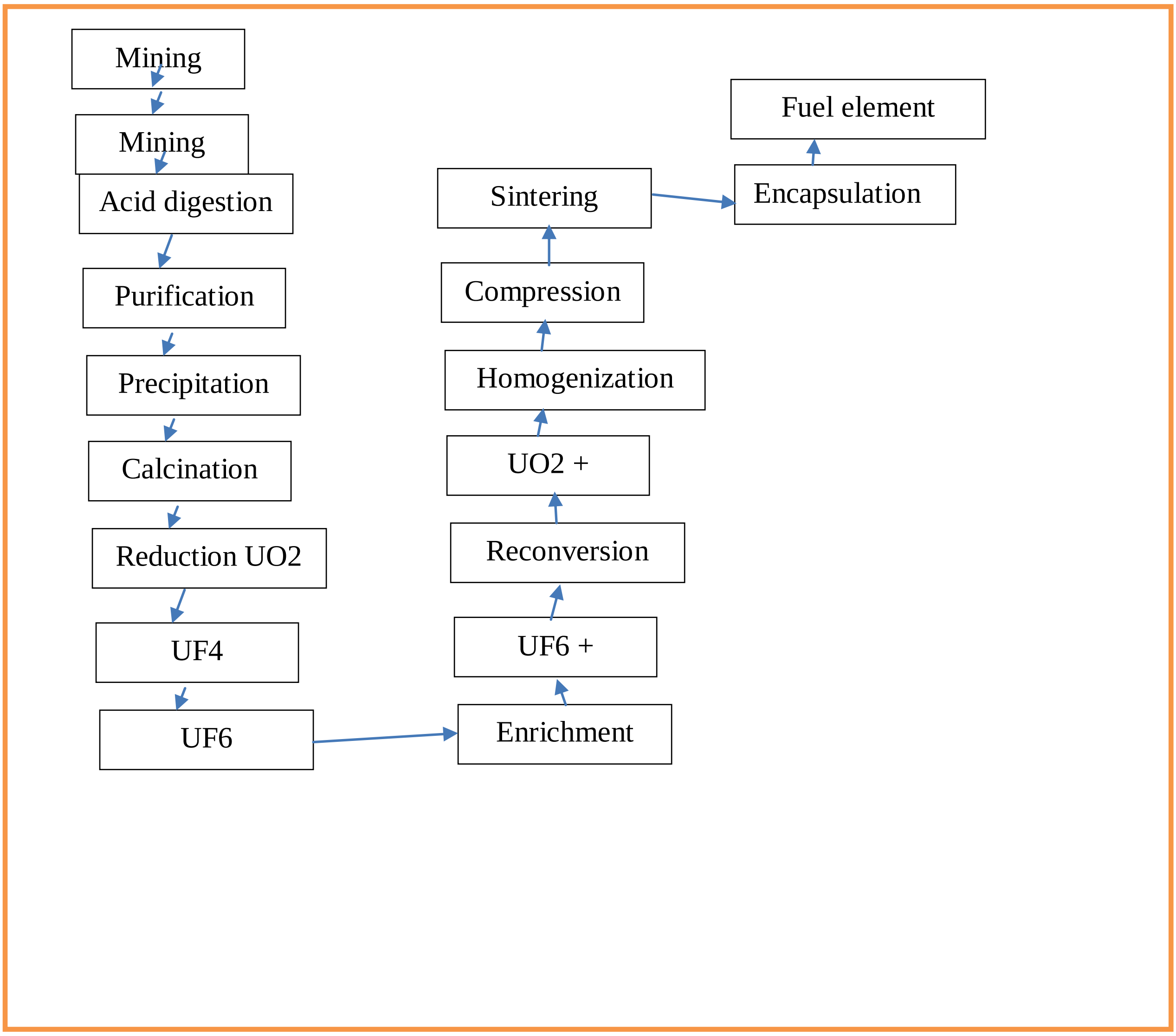

Figure 1: Uranium cycle flow chart (SANTOS, 1992)

The proposed work consists of the production of fuel pellets from UF6, so this material is already enriched, so the study will be developed starting from the Reconversion.

3.1. Production of UO2 powder

The main processes of obtaining UO2 on an industrial scale can be divided into two types.

Wet processes in which the precipitation of a uranium compound from an aqueous solution is used as the intermediate procedure, the intermediate products subsequently being calcined and/or reduced to obtain UO2.

Dry processes, in which UF6 is converted to UO2 through gas phase reactions, with a series of variants as a function of intermediate products.

3.1.1. Reconversion by wet route – Ammonium Diuranate (ADU)

The reduction of the ADU to obtain UO2 was the first to be developed on an industrial scale. The ADU can be obtained from UF6 or a solution of uranyl nitrite UO2(NO3)2. When UF6 is used, it is hydrolyzed by reacting with ammonia, obtaining the DUA, (SILVA, 2011) with the following reactions:

$$2UF_{6_{(G)}} + 7H_2O_{(l)} + 14NH_{3_{(S)}} \rightarrow \underset{(ADU)}{(NH_4)_2U_2O_{7_{(S)}}} + 12NH_4F_{(l)} \qquad (1)$$

The cake is dried and steam and hydrogen treated in a furnace to reduce the ADU to UO2 and thereby decrease the fluoride content to desirable levels when the ADU is obtained from UF6. After the reduction occurs the passivation step, which consists of the controlled oxidation of the UO2 powder, the resulting product is a non-stoichiometric oxide.

Generally, a mechanical conditioning is required to adjust the particle size and the flow ability of the UO2 powders obtained through the ADU method. Typical treatments are grinding and / or pre-compaction, followed by fragmentation and granulation. If organic binders are used in pre-compaction, binder homogenization and possibly a specific removal treatment of these organic materials before sintering of the pellets would still be required. (LAINETTI, 1992)

3.1.2. Reconversion by wet route – Ammonium Uranium Carbonate (AUC)

The container of the precipitation unit is filled with demineralized water. The gases UF6, CO2 and NH3 are fed and dosed through a system of injector nozzles. The operation is performed such that an excess of ammonium hydrogencarbonate NH4HCO3 is always present in solution, so that the AUC is formed immediately from the hydrolyzed UF6. The reaction takes place according to the equation:

$$UF_{6_{(G)}} + 5H_2O_{(l)} + 10NH_{3_{(G)}} + 3CO_{2_{(G)}} \rightarrow \underset{AUC}{(NH_4)_4(UO_2(CO_3)_3)_{(S)}} + 6NH_4F_{(l)} \qquad (2)$$

Drying, decomposition, reduction of the fluorine content (by pyrohydrolysis at about 650°C) and reduction to UO2 (with H2 or cracked ammonia) is carried out in tray ovens or in fluidized bed furnaces. Thereafter, a slight oxidation of the obtained stoichiometric UO2 powder is carried out, an operation known as stabilization or passivation. Otherwise the powder could not be handled in the air, oxidizing to U3O8, due to pyrophoricity (LAINETTI, 1992). The overall reaction that occurs in the reduction:

$$(NH_4)_4(UO_2(CO_3)_3)_{(S)} + H_{2(G)} \rightarrow UO_{2(S)} + 4NH_{3(G)} + 3CO_{2(G)} + 3H_2O_{(G)} \quad (3)$$

3.1.3. Reconversion by dry route – Fluidized bed

UF6 is hydrolyzed with superheated water vapor to obtain solid UO2F2. UF6 is introduced in the gaseous form, either pure or with the aid of N 2, through an orifice located just above the porous plate of the fluidization furnace. The UO2F2 is formed on the surface of the powder particles previously placed in the kiln, "seeds" are placed before the injection of UF6 begins. These seeds may be composed of partially reduced UO2 or UO2F2. The particles baked from the seeds have a relatively large diameter, contributing to the success of the bed behavior when being fluidized, and to reducing the problems inherent in the existence of fines. However, the large particles have an effect of damaging the following reactions of reduction to UO2, since the speed of most gas-solid reactions is slower for larger particles. In the second stage of the Dry Bed process, the UO2F2 is reduced with hydrogen in the presence of water vapor, to increase the reaction rate and avoid the formation of UF4 (LAINETTI, 1992).

$$UF_{6(G)} + 2H_2O_{(l)} + H_{2(G)} \rightarrow UO_{2(S)} + 6HF_{(G)} \quad (4)$$

3.1.4. Reconversion by dry route – GECO

The process is carried out in an inclined rotary kiln, in which the UF6 is injected into the upper end of the kiln together with superheated steam, and comes in contact with H2, injected countercurrent. In the region of injection of UF6, the reaction occurs with H2O (steam), producing UO2F2, which is reduced to UO2 with H2. The Formula of the balance is the same as that of the fluidized bed (4).

3.2. Powder metallugy

Sintering can be defined as a process in which materials in powder form are transformed into a mass under the influence of heat. The process consists of a transport of matter by thermal activation, which leads to the strengthening of the contacts between the particles, altering the geometry of the pores and grains and the reduction of physicochemical gradients. This mass transport aims to reduce the free energy of the system until it reaches equilibrium..

After obtaining the powder of UO2 be by any means of those described in item 3.1. Subsequent steps to obtain the pellets consist in pressing the powder into pellets and sintering the pellets until reaching the required density.

The pressing consists in charging an amount of UO2 powder about 7 grams into a cylindrical die, pressing this powder at a pressure of 400MPa. This step of the fuel fabrication process has dual purposes: to give the pellet the desired shape, and to promote the intimate contact between its particles.

The process vision from the distribution of the particles that constitute the powder inside the green tablet. It is known that powders are composed of multiple generations of crystallite clusters. When these powders are pressed, depending on the binding fragility between these multiple generations of clusters, these crystallites make contact with the neighboring crystallites, and can reach a maximum coordination number of 7 on a green tablet (ASSIS, 2007). The next step is to sinter the pellet to promote the densification and evolution of its microstructure. Both the densification and the microstructural evolution of the pellet during sintering have a very close relationship to the structure established by pressing the pellet to green.

The sintering process was divided into three steps.

3.2.1. Initial stage

At this stage the individual particles of the compact remain perfectly identifiable joined by the growth of the necks, i.e. formation of grain outlines. What maintains the grain boundaries between two adjacent particles are the elastic forces resulting from the surface tension. During the initial stage, the area of contact between the particles increases from zero to approximately 20% of the cross-sectional area of the particle. The centers of the particles approximate slightly, corresponding to a small contraction of up to 5%, causing a decrease in surface activity to occur. As shown in figure 2 and figure 3.

3.2.2. Intermediate stage

At this stage only 5% to 10% of the porosity remains. The "neck" is already sufficiently grown and the pores have the approximately cylindrical shape constituting an extensive network of interconnected channels. This network of pores is considered as a series of fictitious cylinders arranged along three edges. During this stage the test body has a volumetric retraction of up to 30%. As shown in figure 2 and figure 3.

3.2.3. Final stage

At this stage, the surface free energy decreases with the pores taking the spherical shape and located at the vertices or inside the grains. Small pores tend to disappear by migrating gaps (crystalline imperfection involving absence of atoms) into the larger pores (structural defect, absence of material) where the free surface energy is smaller. During this last stage, the densification is very slow and, often, the discontinuous growth of grains occurs. When this happens most of the pores are isolated inside the grains and densification virtually ceases. As shown in figure 2 and figure 3.

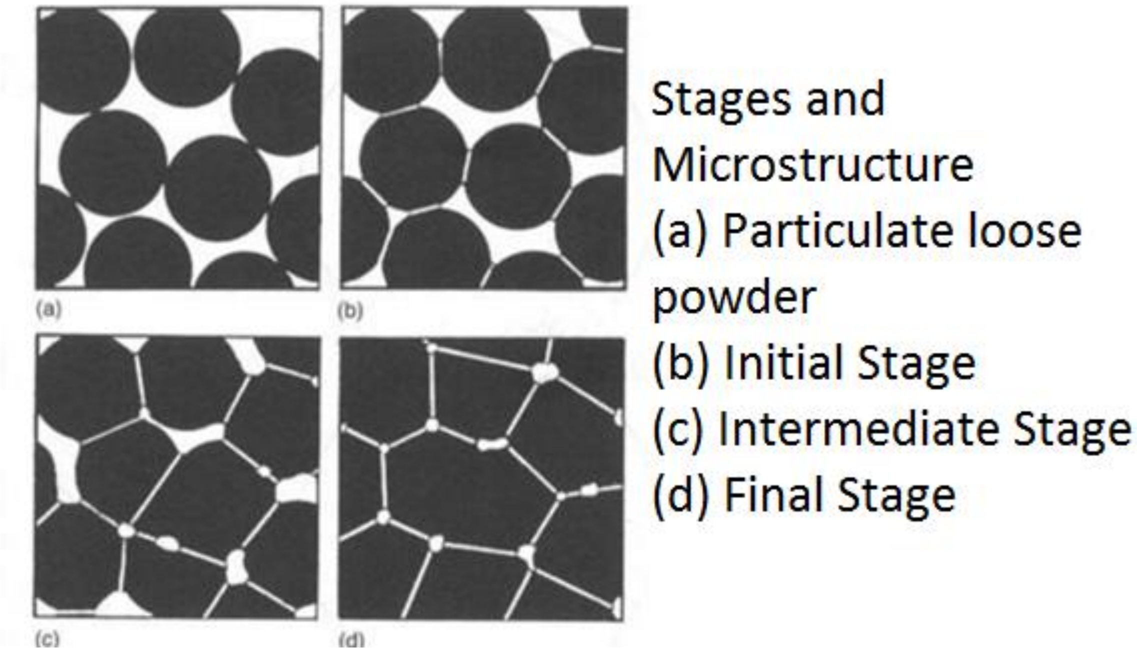

Figure 2: Sintering stages

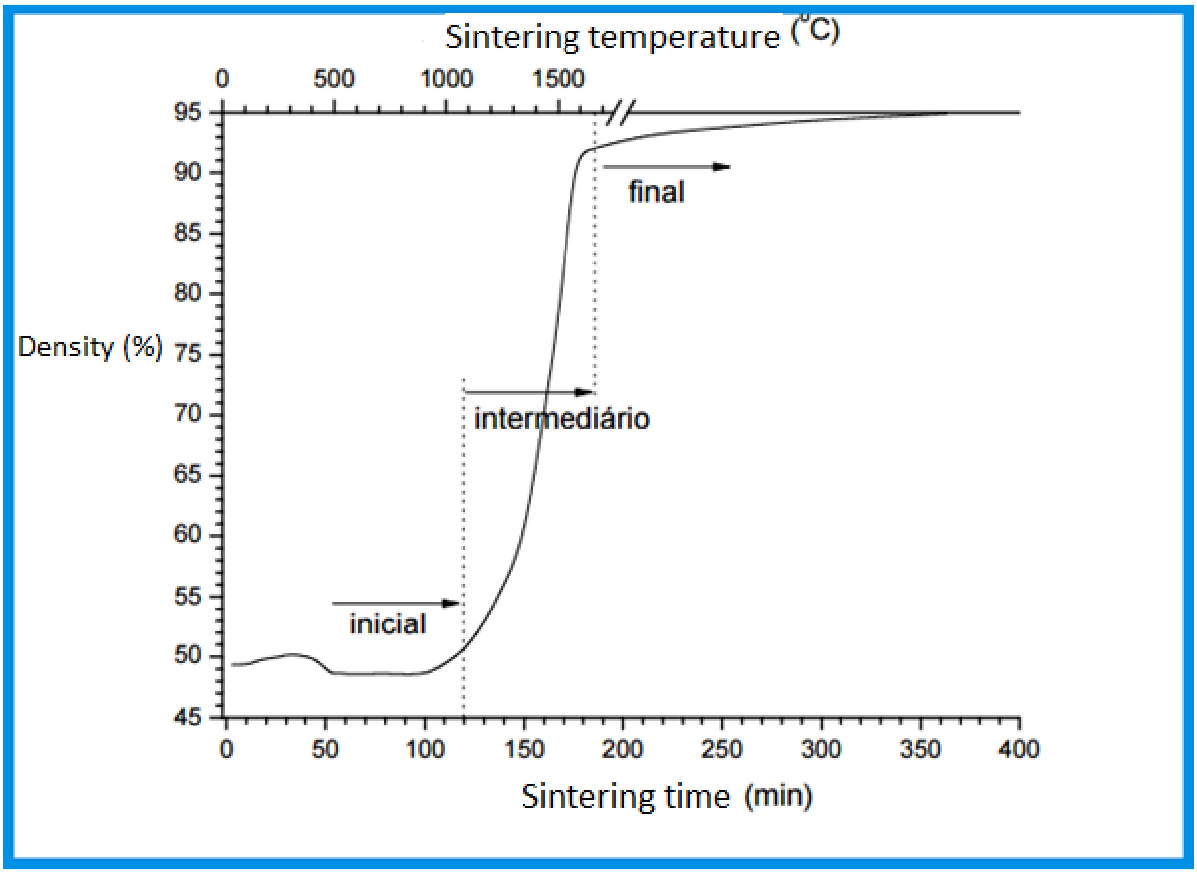

Figure 3: Diagram of temperature, time and density of pellets

With the sintering process there is mass transport (and consequent filling of the voids as the pores and displacement of interfaces as the grain contours) promoting the densification and growth of the grains. Thus, several of the mass transport mechanisms, called diffusion mechanisms, that detail the paths through which mass transport has been proposed, have been proposed (ASSIS, 2007).

- Surface diffusion (movement of atoms on or near surfaces of crystallites and grains). The flow of material occurs on the surfaces of the necks formed between the particles or in the grain contours in the third stage of sintering.

- Diffusion in volume (movement of atoms through the crystalline lattice). This mechanism is highly influenced by the composition of the material, curvature of the surface, and mainly by the sintering temperature. It is the material transport mechanism that requires the highest activation energy values.

- Diffusion in grain outline (movement of atoms through grain outlines). In this mechanism the material is removed from the grain contours, flows along these contours and deposits on the surface of the neck in the region of the boundary between the particles.

- Evaporation-condensation (movement of atoms by gas phase). Evaporation of the material occurs on the convex parts of the surface of the particles; then the vapor phase of the material moves within the pore, finally condensing into the concave portions of the surface of the particles.

3.3. Quality of fuel pellets

The processes have to be inspected to be classified as fit for a combustible element. This quality process is done between the intermediary processes so that in the end much of the pellets made can be routed to the fuel element. Represented in figure 4.

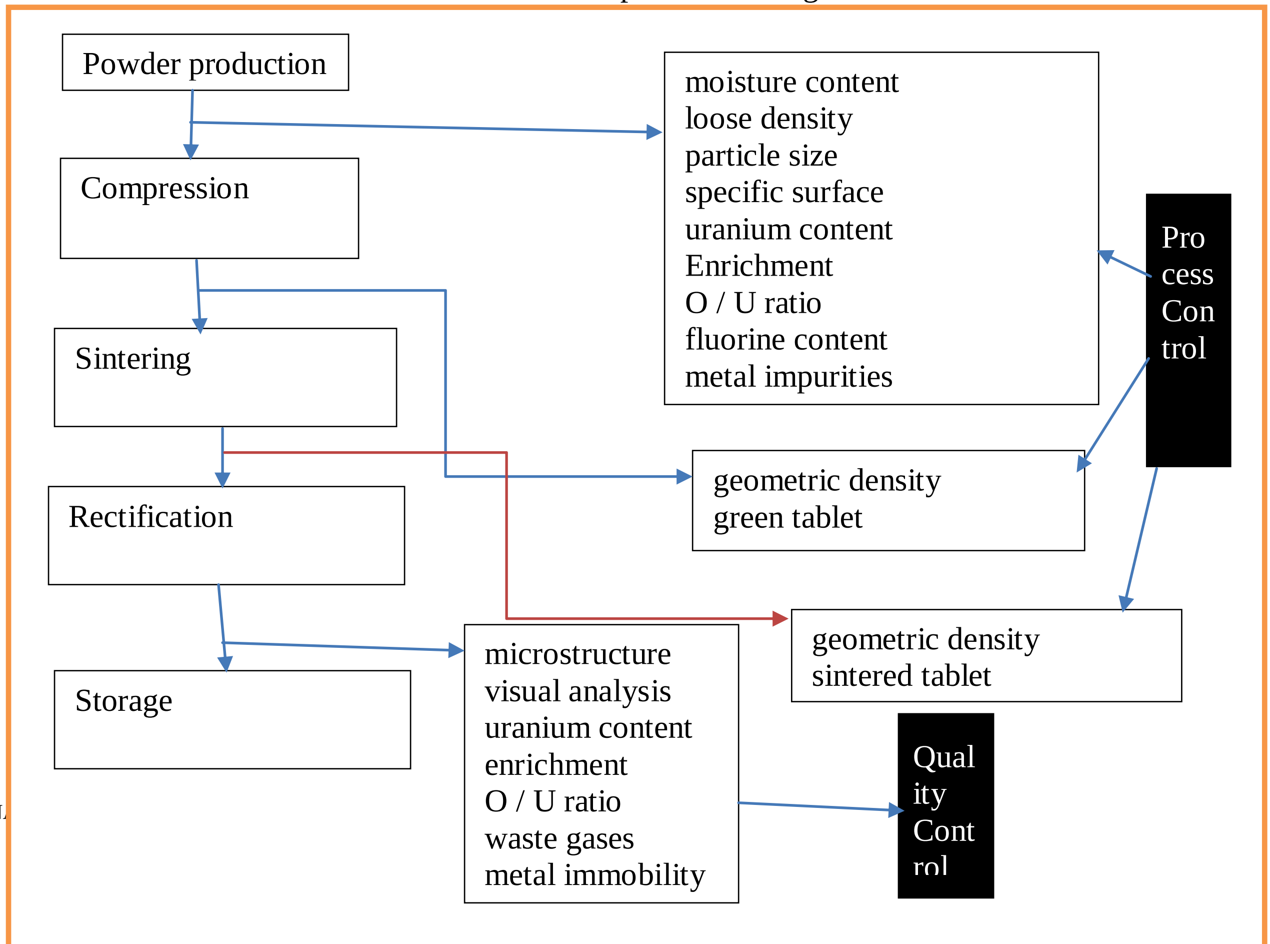

Figure 4: Quality control of tablet production

4. FUEL ELEMENT

A fuel element (FE) is defined as the reactor component, which properly contains the combustible material in a well-defined geometric and structural form. The reactor core consists of a set of fuel elements. Each type of reactor has an appropriate type of combustible material and an appropriate form of fuel element containing such material.

The fuel element of a commercial PWR reactor consists of fuel rods (14 x 14, 15 x 15, 16 x 16, 17 x 17, 18 x 18) with defined spacing. The main components of the fuel element are: fuel rod, control rod, spacer grids, end nipples, and FE securing spring (SILVA, 2011).

The shape of the insert is cylindrical, with cavities at the ends and chamfers at the edges. This shape is made in order to minimize the axial and radial thermal expansions of the insert ends so as to improve the performance of the rod during irradiation (avoid high axial deformation of the insert column and avoid hard contact insert coating at the interface between inserts) since the radial thermal gradient in the pellet is very pronounced. To keep the column of pellets bound within the fuel rod (especially during handling and transport of the FE) and to create void spaces to accommodate the fission gases produced during irradiation, a spring is placed on the top of the rod. This spring also accommodates the differential expansions between the insert column and the coating, avoiding high stresses. Between the spring and the column of combustible pellets an insulating pellet of Al2O3 (alumina), is placed so as to decrease the heat flow of the fuel pellet to the plenum region as well as to avoid reactions between the pellet and the spring, since temperatures in the central region of the pellet can be high (above 600 ° C). The material normally used for the coating tube are zirconium alloys (zircaloy-4, Zirlo ™, Duplex), which have good mechanical characteristics and resistance to corrosion, good performance under irradiation and low absorption shock section. The fuel rods are internally filled with helium gas in order to improve heat transfer from the pellets to the shell (SILVA, 2011).

Absorber rods that enter the FE in order to control the chain reaction in the reactor core occupy some positions of the FE.

A PWR core of 1100 MWe can contain 193 fuel assemblies composed of more than 50,000 fuel rods and about 18 million fuel pellets. Once loaded, the fuel remains in the core for several years depending on the design of the operating cycle. During refueling, every 12 to 18 months, part of the fuel - usually a third or a quarter of the core is removed for storage, while the remainder is rearranged to a location in the core most suited to its remaining level of enrichment.

5. CONCLUSION

It is concluded that a nuclear reactor needs several processes before starting its operation.

The reprocessing method of UO2 used for large-scale production is the Ammonium Uranium Carbonate (AUC), as it is a cheaper and simpler process to be applied. The other conversion processes, excluding ADU, are used in small scale (laboratory) samples.

In sintering the most sensitive range is the intermediate where the densification of the tablet takes place, a very high control is necessary because any unexpected variation will leave the tablet unusable.

REFERENCES

1. LAINETTI, P. E. O. "Desenvolvimento do processo de produção de pós de UO2, a partir de nitrato de uranilo, via atomização". Instituto de Pesquisas Energéticas e Nucleares, São Paulo, 1991. http://www.iaea.org/inis/collection/NCLCollectionStore/_Public/23/061/23061773.pdf
2. SANTOS, G. R. T. "Sinterização de pastilhas de dióxido de urânio em atmosfera de CO2". Instituto de Pesquisas Energéticas e Nucleares, São Paulo, 1992. http://pelicano.ipen.br/PosG30/TextoCompleto/Glaucia%20Regina%20Tanzillo%20Santos_M.pdf
3. SILVA, P. H. P. "Processo de beneficiamento do urânio visando a produção de energia elétrica". Universidade Estadual do Ceará, Fortaleza, 2011.
4. ASSIS, G. "Crescimento de grãos em pastilhas de UO2 para uso como combustível nuclear de elevado desempenho". Universidade Estadual de Campinas, Campinas, 2007.