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FUEL FOR THE NEXT BRAZILIAN NUCLEAR POWER PLANTS

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ABSTRACT

The conclusion of the Angra III nuclear power plant ends a cycle of the nuclear energy in Brazil that started about forty years ago. Nowadays the country is planning the installation of 4 GWe to 8 GWe of nuclear power up to the year 2030. The nuclear reactors considered for this new cycle should take into account the current technologic development and environment of the nuclear market. They certainly will have significant differences in relation to the Angra I, II, and III reactors. Important impacts may result on the nuclear fuel production chain, e. g., case high temperature reactors were chosen, which can deliver electricity and heat. The differences between the fuels of the candidate reactors after Angra III are analyzed and development lines are suggested to minimize these impacts.

1. INTRODUCTION

The next nuclear power plants after the conclusion of Angra III will probably have important design improvements due to safety, economic, and environmental reasons. Passive safety, standardized components and equipments, and longer plant operation life are extensively being taken into consideration in the nuclear market [1,2].

On the other hand, strategic issues demand additional roles for the nuclear energy [3-6]. The restrictions on the emissions of greenhouse gases and the deregulation of the electricity market are examples. Electricity generation in a regulated market has been the primary foundation for the design of nuclear power plants since the decade of 1950s, which favored large centralized plants of power greater than 1 GWe. But the deregulated electricity market may favor smaller than 1 GWe, distributed plants. 7 to 8% of the generated power is lost in electricity transmission lines. Which one is now the best approach? To construct a 1.2 GWe power plant and send the power to distant places through transmission lines or to construct two 0.6 GWe plants far apart each other and closer to the end users? Financing is also an important issue, because nuclear power plants require the greater part of the expenses during their construction. Again, which one is the best approach? To construct a 1.2 GWe power plant or to construct first a 0.6 GWe plant, that immobilizes less funds during some years, and then to construct a second 0.6 GWe plant?

The restrictions on the greenhouse gases emissions may demand heat as an additional product of the nuclear power plants [3-14]. If heat is considered, the impact on the design of nuclear power plant designs is great. The temperature of 600 to 1000 ^{o}C required for industrial heat (mainly for hydrogen production) exclude reactor cores with metallic components and water, which are commonplace in the existing nuclear power plants.

The optimal use of natural resources and minimization of waste are issues that also stress the shift to more advanced power plants. The current water-cooled reactors use only 1% of the natural uranium. This figure ought to be increased to at least 50% to ensure a sustainable use of nuclear energy [5].

This paper discusses the impact on the fuel fabrication of the introduction of new reactors designs in Brazil after the conclusion of Angra III.

2. THE CASE FOR EVOLUTIONARY AND PASSIVE CONCEPTS

These concepts are focused on the electricity market. The evolutionary concept is based on the PWR (like Angra I, II, and III) and BWR designs, which incorporate improvements in safety, simplifications, and margins [1,2,15]. It also uses the previous experiences of existing plants and updated technology especially in the control room. The passive concept is also based on PWR and BWR designs, but uses passive means like gravity, natural circulation of fluids, and storage energy for accident prevention or mitigation. It keeps the reactor core protected without operator action for a long time, is greatly simplified, and can be constructed in shorter times due to modularization and prefabrication.

In the last 20 years the observation, analyses of causes, and mitigation have greatly reduced the failure rate of fuel elements in LWRs [16-21]. In PWRs it has fallen from 13 to 4 failures per 100,000 irradiated rods, whereas in BWRs it has fallen from 3 to 2 failures per 100,000 irradiated rods. The differences between the water chemistry of PWRs and BWRs may result in different remedies to reduce the fuel element failures. In PWRs the coolant is a highly reducing environment, because it contains boric acid (up to 2500 ppm) to reactivity control, LiOH to control the pH, and hydrogen to keep the oxygen level below 0.05 ppm. In BWRs it is not possible to control oxygen by adding hydrogen because it boils away. The oxygen level in the water is 0.3 ppm and in the steam is 200 ppm. They also don’t use boric acid. The coolant pressure and core flow rate in PWRs are 136 atmospheres and 36,000 kg/s, and in BWR, respectively, 68 atmospheres and 13,000 kg/s. The coolant temperature in PWR is in the range 290 – 330 ^{o}C and in BWRs is about 320 ^{o}C.

The main failure mechanisms in PWR fuel elements have been grid-to-rod fretting (~ 51%), failures estimated as related to coolant activities (~31%), failures not examined (~9%), failures due to debris fretting (~6%), failures examined nut not identified (~2.5%), and failures due to pellet-to-cladding interactions (~0.5%). In BWRs the failures mechanisms have been debris fretting (~36%), pellet-to-cladding interaction (~18%), estimated as related to coolant activities (~17%), not examined (~16%), examined but not identified (~14%), accelerated cladding corrosion (~3%), and manufacturing related (~2%) [16,17,19,21].

The remedies for grid-to-rod fretting are being introduced through design modifications to enhance the contact between the grid springs and fuel rod cladding, to balance the fluid flow within grids, and to change the vibration characteristics of fuel rods. This effect is important in PWRs because of the higher coolant flow and pressure. The debris fretting is being assessed by the design of more efficient bottle nozzles and the installation of bottom grids that also act as debris filtering together with measures to reduce the introduction of strange materials in the primary coolant system. The effect of pellet-to-cladding interaction can be reduced by the introduction of a liner in the inner wall of the cladding tubes with different composition and microstructure to improve the ductility. This effect is more prevalent in BWRs because the reactor operation is done more extensively by rod control movements, which result in more power ramps in the fuel rods.

Nowadays the burnup of LWRs is about 40 to 45 GWd/tonU, but the goal is to increase it to the range of 60 to 70 GWd/tonU [17,18]. The cladding tubes should remain intact not only during the in-core service, but also during the in-pool and dry storages. The potential failures mechanisms at high burnups are corrosion, pellet-to-cladding interaction, pellet-to-cladding mechanical interaction, brittle fuel rods failures during LOCA, and fuel dispersion during RIA.

Since higher burnups mean in general longer in-core residence times, corrosion will proceed to higher degrees and may become the limiting factor. Improved zirconium alloys, like Zirlo and TM5, may be useful [19,20]. The fuel enrichment must be increased to reach high burnups, as well as the fuel element power. The temperature of the cladding tubes will be higher in PWRs and higher corrosion rates are expected. More acid boric is necessary to control the reactivity in PWRs, as well as more LiOH to control the pH. A higher fuel rod power may increase the subcooled boiling, which, combined with the higher LiOH concentration, may dissolve the zirconium oxide layer and accelerate the corrosion rate. More burnable poisons may be required in the fuel pellet.

Higher burnups mean increased fast neutron fluence, which favor the dissolution of second phase particles in the zircaloy. It is known that this dissolution can accelerate the corrosion rate, albeit diminishing the nodular corrosion [21].

The hydrogen produced by the cladding oxidation is absorbed by the zirconium alloy. If the hydrogen concentration is high enough, hydrides can precipitate and accelerate the corrosion and embrittle the zircalloy. The embrittlement of zircalloy is a complex process, because it depends not only on the hydride concentration, but also on their distribution and orientation within the alloy [19,21,22]. More hydrogen is picked up by zircaloy with increased burnup. The embrittlement reduces the fracture toughness and is very temperature dependent. It is far less important at 350 ^{o}C than at 100 ^{o}C, which is a concern for the storage after irradiation. It is known that non-uniform distribution of hydrides reduces the ductility and fracture thoughness more than uniformly distributed hydrides [19,21]. The non-uniform distribution of hydrides is driven by the heat flux across the cladding wall. The higher heat fluxes that can be produced by the higher reactivity necessary to reach higher burnups increase the tendency to form a hydride rim. Oxide fragmentation, which is more frequent with thicker oxide layers, tends to promote the formation of hydride blisters, which decreases the ductility and fracture toughness. In recrystallized zirconium alloys the hydride concentration increases the fraction of radial hydrides. The hydrogen uptake and hydride formation contribute to dimensional changes that may limit the burnup.

In the fuel pellet, increased burnups means more fission products, higher fission gas release (also during transient), formation of a rim zone at the pellet periphery, fuel-cladding bonding, and swelling (also in transients). The fuel rod inner pressure will be higher at high burnup. The cladding burst during LOCA will occur earlier. The tendency for cladding ballooning and fuel relocation is also higher. Increased embrittlement of the cladding is expected under these situations. The higher rod inner pressure also increases the strain rate of the cladding, which promotes brittle failures [16].

The pellet rim zone is made of fine grains with large bubbles of fission gases in the grain boundaries. The risk for fuel dispersion is higher in the case of cladding failures. In the high burnup stage most of the fissile material is in the periphery. This zone will experience the highest temperature during a RIA event. Fission gas bubbles in the grain boundaries will expand tending to separate the grains.

The pellet-to-cladding interaction [23] threshold should also decrease due to more yields of cadmium and cesium combined with fuel swelling.

3. THE CASE FOR NUCLEAR PLANTS TO PRODUCE ELECTRICITY AND HEAT

The reactor considered for this case in the near to mid terms is the high temperature reactor. This reactor is moderated by graphite and cooled with gas [3-5,18,24]. The output temperature is above 600 ^{o}C, which promotes high efficiency (~ 40 %) for energy conversion. The experiences with the operation of such a type of reactor are the gas cooled reactors (in Britain), and decommissioned reactors (Dragon in Britain, AVR and THTR in Germany, and Peach Bolton in USA). There are also some current and future projects (HTTR in Japan, HTR-10 in China, PBMR in South Africa, and GT-MHR in Russia). South Africa and China plan to build many high temperature reactors of the pebble-bed type in the next decades. The GT-MHR is considered for destruction of more than 94% of Pu239 and the other geologically problematic actinide species [25].

The core of the new reactors coming to the market is made of spherical particles of UO_2 covered with four layers (Figure 1). UC spherical spheres are also considered. The first layer is made of porous pyrolytic carbon to accommodate the volatile and gaseous fission products, as well as the UO_2 swelling. The second layer is made of dense pyrolytic carbon. The third layer is made of SiC and the fourth layer is made of dense pyrolytic carbon. The purpose of the SiC layer is to retain fission products at high temperatures. A ZrC layer in place of the SiC layer is also considered because it can withstand higher temperatures. These layered particles are known as tristructural-isotropic (TRISO). The TRISO particles are dispersed in a matrix of pyrolytic carbon. This matrix may be in the form of spheres or hexagonal blocks. The TRISO particles are designed to withstand temperatures up to 1600 ^{o}C, which is the maximum possible temperature in the worst postulated accident. The level of safety of the high temperature reactors relies greatly on the ability of the TRISO particles to confine the radioactive fission products under this situation [5,26]. As a consequence, the fuel must be manufactured to precise design and manufacturing specifications. Irradiation tests have demonstrated that the TRISO particles have excellent behavior and capacity to retain fission products up to burnups of 75% FIMA (about 800 GWd/ton) [5].

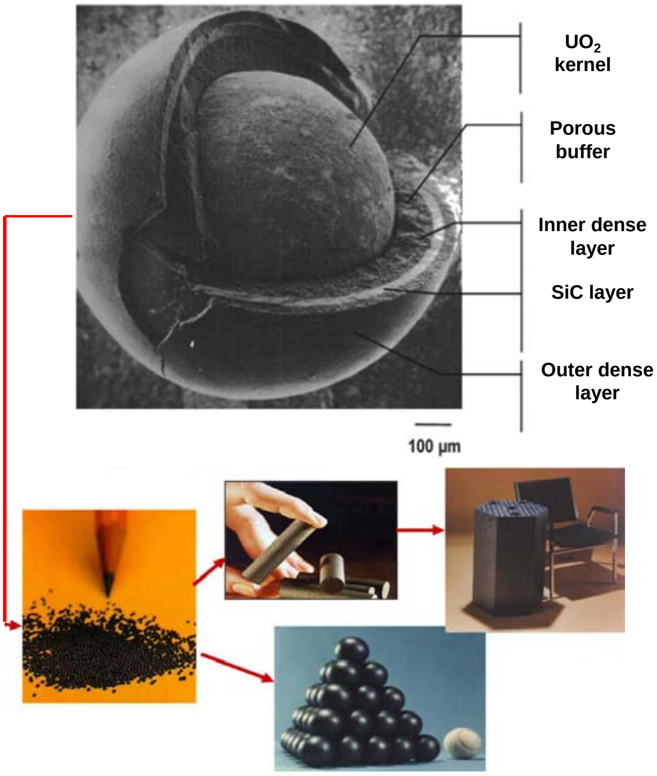

Figure 1 – TRISO fuel for the high temperature reactor (adapted from [5] and [18]).

A pebble-bed reactor of 200 MW has about 450,000 spheres of 6 cm diameter. These spheres have a non-fuel 5mm thick outer layer. Each sphere has about 150,000 TRISO particles of 1 mm diameter. The UO_2 kernel has 0,5 mm diameter. Typical layer thicknesses are: first buffer porous pyrolytic carbon with 50% of the theoretical density – 90 μm, second dense pyrolytic carbon – 40 μm, SiC – 35 μm, outer dense pyrolytic carbon – 40 μm. About 68 billions of TRISO particles are loaded in the reactor core. A robust and reliable manufacturing process for the fuel is necessary to produce such an amount of particles and spheres according to the specifications.

3.1 Manufacturing of TRISO Particles

A sol-gel process is used to obtain the UO_2 spherical particles [5,18,27-29]. A sol is prepared with $UO_2(NO_3)_2$, polyvinyl alcohol (PVA), and other additives. The sol is dropped from small nozzles to form microspheres, which are then gelled in an ammoniacal solution and washed also in ammoniacal solutions. The gel microspheres are dried and calcined at temperatures higher than 700 ^oC. High spherical and density kernels of UO_2 are obtained by sintering in reducing atmosphere at 1700 ^oC. The uranyl nitrate solution can be obtained by the dissolution of U_3O_8 powder in nitric acid. U_3O_8 can be obtained by the calcination of UO_2.

The low density buffer layer of pyrolytic carbon is obtained by the decomposition of acetylene (C_2H_2) by the chemical vapor deposition (CVD) at temperatures from 1300 to 1400 ^oC. The density of this layer is controlled by the gas concentration and the temperature. The dense pyrolytic layers are obtained by the decomposition of a mixture of acetylene and propylene (C_3H_6), also a CVD process. The temperature from 1300 to 1400 ^oC is the controlling parameter for these layers. The deposition rate is lower compared to the one of the buffer layer. The SiC layer is also obtained by a CVD process by the decomposition of methyl trichloro silane (CH_3SiCl_3) with H_2 at 1600 ^oC.

3.2 Manufacturing of Fuel Compacts

This process consists in homogeneously dispersing the coated particles in a graphite matrix. The process steps are granulation of graphite, mixing the granulates with the coated particles, pressing, and sintering. A graphite powder is resinated with a phenolic resin to obtain small granulates, which are sieved for size control. The coated particles are mixed with the granulates in a special equipment to obtain a uniform dispersion. This dispersion is then pressed in the required geometry. The last step is the sintering of the compact in three stages: to polymerize the resin at 200 ^oC, to degas the volatile organic products from the resin under N_2 at 800 ^oC, and to harden the matrix in vacuum at 1800 ^oC [4].

3.3 Fuel Characterization

Due to the large number of kernels and fuel compacts, the quality control should be performed entirely on statistical basis.

The same methods applied to LWR UO_2 pellets are used to characterize the kernels, such as thermo gravimetric analysis for the O/U ratio, He pycnometry and Hg porosimetry for density, as well as the chemical analyses. Specific characterization tools are necessary to control the diameter and sphericity of the kernels and the coating properties (thickness, density, and degree of anisotropy in dense pyrolytic carbon layers).

Other methods are also under consideration for characterization of the coating layers, like scanning and transmission electron microscopy, X-rays diffraction, electron probe micro analysis, secondary ion mass spectroscopy, and electron spectroscopy for chemical analysis [4].

The quality control of the fuel compacts is done by dimensional measurements, weight, fuel particle failure, compact disintegration, burn leaching, and fuel particle homogeneity.

4. CONCLUSIONS

In the case of LWRs, the trend in fuel technology is to reach higher burnups. The integrity of the cladding and structure components of the fuel element must be assured at higher burnups with more deteriorated materials due to mechanical, chemical, and metallurgical mechanisms. The understanding of these mechanisms will result in modifications of the fuel element design, development of improved materials, as well as modifications of manufacturing routines. In view of the good performance of fuel elements in LWRs, the trend of incremental changes in design, materials, and manufacturing is the most probable one to reach higher burnups.

In the case of high temperature reactors, the technology of fuel manufacturing is very different from the one of LWRs. A new manufacturing plant is necessary to obtain the kernels, their coatings, and the fuel compacts. Together with a new technology, it is also expected that this manufacturing plant will also experience many incremental modifications due to the natural improvements in the manufacturing routines and the gain of operational experiences. The wet technology to manufacture the kernels generates large amounts of fluid effluents, which may become a big problem in large scale production. There are experiences in Brazil with the manufacturing of kernels in pilot scale, but very limited experiences with the coatings and fuel compacts manufacturing technology. It is strongly recommendable that these technologies be part of the research and development lines of research groups in Brazil. The same is true for studies concerning the performance of the TRISO fuel and nuclear grade graphite. Cross-cuttings between the LWR and the high temperature reactor technologies, such as the obtainment of kernels directly from the UO_2 powder, should also be investigated to minimize the cost of fuel manufacturing.

The cases for plutonium recycling (MOX fuel) and small nuclear power plants (with power less than 30 MWe) were not considered in this paper.

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