IAEA-TECDOC-286

# UTILIZATION OF PARTICLE FUELS IN DIFFERENT REACTOR CONCEPTS

SURVEY OF WORLD EXPERIENCE

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# FOREWORD

To date, particle fuel is only used in high temperature reactors (HTR). In this reactor type the particles exist of oxide fuel with a diameter of about 0.5 mm and are surrounded by various coatings in order to safely enclose fission products and decrease the radioactive release into the primary circuit.

However, it is felt that fuel based upon spherical particles could have some advantages compared with pellets both on fabrication and in-core behaviour in several reactor concepts. This fuel is now of general interest and there is a high level of research and development activity in some countries.

The development of gel process for the fabrication of spherical particles began in the 1960s. The first international symposium on sol-gel processes was organized by CNEN in 1967. It was followed by an Agency's specialists' meeting in 1968. Three years later the IAEA convened a panel at Vienna to examine the status and to evaluate the likely future role of sol-gel processes in fuel fabrication.

In order to collect, organize additional information and summarize experience on utilization of particle fuels in different reactor concepts, a questionnaire was prepared by the IAEA in 1980 and sent to Member States, which might be involved in relevant development. The survey has been prepared by a group of consultants and is mainly based on the responses to the IAEA questionnaire. It will be used for Member States, institutes and experts interested in the topic.

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## 1. INTRODUCTION

Actual nuclear reactors are almost exclusively equipped with fuel elements containing ceramic fuel. Metallic fuels have been used in gas cooled power reactors of the Calder Hall type, in material testing reactors, and in experimental fast breeders; cermet fuel elements are applied to material testing reactors and high-flux reactors.

The fuel elements of light water reactors are built up of a number of fuel pins which are arranged to quadratic bundles. The characteristics of a LWR fuel pin are pellets of slightly enriched uranium oxide and a Zircaloy cladding. Likewise the annular fuel elements of heavy water reactors consist of fuel pins, made of Zircaloy and uranium oxide pellets. In the hexagonal fuel elements of fast breeder reactors oxide pellet fuel, clad with steel, is utilized. The fuel concept of the advanced gas-cooled reactor (Hinkley Point B) is a combination of oxide pellet fuel and steel cladding as well. The smallest fuel unit of the gas cooled high temperature reactor is not the pellet, but the microsphere. The kernels consist of oxide or carbide fuel, each kernel being enclosed in several spherical layers of pyrolytic carbon and one layer of silicon carbide in most of the cases. These particles are overcoated with carbon and subsequently compressed with carbon to globular fuel elements or filled into the fuel channels of a multihole graphite block.

Particle fuel can be fabricated by agglomeration of powders, fusing granulated powders in a drop furnace or by forming and solidifying droplets on wet chemical route. Intensive work on wet route techniques for particle fabrication brought to light a series of advantages inherent in the gel method. These benefits are especially obvious when plutonium bearing fuel or highly active reprocessed material is to be treated. For these reasons fuel based upon spherical particles produced by a gel method offers a promising potential also in the field of commercial LWRs and FBRs. This might be accomplished on one side by substituting spherical particles for powders which are difficult to handle in the production of pellet fuels and on the other side by avoiding pellet fabrication at all by filling fuel pins with appropriate mixtures of spherical particles directly.

The development of gel process for the fabrication of spherical particles began in the 1960s. The first international symposium on sol-gel processes was organized by CNEN in 1967. It was followed by an Agency's panel in 1968 and an US symposium at Gatlinburg in 1971. Three years later the IAEA convened a panel at Vienna to examine the status and to evaluate the likely future role of sol-gel processes in fuel fabrication. In order to collect, organize additional information and summarize experience on fuels based upon spherical particles, a questionnaire was prepared by the IAEA in 1980 (Appendix A) and sent to Member States, which might be involved in relevant development.

This survey has been prepared by a group of consultants (Appendix B) and is mainly based on the responses to the IAEA questionnaire.

## 2. FABRICATION OF SPHERICAL PARTICLES

Spherical particles of nuclear fuel can be fabricated by agglomeration of powders (dry route) or by forming and solidifying droplets (wet chemical route).

The powder agglomeration processes use finely ground powders of respective oxides which are mixed with carbon black and slight quantities of binders and water or organic solvents. This powder is granulated to form seeds which are then grown to the correct size in a vibrating pan fed with fresh powder. After drying, carbonising the binder and burning out the carbon, the kernels are sintered to oxide. This process has been used, inter alia, by the Dragon Project to produce HTR fuel [1].

Dragon Project also developed an improved fabrication route based on the formation of green spheres by the binderless agglomeration of submicron powders [2].

To produce carbide particles there were used methods such as sintering of converted kernels at 2000°C and subsequently dropping them through a vertical graphite tube furnace heated to about 2600°C or through a plasma [3.4]. In another process granules were overcoated with carbon black and then melted in bulk at 2500°C in graphite crucibles [5]. Carbide fuel kernels have been used in the Dragon, Peach Bottom, AVR and Fort St.Vrain reactors.

Uranium oxide and uranium carbide kernels have been produced at densities up to 95%, in sizes ranging from 50 to 1500 $\mu$m dia. The disadvantages of powder agglomeration processes are that the quality of the powder must meet stringent requirements in regard to fineness and sinterability, that a wide grain spectrum is often produced and that with oxide kernels low densities of about 80% th. are attained in general [3,6]. A process which was suitable only to produce kernels of about 1 mm dia or more dealt with compressing of powder to a spherical shape in a miniature die.

To produce spherical particles there exist several wet chemical processes, all of them having in common that solutions or sols of fissile or fertile materials are dispersed into uniform droplets. The spherical shape of these droplets is fixed by gelation which can be achieved by either a precipitation or a dehydration reaction. After washing of the gel spheres a heat treatment, consisting of drying, calcining and sintering is carried out.

The ORNL sol-gel process [7] was originally developed to prepare $(Th,U)O_2$ microspheres (SOLEX-process) and was modified to produce spherical particles of $UO_2$ (CUSP-process) and $PuO_2$ (improved APEX-process).

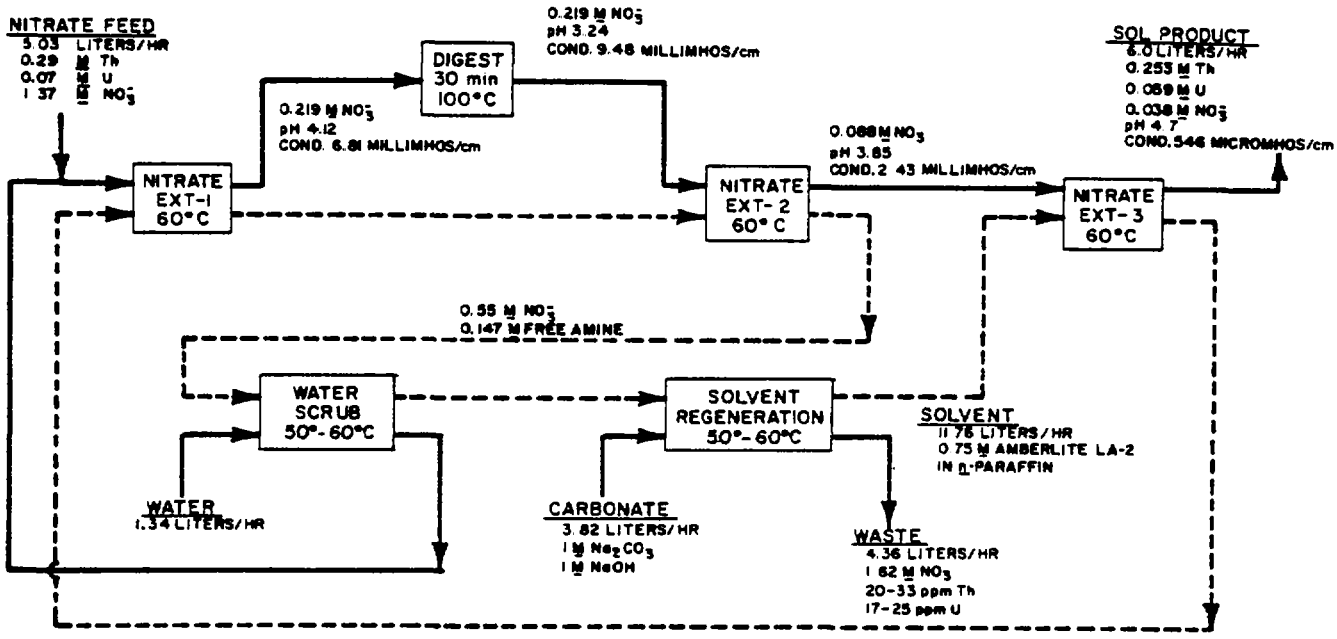


Fig. 1. Countercurrent Flowsheet for the Preparation of 120 kg of 81% $ThO_2$-19% $UO_3$ Sol by Amine Solvent Extraction

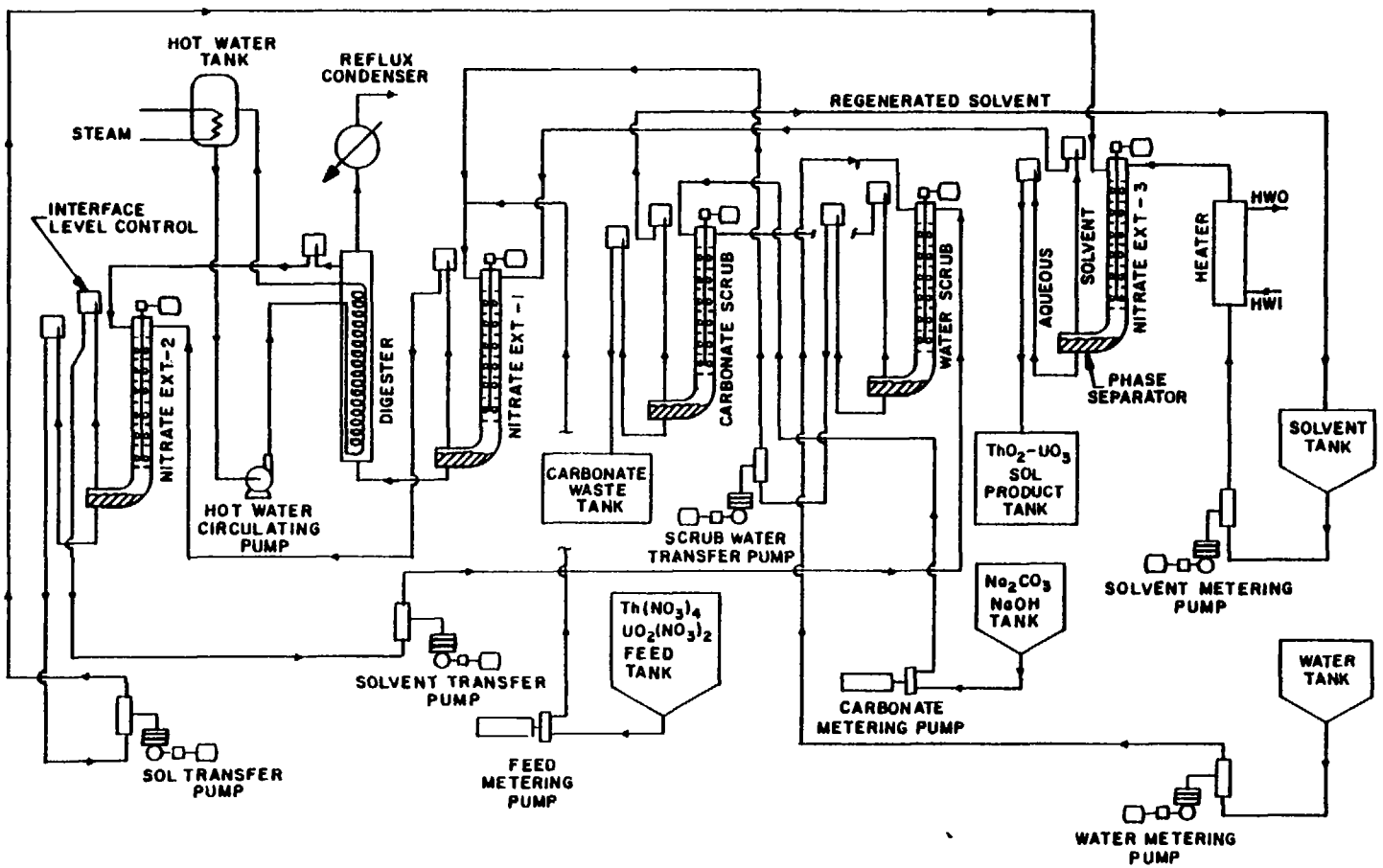


Fig. 2. Equipment Flowsheet for the Preparation of $ThO_2$-$UO_3$ Sol by Amine Solvent Extraction

Sol preparation according to SOLEX-process was by means of amine extraction from ague solutions of thorium-and uranyl nitrates (Fig.1.2). The sol was about 0,3M(Th+U) as-prepared and was concentrated by evaporation to 1,5M(Th+U) for use in sphere forming. In the ORNL microsphere forming process (Fig.3), gel microspheres are produced as water is extracted from droplets of sol by 2-ethyl-1-hexanol (2EH). Addition of surfactants to the 2EH is required to facilitate formation of $ThO_2$-$UO_3$ gel spheres.

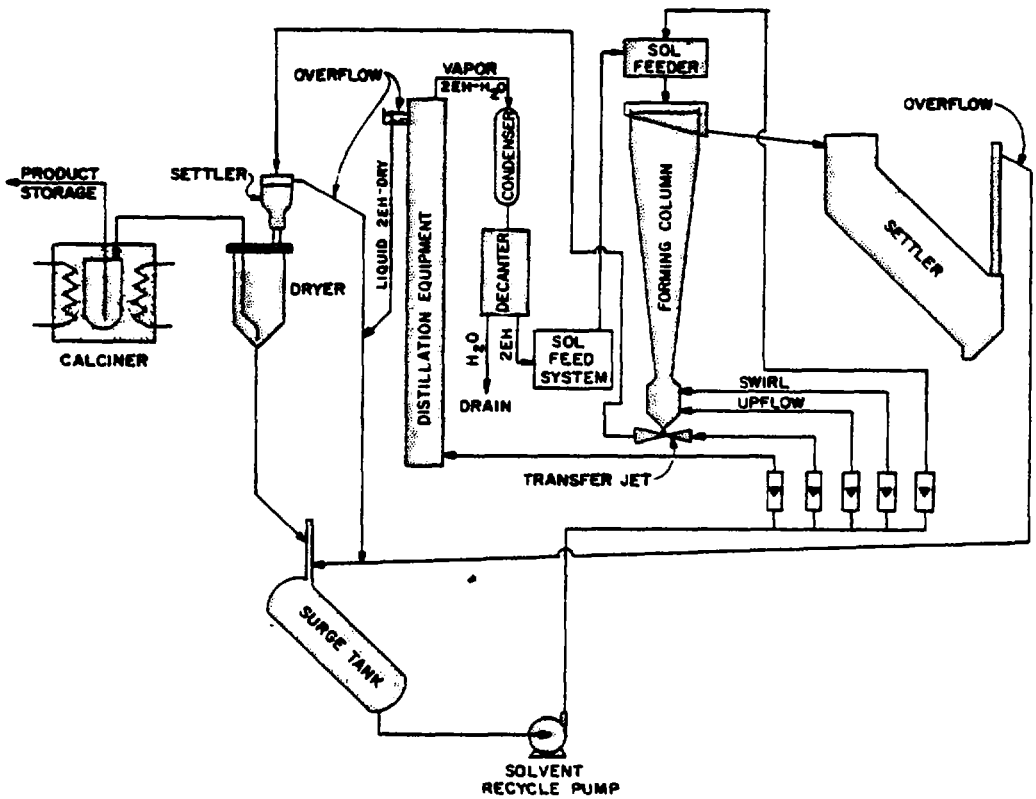


Fig. 3. Equipment and Process Flowsheet for the Preparation of $ThO_2$-$UO_3$ Gel Microspheres

The device used for sol dispersion was a multiple two-fluid nozzle. The hold up time in the sphere forming column was about 45 minutes for the production of a 450 $\mu$m dia. product. Microspheres were collected by the settler, brought into the dryer, filtered and dried in an argon-steam purge to 200 - 250°C. Subsequently the microspheres were calcined at 1150° in air to remove carbon and to densify the spheres. Reduction of the $UO_3$ to $UO_2$ was accomplished by heating in 4% $H_2$-Ar at 1150°C. The dried gel shrinks about 63% by volume, and 30% diametrally. The calcined products had densities of 95 to 98% of theoretical, 20 to 40 ppm carbon content and O/U ratios of 2.001 to 2,020.

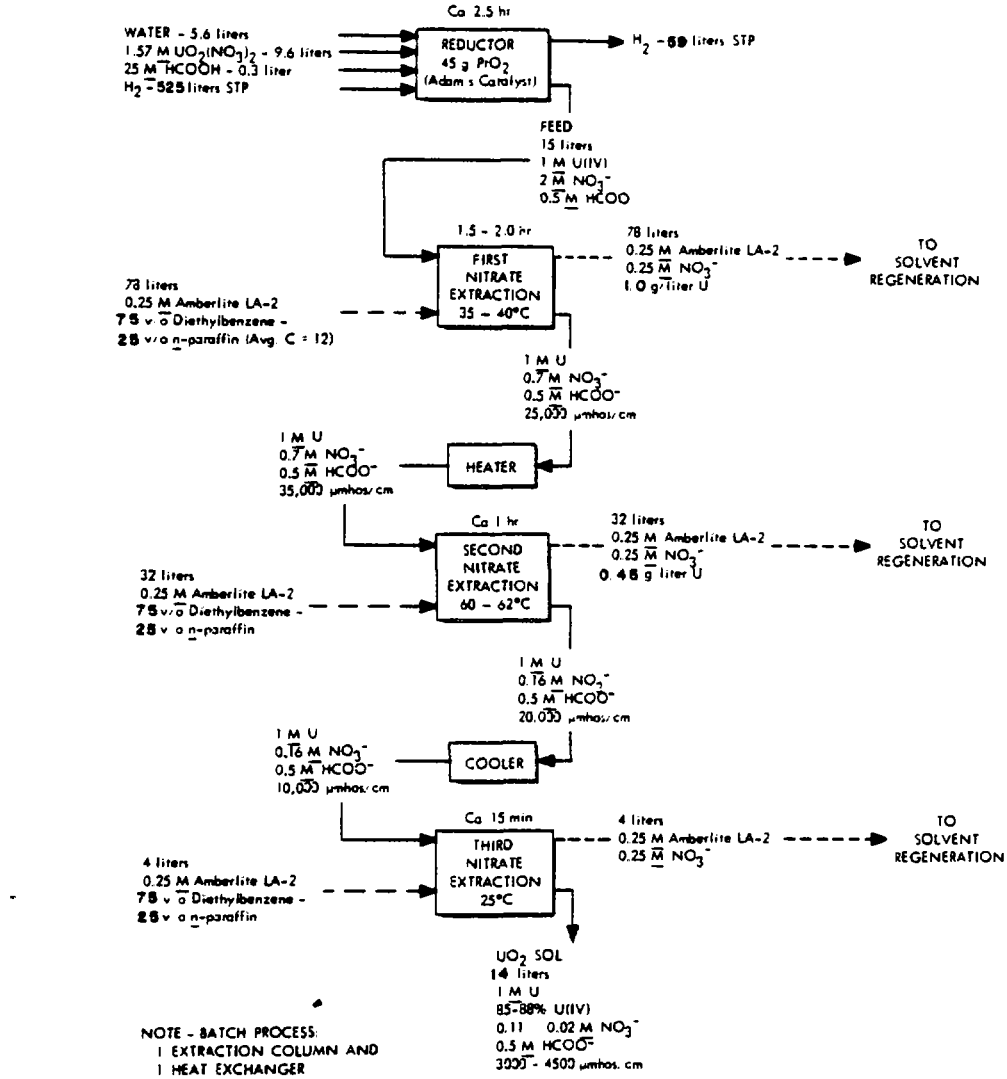


**Fig. 4. Chemical Flowsheet for Preparing $UO_2$ Sol by the CUSP Process**

The first step to produce $UO_2$ microspheres by the CUSP process (Fig.4) was the reduction of a solution of uranyl nitrate and formic acid, using hydrogen in the presence of a platinum oxide catalyst. The extent of reduction to U(IV) is monitored by a platinum VS glass electrode pair. Vigorous agitation is required during reduction to avoid gelation of the solution by local formation of ammonia. When reduction is complete the U(IV) solution is drained from the reductor, leaving the Pt on the stainless steel filter. The catalyst is reoxidized with $HNO_3$ for reuse.

In the next step (Fig.5) a stable 1M $UO_2$ sol was prepared by Amberlite LA-2 (a secondary amine). Some uranium is entrained in the amine extractant in the form of negatively charged colloidal particles. A three-stage extractant system consisting of two cleanup stages and an

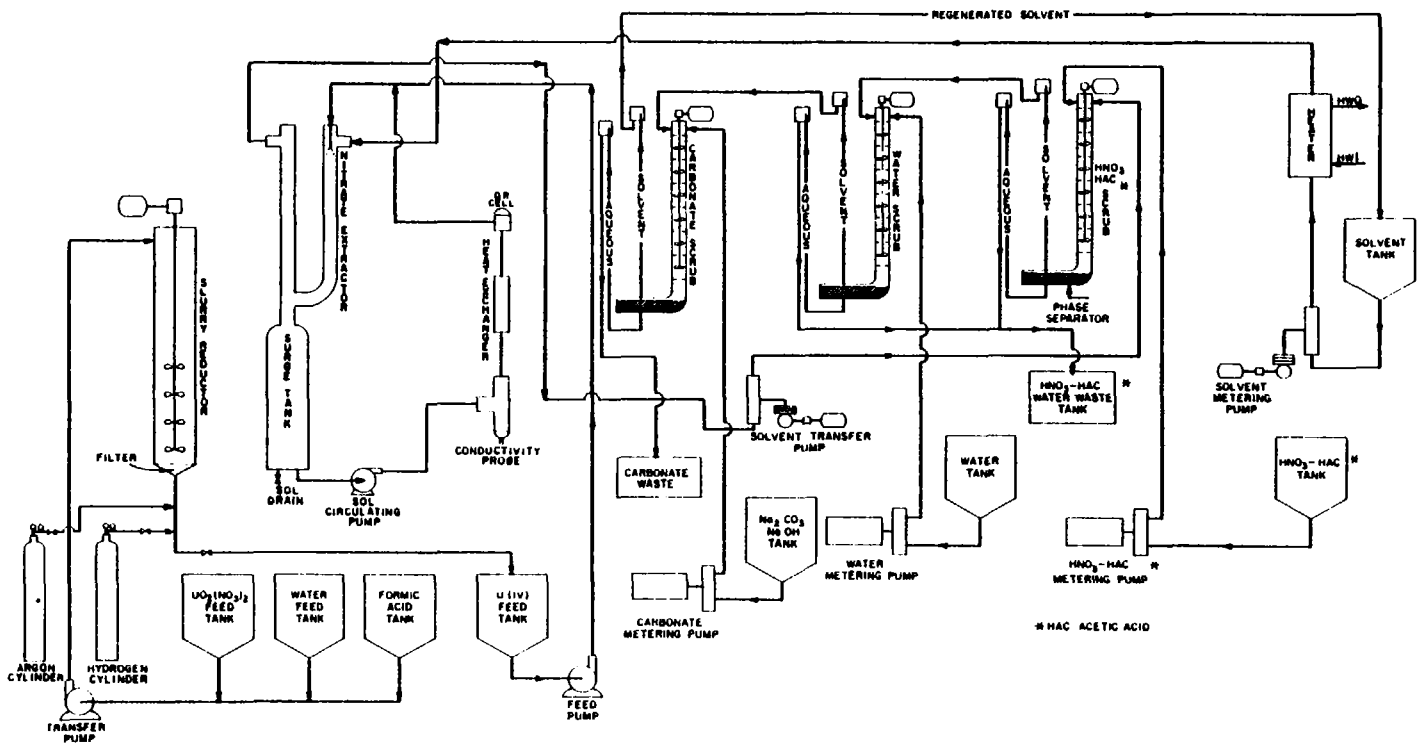


Fig. 5. Equipment Flowsheet for the Preparation of $UO_2$ Sol by the CUSP Process

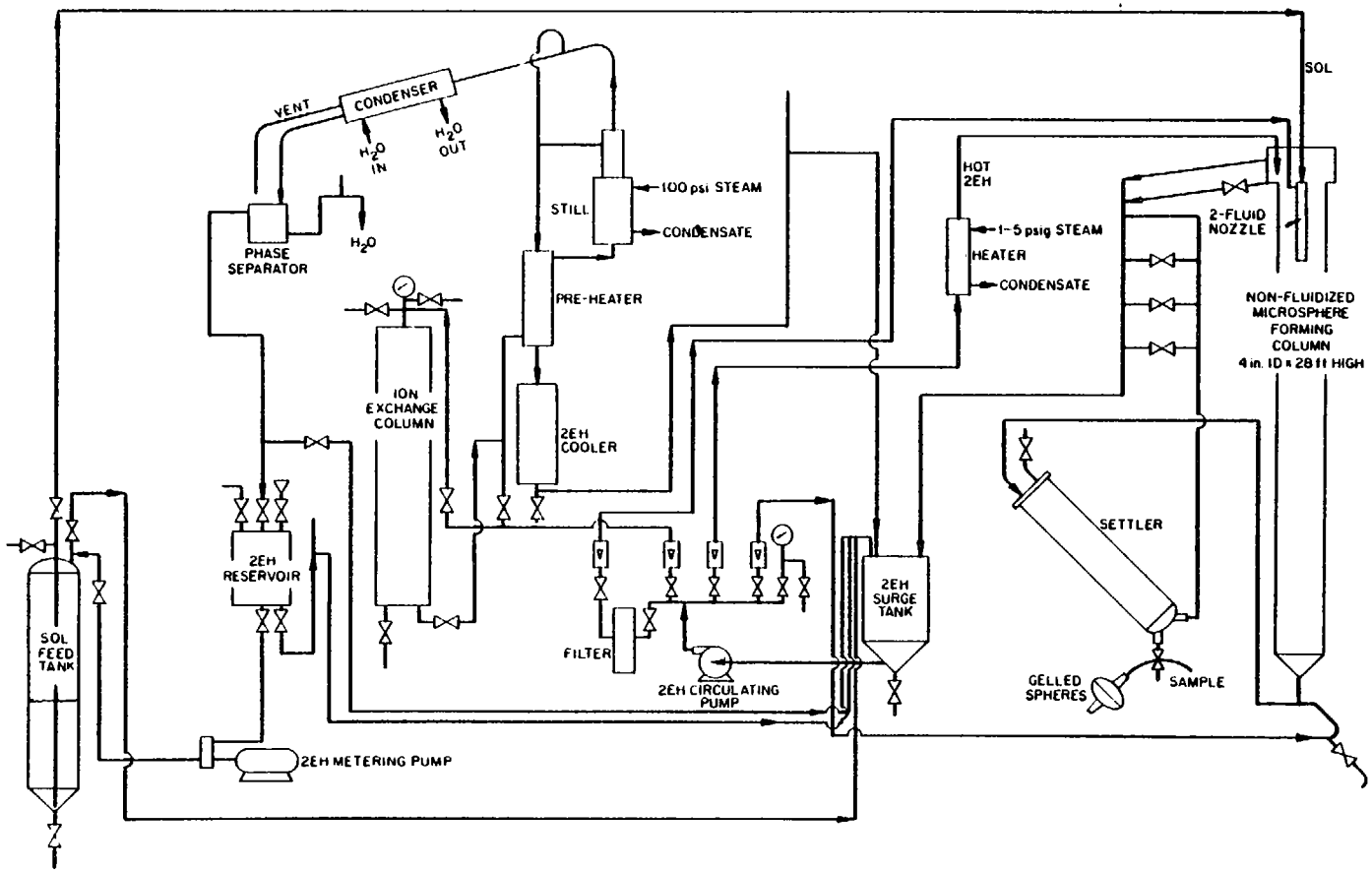


Fig. 6. Equipment Flowsheet for the Unfluidized Microsphere Forming System

amine regeneration stage was found to satisfactorily recover the uranium and regenerate the amine. Microspheres were formed from the $UO_2$ sol in a 28 feet high unfluidized column (Fig.6). The temperature of the 2-ethyl-1-hexanol was 50 to 80°C. To avoid impurity build up in the 2EH, which had been shown to cause serious problems, 2EH was continuously purified, using a weak-base ion exchange resin. Water content in 2EH was controlled by distillation. Finally, again the green microspheres were subjected to extensive drying and firing procedures.

The ORNL plutonia sol-gel process (improved APEX process) is characterized by: (1) extraction of $HNO_3$ from an agueous $Pu(NO_3)_4$-$HNO_3$ feed with a long chain alcohol, e.g. n-hexanol until an $NO_3$/Pu ratio of 0,9 is obtained; (2) seeding the feed solution with sol before or during extraction to produce a micelle structure of $PuO_2$ crystallites; (3) thermal denitration of the sol by drying and baking it; (4) resuspension of solids in water. The sol preparation flowsheet is shown in fig.2. The extractibility of Pu limits feed Pu concentration to 0,1 to 0,2 M if excessive Pu extraction and consequent recycle is to be avoided. Extraction with n-hexanol rather than an amine is used because alcohol gives excellent phase separation, and extracted Pu can be recovered readily by water stripping. The seed polymer must be added to the feed solution before the $NO_3$/Pu ratio is less than about 4,6. Minimum seed concentrations are 20% of the total Pu by weight. Following the final extraction to a $NO_3$/Pu ratio of 0,8-1,2 the sol is evaporated to dryness and heated for a minimum of 15 minutes at 230°C in dry air. Solids are resuspended in water to form a 1-2 M $PuO_2$ sol (0,1-0,2 $NO_3$/Pu mole ratio) which can be mixed with CUSP $UO_2$ sol and formed into microspheres by the 2EH process. To improve size uniformity of microspheres vibration nozzles have been developed. The sol is vibrated by a piston in the sol at the entrance to the capillary or orifice where sol enters the drying 2EH.

The gelation processes developed at ORNL are rather complicated and had difficulties with particle sizes, greater than 600 $\mu$m, particularly with urania. These processes are scarcely used nowadays and the chemical gelation processes, developed in Europe, are preferred, which use ammonia to cause rapid gelation of droplets. Ammonia gelation can be accomplished either externally, via ammonia gas and ammonium hydroxide, or internally via an added ammonia generator such as hexa methylene-tetramine. The use of an organic polymeric thickener in order

to provide a gelling support in which the precipitation of alkale insoluble heavy metal compounds takes place is a major distinctive feature of the gel precipitation process with external gelation. The process is a very versatile one to produce all types of reactor fuel and fertile materials in current use with only slight changes in composition.

The NUKEM-process [5,8] has been used mainly for the production of $(U,Th)O_2$, $ThO_2$, $UO_2$, $UC_2$ and $UC_xO_y$ kernels. To fabricate oxide kern the uranyl nitrate solution or/+ thorium nitrate solution of PM 3,5 to 4,0 is mixed with polyvinyl alcohol solution (PVA), other additions and water. The feed solution is passed through nozzles, developing jets of liquid in air which form uniform droplets under the influence of an electromagnetic vibration system. The droplets are solidified by reaction with ammonia gas and are collected in ammonia solution. The quality of the spherical form depends on the type and quantity of the auxiliary materials e.g. PVA, dioxane, acetamide, urea or glycine. Then the particles are washed free of $NH_4NO_3$ with ammonia water, dehydrated with isopropanol and dried at 80°C in a vacuum with recovery of the isopropanol. The particles are calcined in air at 300°C to remove the auxiliary materials and to decompose thermally ammonium diuranate and thorium oxide hydrate. The kernels are then sintered in hydrogene to heavy metal dioxide of 97-99% of the theoretical density.

This process is also applicable to produce carbide or oxicarbide fuel by appropriate addition of carbon black to the uranyl nitrate solution. The kernels are calcined and converted under argon in a sintering furnace and are finally sintered at 1850°C. In order to prepare molten $UC_2$ kernels the dried kernels are calcined under argon and then are coated individually with carbon black in a drum. These coated kernels are converted in bulk to carbide in a graphit cucible under vacuum within about two hours, and are finally melted at a temperature of 2500°C. The carbon black layer is then removed by screening.

A typical feed solution composition for the preparation of particles of $UO_2$-20% $PuO_2$ according to the SNAM process [9] is the following: $UO_2(NO_3)_2$ 0,52M, $Pu(NO)_3)_4$ 0,13 M, tetrahydrofurfuryl alcohol 30%, hydroxypropylymethyl cellulose 0,3%, free $HNO_3$ 0,5M. The gel particles obtained from dripping in $NH_4OH$ this feed solution are aged in $NH_4OH$, washed, dried, and calcined in air at

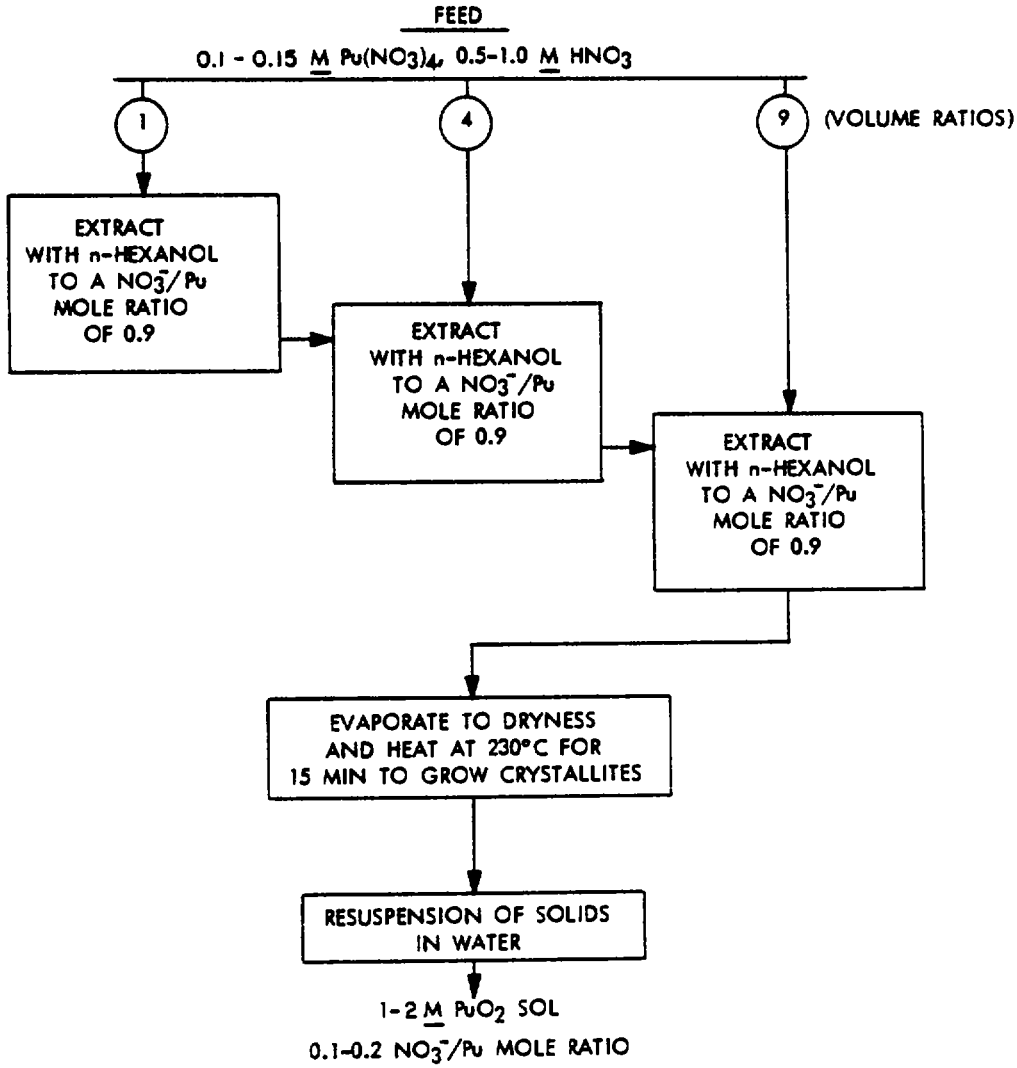


Fig. 7. Modified APEX Process for the Preparation of Low Nitrate $PuO_2$ Sol

450°C and sintered in Ar-4% $H_2$ at 1300°C to yield $UO_2$-$PuO_2$ particles having densities higher than 95% th.

A precipitation process has been developed at KFA Julich which is expected to be most suitable for $^{233}U$ refabrication. This process does not involve any polymeric thickener [10,11]. An aqueous solution of $UO_2(NO_3)_2$ and $Th(NO_3)_4$ is preneutralized by gaseous $NH_3$ at 95°C. The 2-3M sol is dispersed into droplets and the precipitation reaction is completed in $NH_4OH$(1%). Processing of solutions of $UO_2(NO_3)_2$ free of thorium is effected by addition of urea and $NH_4OH$ to the broth. Droplets are gelled in $NH_4OH$ 7M and aged at 90°C for 30 minutes in $NH_4OH$ 7M. The fresh particles are dried at 180°C and subsequently reduced and sintered as usual.

The chemical principle of the gel precipitation process with internal gelation is the precipitation of oxyhydrates from nitrate solutions by means of ammonia, which is generated in the nitrate solution by the thermal decomposition of hexamethylenetetramine. In the solution urea is present to protect uranyl ions against premature precipitation by complex formation. The starting solution according to KEMA UI (VI) process [12] is a substoichiometric solution of uranly nitrate, about 3 m U/l with a $NO_3$/U molar ratio of about 1,5 which can additionally contain plutonium or thorium nitrates. This solution is mixed with 1,4 volumes of a solution of hexamethylenete tramine and urea (both 3 m/l) under cooling below 10°C in order to keep the decomposition rate of hexa, which is induced by the heat of neutralisation, as low as possible.

The feed is dispersed into droplets of the required size which are gelled in a water immiscible organic liquid of about 90°C. At this temperature the decomposition of hexa supplies ammonia to the system resulting in a conversion of uranyl-urea complexes into uranylhydro-complexes, followed by the precipitation of $UO_3$ hydrates. The ammonium nitrate, formed during the gelation reaction, residual hexa and urea are removed by leaching the fresh microspheres in a column with ammonia solution. The ammonia solution converts the $UO_3$-hydrate droplets into ammonia-uranate spheres. The spheres are dried in air at 70-80°C and then are calcinated at 450°C in air to remove the volatile compounds. Finally this material, a mixture of $UO_{2,9}$ and $UO_3$, is reduced and sintered during approximately 5 hours at 1600°C in a reducing atmosphere.

EIR adopted the process for the manufacture of mixed carbide microspheres [13]. The feed consists of a 1,5M(U,Pu)- nitrate solution which is added to a mixture of hexamethylene tetramin and urea containing finely dispersed carbon black. The feed is then spread into very fine droplets by means of special nozzles (Fig.8). A very narrow size-distribution can be obtained with vibrated nozzles or with a sheared system. The gelation occurs in silicon oil at 80-100°C, followed by three wash-treatments with $CCl_4$ to completely remove the silicon oil. The fresh microspheres are leached in an expanded bed column with 3N-ammonia solution. After drying at 150°C, the kernels are precalcined at about 550°C under an inert or reducing atmosphere, in order to remove traces of organic matters. The final step consists of a carbothermic reduction and sintering up to temperature of 1800°C, using a 17 hour cycle to heat, hold and cool down.

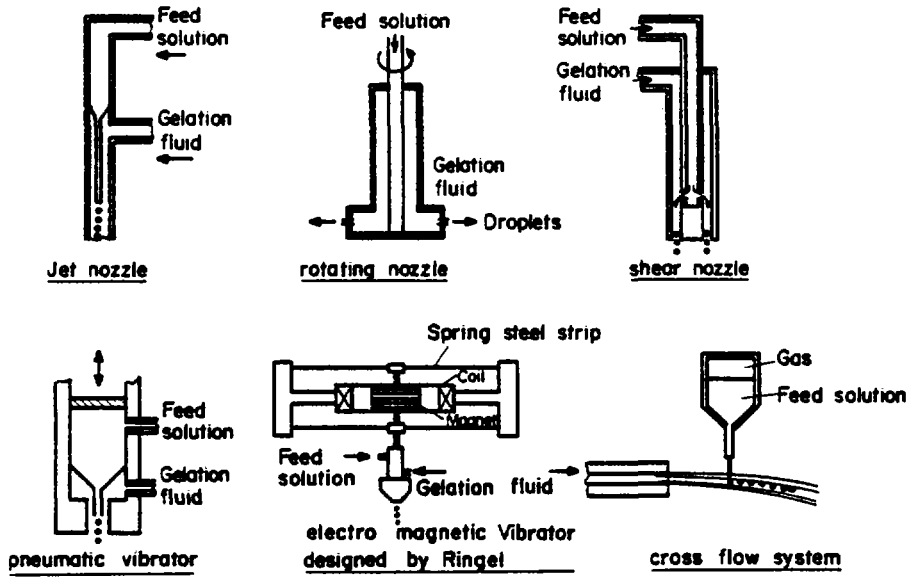


Fig. 8. VARIOUS DROPLET GENERATORS

An improved internal precipitation process applicable to all kinds of particle fuel has been developed in Germany [14]. The nitrate solutions of U,Pu and Th generated in the reprocessing can be used as starting products. At the conditioning stage the heavy metal solutions are concentrated separately up to 800g U/l, 600g Pu/l and 800g Th/l, respectively, and denitrated simultaneously by addition of formaldehyd up to a $NO_3$/Me ratio of about 1,7. This stoichiometric ratio can be further reduced by neutralization with ammonia. The solutions so obtained are made to form complexes with urea, cooled to about 5°C, mixed in the ratio desired for mixed oxide fabrication and added to HMTA. At this point of the process graphite or soot is added for carbide or oxicarbide fabrication. The particles are formed by dropping the conditioned solution into tetrachlorethylene heated to about 95°C. Tetrachlorethylene offers the advantage of being non-inflammable and easily volatized, so the kernel wash with organic solvents required in oily heat transfer media can be avoided. The kernels, insensitive to pressure reach vibrating sieve grooves where the liquid is separated and recycled and the particles are dried. In the subsequent washing stage a 2% ammonia solution heated to 35°C is pumped in a counter current flow to the particles in a vertical column (Fig.9). Drying and sintering of the particles are in the usual way.

A special wet-chemical process is the resin process originally developed by ORNL [15] in which cation exchange resin beds of suitable size are loaded with uranium from uranyl nitrate solutions.

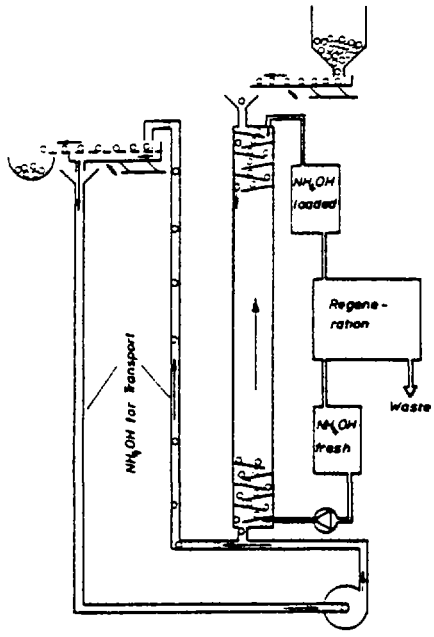


Fig. 9. Technological principle of continuous particle wash

Spherical particles can also be produced by polycondensation processes with solidification of two organic compounds containing uranium, thorium or boron in liquid state [16]. These processes necessitate the decomposition of the resins before sintering of the particles.

## 3. PARTICLE FUEL FOR DIFFERENT REACTOR LINES

A special feature of the gel processes for the production of spherical particles is their versatility. By only small changes in procedure it is possible to produce with the same equipment oxides and carbides, all of the fissile and fertile chemical elements which can be used as a nuclear fuel in different reactor types. Owing to its simplicity the gel process is especially suited for the production of refabricated fuel. The spherical particles are processed to fuel elements according to the requirements of the differrent reactor lines.

On an industrial scale particle fuel is only used in HTRs to date. Coatings of pyrocarbon are deposited onto kernels in fluid bed reactors. Typically acetylene is used to make low density buffer coats and methane and propylene to make high density isotopic coats. Additionally silicon carbide coatings are deposited by CVD techniques to improve fission product retention of coated particles. The coating units are operated so that multilayer coats are deposited consecutively without

interruption of the process until layers are deposited. These coated particles are dispersed in a graphite matrix and fuel element spheres are formed by pressing and heat-treatment (AVR, THTR) or compacts are made for the block elements (GAC).

Starting from gel spherical particles the manufacture of fuel pins for LWRs and FBRs can be ahcieved by filling proper mixtures of particles of different sizes into rods (sphere-pac technology) or by compressing of calcined gel-particles to pellets followed by the usual rod-filling.

Sphere-pac technique requires at least two sizes of spheres to achieve a smear density of about 80%[17]. This smear density is suited for FBR pins. Usually the infiltration method is applied. First the large spheres are loaded into the rod in an efficiently packed geometry by low-to medium- energy vibration. Then the fuel column is fixed by a sieve and the small sized fraction is infiltrated into the column by vibration filling the voids between the large spheres. Usual particle sizes are 800 and 30 $\mu$m diam.[18.19].

When the fuel has been compacted, the pin is evacuated, filled with helium gas and the end cap welded on. After decontamination the pins are submitted to a series of quality control operations including visual inspection and gamma scanning. A great number of sphere-pac uranium/plutonium oxide and carbide pins have been produced and irradiated up to now. The irradiation tests indicated that these fuels perform at least as well as pellet fuels under the same conditions and in some respects - (fuel cladding interaction, heat transfer, overpower transients), better.

Spnere-pac requires tnree sizes of spheres to achieve a smear density approaching 88%, these sizes should have diametral ration of about 40:10:1. The preferred method is to incorporate all the fissile content into the two large sizes which allows the fines to be made in a separate operation using only fertile isotopes - the "fertile fines" method [20]. However, the sphere-pac process is not limited to use fertile fines, and all sizes of spheres can have the same composition.

Simultaneous loading of all three (or four) size fractions shows promise of overcoming the problem of excessive loading times for LWR-rods, caused by the infiltration method. Conventional power

blending methods permit extensive segregation for the wide, range of sphere sizes. A particle blending technique amenable to remote operation is being developed [21].

Possible variations in density within the particle column can be detected by gamma-scanning, e.g. with an Ir-192-source. If properly set the deviations are lower than with pellet columns due to dishing of the pellets [14]. For LWR applications fuel rods have been filled with gel spheres of 30, 300 and 1200 $\mu$m diam and with particles of about 10, 100 and 1000 $\mu$m diam. [20,22].

A hybrid fuel pellet fabrication concept, also called "spere-cal process", in which dried and calcined gel microspheres are pressed into pellets is being developed in some countries. This method combines the front end of the gel-sphere flowsheet with the back-end of the standard pelleting flowsheet and readily meets LWR requirements of high fuel density. Further this approach has the benefit of yielding a fuel form that is already licensed which seems to be of importance in regard of both LWR and FBR lines [11,23,24,25,26].

There are a number of advantages of particle fuel compared with pellet fuel produced from powder in the usual way. These benefits obvious in lab and pilot plant production have to be proved yet on an industrial scale. The particle route is simple, it requires fewer steps for fuel fabrication than powder processes (Fig.10). The fissile and fertile material contained in the feed solution is dispersed directly in the final form of the particulate fuel. Further, the process is very versatile to produce all types of ceramic nuclear fuel essentially with the same equipment. The simplicity of the wet fabrication route of particles gives use to numerous advantages on safety and economies of plutonium bearing fuel production.

The gel particle route is especially suited for the fabrication of fuel from nuclear materials coming from reprocessing facilities. When plutonium of LWR origin or multicycled fuel is used for fuel fabrication experience has shown that mechanization of the production line including remote operation is necessary together with extensive use of gamma and neutron shielding. Fabrication of microspheres offers an excellent potential in this respect, for the materials are contained in liquids up to the thermal stages. This together with the free flowing

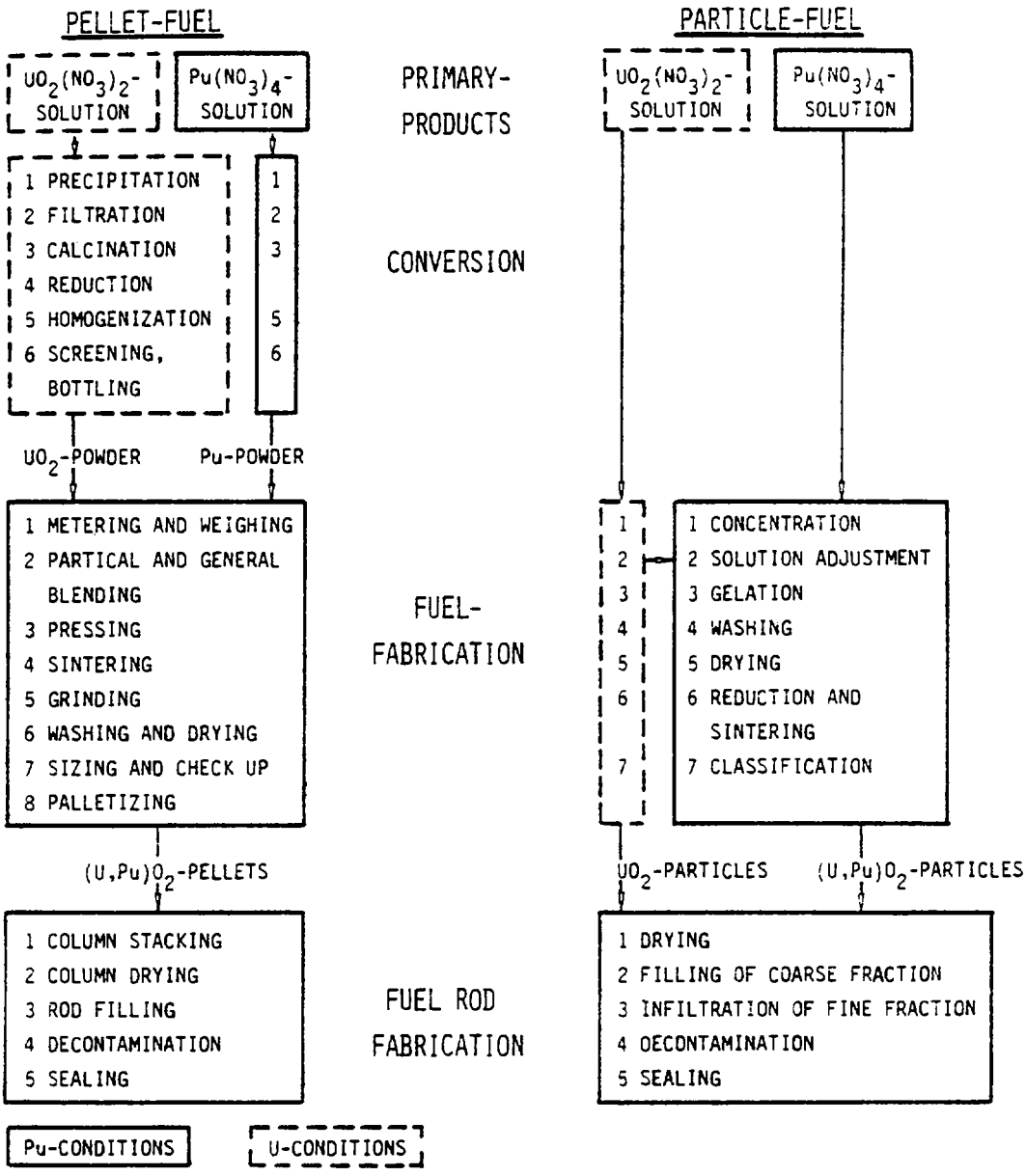


Fig. 10. Flowsheet of fuel rod fabrication with pellet or particle fuel

characteristics of the dry fuel, facilitates automatic remote processing. Wet processing reduces dust load drastically, for powder milling and powder granulation is unnecessary and mechanical finishing is eliminated on the sphere-pac route. This minimizes operator dose, facilitates primary shielding and eases decontaminaition and maintainance of equipment. Recycle is reduced in comparison with pellet manufacture when the pellet product specification necessitates grinding and inspection operations. Grinding gives rise to Pu active slurry and inspection results in some recycle. Transfer and handling problems are

reduced for uniform spheres present fewer handling difficulties than powder, pellets or irregular granules. The necessary equipment is simpler and more reliable. The sintering temperatures of particles are lower because the small crystallites produced in the gels sinter more easily to a high density. Coprecipitation of uranium and plutonium results in an excellent product quality regarding plutonium homogeneity. Disturbances during reprocessing owing to $PuO_2$ enriched grains which are insoluble in $HNO_3$ are avoided by the formaition of homogeneous mixed oxides or carbides.

Compared with pellet fabrication the particle route is not only safer to the operator and the environment but it is also more proliferation and diversion resistant. The feed preparation flowsheet is compatible with co-stripped uranium and plutonium streams from the reprocessing plants, thus leading logically to an integrated fuel reprocessing and fabrication plant. No intermediate conversion to the oxides is involved, thus avoiding handling and storage of pure plutonium oxide. In addition wet processes can accomodate feeds which are spiked by partial fission product recycle. Again this prevents unauthorized access to the fissile materials.

Irradiation tests did not indicate disadvantages in the operating benavior of particle fuel columns compared with pellet rods. Sphere-pac pins showed extremely smooth diameter profiles in contrast to the pelletized pins, which were characterized by steep local changes in diameter [27]. The microspheres tend to form a flexible annulus at the inner cladding surface and any cracks within the restructured fuel do not extend radically to the clad wall [28]. Hard mechanical interactions between fuel and cladding was not observed on any of the 70 cross section micrographs of spere-pac pins irradiated in the HFR at Petten [22]. However, at several locations of the cross sections of the pellet pins fragments of the restructured pellets were pressed into the zircaloy cladding by the adjacent pellet surface. Irradiation tests on mixed oxide fuels have shown that sphere-pac fuels are much less reactive in PCCI than pellet fuels [7].

It has been observed that the heat transfer is better for particle columns than for pellet pins, so that after restructuring the particle fuel has a higher thermal conducitivity [7,29]. Further the

occurrence of hot spots in sphere-pac rods is very improbable because of good fuel homogeneity [30].

## 4. ACTIVITIES

The work completed in the respective countries, the current activities and the future plans are summarized subsequently.

### 4.1 BELGIUM

Between 1960 and 1975 CEN/SCK and Belgonucleaire studied particle fuel elements in regard of the possibility to use them in water reactors and HTGRs.

For water reactors the research was aimed at replacing pellets by vibrocompacted powders, mainly in view of recycling plutonium in these reactors. Several concepts were considered and it was tried to produce three different geometric fractions of powders. The irradiation benaviour of particle fuel proved to be excellent, in both test and power reactors. The studies were ultimately abandoned, mainly on economic reasons, but also on account of the dust formation during thepulverization process and the sifting of plutonium - bearing powders.

Studies an HTGR-fuels were carried out mainly under the Dragon Project. Microspheres of urania and $(U,Pu)O_2$ were produced by agglomeration of powders. These kernels were coated in the usual way and compacted. The irradiaition benaviour of these particles proved to be excellent. Coating tests were also carried out with aluminium nitride and $Al_2O_3$ or MgO. These investigations were abandoned owing to the cessation of the Dragon Project.

The particle fuel studies were resumed in 1979 with a different aim - namely fast reactor applications. Particles of mixed uranium - plutonium oxides, containing up to 35% $PuO_2$ are produced by a wet process with external gelation. The particles are compacted into pellets and then sintered. Fuels with lower plutonium content can be fabricated by mixing the plutonium-bearing particles with depleted urania powder before forming them into pellets.

The fuel element fabrication sequence differs in no way from fabrication based on sintered powders. The work has been carried out onthe laboratory scale; at this stage the fabrication process is not continuous.

This concept "COGEPEL" - Process will be perfected within the framework of Belgium's fuel fabrication programme. Tests on fuels are to be carried out in Belgium in the BR2 test reactor. The possibilities for full-scale testing in fast reactors are limited on account of high costs, safety regulations and lack of space.

## 4.2 FRANCE

Between 1968 and 1979 France carried out considerable research inthe field of spherical particle fuels for HTGRs, together with some exploratory work on the applicability to gas-cooled fast reactors. This research was at first part of the French contribution to the Dragon Project. The Dragon fuel fabricated in France by the companies SFEC (kernels) and CERCA (coated particles and compacts) was irradiated in the form of particles, compacts and fuel elements in the French reactors EL0E, OSIRIS, PEGASE and also in the Dragon reactor. As part of the collaboration with General Atomics from 1973 onwards, research was done on the high-enriched uranium-thorium fuel cycle. This work dealt with the fabrication of fissile and fertile coated particles, the manufacturing of cylindrical compacts and irradiation testing. Finally fabrication and irradiation tests were made for a U-Th cycle fuel, made up of fissile particles with uraniumoxycarbide kernels, obtained by calcination of uranium-impregnated ion-exchange resin spheres and TRISO coatings, as well as fertile particles with $ThO_2$-kernels and BISO coatings. The overall experience gained in France in the field of coated particle fuels for HTGRs has proved positive in all respects. The decision to discontinue the French work on high temperature reactors in 1979 had nothing at all to do with the technical aspects of particle fuels.

Spherical particles of oxide fuel for fast reactor applications have been manufactured by dry agglomeration of powders and by a wet process developed by General Atomic. Experimental fuel elements have been made by vibro-compaction of spherical particles obtained by the dry technique. Irradiation of these pins in a thermal reactor at high linear

power has produced considerable fusion of the fuel and this was the reason to drop the idea of vibro-compacted elements.

## 4.3 F. R. G.

Work on particle fuels has been concentrated upon the fabrication of fuel elemtents for HTRs with HEU/Th cycle and MEU/Th cycle. It was directed at the fabrication of fuel elements for AVR (Julich HTR test reactor) and THTR and at the development of fuel elements for advanced HTRs, i.e. for direct cycle plant and process heat plant as well as for block fuel elements according to GAC design. Efforts also included irradiation and PIE of fuel under development programmes for THTR (completed in 1972), for reactors fuelled with block fuel elements (programme discontinued in 1977), for direct cycle plant and process heat plant (completed in 1980).

AVR and THTR fuel elements were tested according to requirements and approved. The development of HEU/Th fuel elements for direct cycle plant and process heat plant is almost completed. The elements display a positive irradiation benaviour.

Current activities on particle fuels relate to all of the three main types of reactors. THTR fuel elements are fabricated on an industrial scale. $(U,Pu)O_2$ fuel is produced and tested for application in FBRs and LWRs with thermal Pu recycle. $(U,Th)O_2$ is investigated in view of utilization in LWRs with thorium cycle, and carbide fuel is developed for advanced breeder reactors.

The reference fabrication process of spherical particles is the gel precipitation process with external gelation. The main stages of HTR-fuel fabrication are coating of spherical particles, mixing of coated particles with graphite powder containing a binder, compressing into spheres, coking and calcining. LWR and FBR fuel elements are produced by compressing of uncoated particles into pellets, sintering of pellets and assembling of fuel rods and fuel elements in the conventional way. The quality control of the fuel is pursued in compliance with the respective specifications for HTR (mainly THTR), FBR (mainly KNK II or SNR-300) and LWR. Irradiation testing is executed under largely realistic conditions, i.e. according to the irradiation history to be expected in the power reactor.

Technical problems of LWR/FBR particle based fuel to be solved are:

- upgrading to production scale
- continuous fabrication process
- demonstration of the operating behaviour in material test reactors prior to use in operating reactors
- changing to remote-controled automatic plant operation

Future plans concerning the HTR line deal with the use of low enriched fuel in the AVR and with the development and fabrication of LEU fuel elements for a THTR 300 follow-up project.

## 4.4 ITALY

Two wet-route processes have been developed in Italy in the early 1960s with the aim to produce HTR coated particles and vibrocompacted LWR/FBR fuel. The process developed at CNEN has a typical "Sol-Gel" approach, using aliphatic amines and catalytic reduction to obtain the gel, whereas the process developed at Agip Nucleare is a "Gel Supported Precipitation Process" (GSP process), characterized by organic polymer gelation together with the precipitation of heavy metal compounds in an aqueous medium. The GSP-process proved to be the preferable route and further development resulted in a continuous production technique, which has been successfully established on pilot plant scale for the production of uranium-, thorium-and plutonium oxide microspheres.

Urania- thoria microspheres have been manufactured at a 10 kg/d pilot plant of Agip Nucleare and a plant of slightly higher capacity had been built at the BNFL Springfield Works. The irradiation experience on GSP particles in the HTR fuel field was gained by the collaboration with Dragon Project, BNFL and BN. There was no difference in irradiation behaviour between GSP particles and particles manufactured by the Dragon powder agglomeration route. Kilogram scale batches of urania-plutonia particles have been produced on a pilot plant at the CNEN Plutonium Laboratory for irradiation tests of vibrocompacted fuel pins. Several irradiation tests have been performed on spherepac fuel pins under thermal and fast reactor conditions. The main facts observed were:

good dimensional stability of the spherepac stack, fuel relocation and structure modifications comparable to reference

samples of pelletized fuel, chemical attack of the cladding due to moisture and impurities in some samples.

Despite the high potential of the vibrocompacting technique for the fabrication of fast reactor fuel, at present Italian view is that the requirements of future plutonium fuel fabrication plant may be more easily achieved by a combination of gel-supported precipitation conversion and pellet preparation by cold pressing and sintering.

CNEN and Agip Nucleare are carrying out a RD programme based on:

- optimisation of the gel supported precipitation process to produce granules of urania and urania-plutonia as feed for pellet pressing
- installation and operation of a pilot demonstration unit
- production of representative batches of $(U,Pu)O_2$ pellets
- irradiation experiments.

In addition a research programme is presently pursued on the preparation of (U,Pu)C particles.

## 4.5 THE NETHERLANDS

The very first sol-gel process for nuclear fuel was developed by KEMA (Keuringsinstituut voor Elektrotechnische Materialen, Arnhem) between 1957 and 1959. It was based on the dispersion of an aqueous colloidal sol in an organic liquid and external gelation of the droplets by ammonia dissolved in the organic. Attempts to increase the diameters of urania/thoria spheres above 400 $\mu$m fuel size led to a new KEMA process of internal gelation in 1964. Ammonia was added to the sol in a masked form and set free by heating the droplet, thereby causing gelation. The production of pure $UO_2$ kernels was enabled in 1968 by the development of the KEMA U (VI) process. With this process support was given to several foreign projects in Europe, and more recently at ORNL.

In 1969 a joint research programme was started to demonstrate the feasibility of the fabrication of urania spherical particles and to investigate the performance of these vibratory-compacted spheres, filled into Zircaloy tubings, during irradiation under LWR conditions. Process development at pilot plant scale was carried out by Interfuel. Three

particle sizes 1, 0.1 and 0.01 mm have been used. Irradiation programmes have been carried out in the HFR Petten, in the Halden Boiling Water Reactor and in the Dodewaard BWR.

The first irradiation experiment in the HFR comprised 3 fuel pin bundles, each bundle consisting of 3 Zircaloy clad pins, one pin filled with vipac-sharp edged $UO_2$ particles and two pins filled with sphere-pac $UO_2$[31]. The second fuel performance experiment in the HFR also comprised three fuel pin bundles under PWR conditions. The fuel types involved were $UO_2$ sphere-pac, $(U,Pu)O_2$ sphere-pac and $UO_2$ pellets[22].

In the Halden BWR four sphere-pac fuel pins and three pellet fuel pins are under irradiation. The pins are partly pressurized and partly equipped with fuel and cladding elongation detectors. Four and seven sphere-pac fuel pins resp. have been irradiated in two previous runs. The full size prototype fuel elements, each with 35 fuel pins, 2m long, have been irradiated in the Dodewaard Reactor. The sphere-pac fuel behaviour under power reactor operation conditions will be examined and compared with that of the pellet fuel irradiated under identical conditions.

In 1981 a revised development programme started aiming to optimize the process as such minimizing the chemical consumption and waste streams and optimizing process control. Optimization will be supported by fundamental studies on process mechanisms. Further it is planned to construct a fuel fabrication demonstration facility for $UO_2$ fuel.

Current activities deal with the production of oxide particles by a sol-gel process with internal gelation. Particles of 1, 0.1 and 0.01mm diam. are mixed and compacted in Zircaloy fuel pins by vibration. The quality of the sol-gel kernels is controlled in all stages of fabrication including chemical analyses, determination of crystallite size, pore size, pore size distribution, diameter, density, U/O-ratio, impurities, homogeneity, moisture content, grain size, particle size and distribution, stack density and stack homogeneity.

Irradiations of this fuel specified for LWRs is effected under LWR conditions. Pool examination of the elements as well as PIE are performed, hotcell examination will start in the near future.

Future plans include fundamental studies on the sol-gel process, the production of a number of fuel pins and fuel elements for test irradiation including PIE, and the optimization of a new pilot plant.

## 4.6 SWEDEN

Besides some RD fabrication experience in the early 1960s, there exists extensive experience on irradiation testing, including PIE since 1961 up to the present. This work consisted mainly of contributions to Dragon development programme and to work in support of the FRG programme. Fission product release during operation as a function of both fabrication and operation parameters has been the main object of the studies. Currently various types of oxide and carbide particle fuels are irradiated and examined on behalf of commercial partners.

## 4.7 SWITZERLAND

Work has been going on at EIR Wurenlingen since 1967 on the development of sphere-pac fuel produced by a wet(gelation) process. It covers the fabrication of uranium-plutonium carbide fuels (max.1/2 kg per week), the fabrication of test fuel pins, the carrying out irradiation tests in the EIR SAPHIR reactor and foreign reactors, PIE and evaluation of the performance of the pins in the EIR hot cells. The initially used Oak Ridge sol-gel process was deplaced by an internal gelation process which had been modified to include plutonium and carbon-containing feeds. Some work has also been done on $UO_2$ and $(U,Pu)O_2$ as well as on the production of pellets using gel produced feed particles. There exists an alpha-box line, containing a considerable amount of analytic equipment. Facilities for fabricating Pu containing test fuel pins are being enlarged to accept pins 2,5m long.

Production of 2-3 pins per month with full quality assurance is possible.

The fabrication of (U,Pu)C sphere-pac has been demonstrated on a lab-scale with good control over fuel characteristics. Approcimate 20 test fuel pins have been fabricated all without major difficulties. The irradiation tests indicated that these fuels perform at least as well as pellet fuels under the same conditions and in some respect (fuel clad mechanical interaction) better. Interest in sphere-pac carbide fuel

would also be based on the fabrication advantages. The pyrophoric dust problem is largely eliminated, the fabrication is simpler and there is great potential for remote fabrication, increasing security and improving operator safety, which seems to be particularly important for fuels containing recycled plutonium.

Current work deals with the fabrication of a batch of 20 fuel pins for a fast flux comparison with pellets. (U,Pu)C particles of two size fractions(700$\mu$m, 40-80$\mu$m dia) ard produced according to EIR modified internal gelation process. Vibrofilling in stainless steel claddings is carried out using tne infiltration technique. Smeared densities range between 77 and 80% of theoretical depending mainly on particle density. Tne pin fabrication is extensively controlled by numerous quality tests. Irradiated sphere-pac pins are examined in the EIR hot cells. The operation may include: axial -scanning, equipment dismantling, measurement of flux monitors, pressure test, pin dimensional checks and visual examination, leak testing, gas puncturing, sample cutting, metallographic examination, autoradiography,, micro-drilling and chemical analyses, burn-up analyses, clad microhardness, radial -scanning (fission product distribution), gross porosity redistribution, special examination of the cladding using SIMS. Other analyses carried out at other institutes have been clad carbon profiles, microprobe analyses of fission products, Pu redistribution and pin profilometry. As already mentioned, the production is at lab scale. There exists no commercial fuel facility in Switzerland. EIR has several formal and informal collaborations with fuel developers in other countries. No industrial plans exist in Switzerland to produce sphere-pac fuels, but EIR is interested in contributing to commercial developments, which are under way in several countries.

For EIR the current plans are to continue with carbide process development and fabrication of fuel for irradiation testing at the present level for the next future. Development of a sphere-pac pin behavioural model will be continued. A continuing irradiation programme would include a mixture of detailed parameter studies and larger scale bundle tests provided partners for the latter can be found.

## 4.8 UNITED KINGDOM

Work on HTR concepts commenced at UKAEA, Harwell, in the early 1950s and led to research into helium cooled concepts based on coated fuel particles in many countries and by the OECD at Winfrith, England. Simultaneously with work on HTR part of the early UK work on particle fuel was devoted to a $CO_2$ cooled VHTR concept in which coated particles were carried in fuel pins formed of dense self-bonded SiC. Eventually it was decided to drop this development in order to concentrate upon the helium cooled concepts. Considerable effort on the development of particle fuel concepts followed the successful operation of the Dragon Reactor. In the late 1960s materials research was greatly increased and coupled with designs investigation to support the construction of a 300 $MW_{(e)}$ prototype. The developments were directed towards the introduction of a Low Enriched Uranium Cycle compatible with existing UK enrichment capabilities and towards the improvement of the manufacturing characteristics. Different types of fuel elements were considered with the aim to reduce the high fabrication costs of the original Dragon prismatic fuel element design. The manufacturing development, basic research, test irradiations, PIE was finished in 1975 when it was decided to concentrate resources on AGR and FBR.

The UK development work on the fabrication of FBR fuel has been devoted primarily to particle fuel since 1971, as it was considered that a wet chemical route, avoiding the filtration of fine powder precipitates, offers the best potential for remote operation and hence reduced operator dosage in the fabrication of plutonium fuels. Development of the preferred gel precipitation process has involved basic chemical flowsheet studies and chemical engineering development, using inactive pilot plants with urania and urania/thoria. There is also a laboratory scale facility for the manufacture $(U,Pu)O_2$ spheres in batches of 700 g. The objective of this work has been the production of large (300 $\mu$m) and small (30 $\mu$ m) uranium/plutonium oxide spheres for vibrofilling fuel pins by low-energy infiltration technique, which has been achieved on the laboratory scale.

About 200 kg of $(U,Pu)O_2$ microspheres have been produced with densities, greater than 96% of theoretical, which are free of cracks. Using this material, over a hundred fuel pins have been made for irradiation in the Prototype Fast Reactor with a nominal smear density of

80 per cent. The normal (pellet fuel) tolerances for weight of fuel in the pins and smear density were readily met. The first gel vipak fuel pins were loaded into PFR in 1980, the number is expected to build up to about 300 during 1981, and the lead pins are scheduled to achieve the target burn-ups of 10% about the end of 1981.

The work to date has been carried out on a batch process at the laboratory scale. A pilot plant operating a continuous process with a throughput of 1 t/yr has been built and is being commissioned with uranium. Production of mixed oxide spheres is due to commence in mid 1981. Current irradiation programme will be followed by an endorsement programme at the thousand pin scale on material fabricated in the gel pilot plant.

The gel vipak fuel concept is being developed in the UK as an alternative to conventional pellet fuel for use in the FBR. The present reference fuel design for CDFR is based on annular pellet fuel, but this will be reviewed in the light of experience with the Gel Pilot Plant, the result of the irradiation programme in PFR and reprocessing studies.

The main problems to be resolved in the future are to demonstrate the viability of the process on the pilot plant scale and the ability of the fuel to achieve irradiation conditions which are economically attractive when the whole fuel cycle is considered.

Future plans imply the development of a route for the direct pressing of gel precipitated spheres into pellets. Compared with vibrocompacting, this route would have more limited advantages in respect to fabrication, but the irradiation performance of pellet fuel is already well established.

4.9 U.S.A.

The sol-gel technology for the production of particle fuel was vigorously pursued in the U.S. until 1972.

At that time the fast breder reactor programme concentrated on pellet fuel and government support for particulate fuel for FBRs was terminated. Sol-gel development was continued in the field of high-temperature gas-cooled reactors. Extensive progress was made

regarding sphere formation processes, control of sphere size and sphere handling, transport and inspection. The renewed U.S. interest in gel-sphere-pac in 1977 came from two directions: the concern for nonproliferation-type flow-sheets, which pointed towards full remote fabrication and the desire for improved fuel-cladding behaviour to allow more severe thermal ramping during reactor operation.

The sol-gel process with internal gelation has been used to produce fuel particles. About 23 pins with $(U,Pu)O_2$ fuels were irradiated in EBR-II. The performance of particle fuel was found to be directly comparable to pellet fuels with the same smear density.

Current activities refer to HTGR, LWR and FBR. The processing of spherical particles is an ongoing programme for HTGR, covering replacement cores for the 300 MWe Fort Saint Vrain Reactor and fuel rods for advanced developments. ORNL is producing microspheres for sphere pac, fuel pins. Work at Exxon Corporation is on a pilot plant scale for LWR. A hybrid fuel pellet fabrication concept, in which dried sol-gel microspheres are pressed into pellets and sintered, is being developed for both LWR and FBR, in addition to the sphere-pac technique.

The irradiation testing of spherical particle fuels includes a wide range of irradiation conditions depending upon the type of reactor considered. The PIE of the fuels is a continuing activity at several centers where hot-cell facilities are available (ORNL, HEDL, ANL, GA, Idaho Nat.Eng.Lab.) Future plans: to continue the present programmes for several years until the relative advantages and disadvantages of spherical particle fuel are determined more clearly.

The scientific and technical problems to be resolved include in U.S. view:

- for HTGR coated particle fuel: the development of more resistant coating and more retentive fuel particles, the threshold limits for temperature, time and burn up

- for the LWR: improvement of fabrication technique and quality, control of sphere-pac fuel; development and testing of spherical process for the fabrication of fuel pellets from gel microspheres;

- development of carbide fuel spherical particles for the FRR: development and testing of mixed oxide and mixed carbide fuel particles; irradiation of spherepac fuels in a fast reactor core with alternate cladding alloys; problems of cladding failure on behaviour of spherepac fuel; to consider hybrid fuel concept.

4.10 USSR

In the USSR the research work on fabrication, test, irradiation and post-irradiation examination of fuel based upon spherical particles are carried out to improve fabrication technology and substantiate efficiency of such fuel in HTGR as well as in FBR reactors concepts. In the All-Union Institute of Nonorganic Materials considerable amount of work has been done in physical and chemical investigation, technology and instrumentation development for the process of oxide microspheres fuel production using both dry and wet techniques. The research work fulfilled covers the process of vibration compaction securing required density and uniformity of fuel distribution in FBR concept.

At the present time the research work on laboratory scale is basically being carried out with nonenriched oxide fuel with internal gelation to improve fabrication technology of fuel for HTGHR and FBR reactors.

5. ECONOMIC AND SAFETY STUDIES

In most of the countries no detailed economic and safety studies on fuel, based upon spherical particles, have been carried out up to now.

A cost evaluation, comparing the powder agglomeration route with the gel supported precipitation process (GSP) has been elaborated by Agip Nucleare in 1972. Difference in costs for raw materials, utilities and equipment mean that for small scale production (2t U/yr) the costs of the two processes are very similar, but for larger scale production (100 t U/yr) the GSP process should be cheaper about 20%.

In 1974 a detailed calculation on GSP spherepac fuel fabrication process has been made by Batelle Pacific Northwest Lab. Fabrication costs were calculated for twelve cases. Three different processes -

spherepac, vipac of sharp edged particles and pellet were examined. Each process was calculated for PWR and BWR fuel element manufacture with both uranium and plutonium enrichment.

The spherepac process is cost competitive with the pellet process. The use of plutonium enrichment is a cost advantage for the spherepac process since an expensive nitrate to oxide conversion step is not required. The relatively high fixed cost components of the spherepac process indicate that it may enjoy a high economic gain in scaling up to larger plants.

There are extensive studies in the U.S.A. on the economies and safety aspects of fuels based upon spherical particles e.g. at GAC, Exxon Corp., GE, Westinghouse, ORNL and BNWL. Fabrication cost estimates in UK for spherepac fuel, show that the cost is comparable to that of pellet fuel.

In general the basis elements, required for thorough studies on economies and safety aspects, will have to be furnished by the research work at present under way. But there is a broad consensus on the advantages of the spherepac and the hybrid process over the standard pellet route which are particularly obvious for fuels containing plutonium:

1) Only liquids of free-flowing spheres are to be handled
2) The direct transformation of nitrate solutions into mixed oxides
3) The dust-free operations involved
   Handling of powders results in dust contaminaitions which produce large amounts of plutonium-containing waste as well as considerable radiation exposure of the personal during both fabrication and maintenance operations
4) Mechanical finishing is eliminated on the spherepac route. Grinding of hybrid pellets can be avoided or is minimized by sintering to size. Less scrap.
5) The versatility of the process in being able to handle U, Th, Pu and mixtures thereof. By slight variations in process handling both oxides and carbides can be produced with the same equipment.
6) Lower sintering temperatures, because the small crystallites, produced in the gels, sinter more easily to a high density.

7) Genuine mixed oxides $(U,Pu)O_2$ are formed by coprecipitation from one solution. No disturbances during reprocessing, owing to $PuO_2$ enriched grains which are insoluble in $HNO_3$.

8) Performance benefits: fuel-cladding interactions are low; no hot spots due to plutonium homogeneity.

9) The absence of fuel powders and dust improves considerably the security of fabrication. In the case of an important accident the fuel powder and the fuel dust would be the main contributors to contamination of the operating rooms and environment.

## 6. REMOTE FABRICATION AND AUTOMATIC HANDLING

When plutonium of LWR origin is used for fuel fabrication experience has shown that full mechanization of the production line including remote operation is necessary together with extensive use of gamma and neutron shielding. Fabrication of microspheres on the wet route offers an excellent potential for the remote, automated fabrication of fuels:

- the wet process is simpler, requiring fewer steps for fuel fabrication than powder processes
- gel granules are inherently free flowing; preslugging and granulation which are necessary for powder to produce free-flowing feed material for automatic pellet presses may be eliminated
- powder milling, producing dust and introducing impurities into the fuel material is unnecessary
- pellet grinding of hybrid fuel may also be eliminated by sintering to size
- microsphere processes are especially suited for fuel refabrication, since granules are produced directly from nitrate solutions. The free flowing granules are suitable for pneumatic transport
- since the gel-granules are relatively dust free, the spread and build-up of contamination would be minimized. This would reduce both personal radiation exposure and equipment decontamination time.

Automation and remote operation are indeed the final goals of most of the present activities. There exists full conformity of opinions

that fuel fabrication on particle route can easily automated and remotely controlled - more easily in any case than the powder to pellet route. In Germany studies on remote control and automatic handling in plants, producing spherical particles, were conducted until 1976 for refabricating fuel for HTR with HEU/Th cycle. The present knowledge indicates that basic problems will not arise. Remote fabrication of 1 mm spheres of $(U/Pu)O_2$ has been demonstrated at lab scale within the Dutch programme.

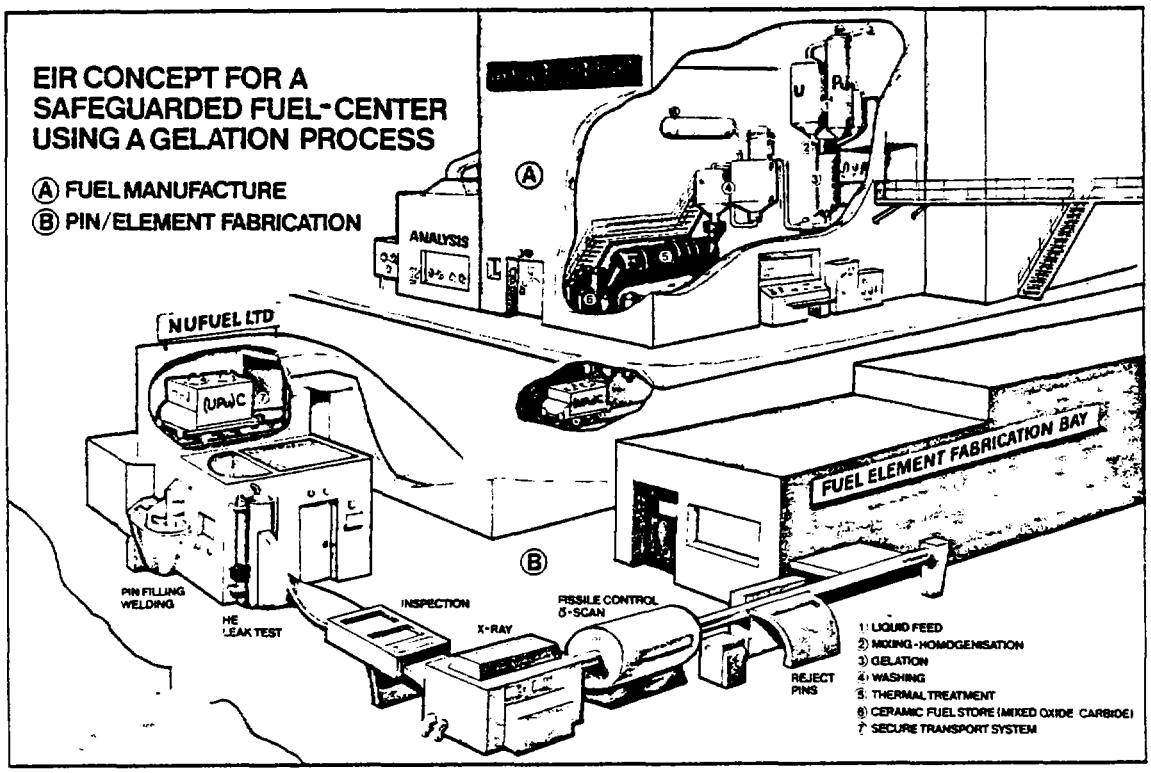


Fig. 11.

## 7. CONTRIBUTION TO SAFEGUARDS ISSUES

Compared with pellet fabrication the particle route is not only safer to the operator and the environment but it is also more proliferation and diversion resistant. The feed preparation flowsheet is compatible with co-stripped uranium and plutonium streams from the reprocessing plants, thus leading logically to an integrated fuel reprocessing and fabrication plant (Fig. 11). There is no need to handle pure plutonium in the wet particle route, for the particles are produced by coprecipitation of a mixed uranium-plutonium nitrate solution which could be obtained directly from a reprocessing plant. As a result of the wet process simplicity the holdup of plutonium in process equipment is

greatly reduced. The wet route would also be of special interest for the handling of strongly emitting multicycled plutonium and of spiked, i.e. intentionally contaminated fissile material which could be used in non-proliferation type fuel cycles and would require fully remote refabrication.

Co-conversion by sol-gel technique as a method to reduce the presence of plutonium in the fuel cycle in separated form has been analyzed in INFCE Working Group 4, document.

## 8. SCALE OF THE WORK

The actual fabrication scale of fuels based upon spherical particles is very different. Fuel for high temperature gas cooled reactors is fabricated and utilised on an industrial scale, but at present only in the U.S.A. and in the FRG. Pilot plant scale has been reached in some countries in the production of spherepac fuel and hybrid fuel (particles, produced by a wet process, are pressed to pellets) for light water reactors. FBR fuel, both spherepac and hybrid, is produced on laboratory scale, but pilot plant stage will be achieved soon.

In UK a pilot plant, operating a continuous process with a planned throughput of 1 t/yr, has been built. Production of mixed oxide spheres is due to commence when commissioning of the plant will be finished. It must be pointed out that the dimensions of plants are limited by criticality considerations and for that reason some engineering components, will be at prototype scale soon. On an industrial scale more of such relatively small producion units in parallel are to be used.

## 9. PROSPECTS FOR FURTHER TEST APPLICATIONS IN OPERATING REACTORS

In the field of spherepac fuel it is planned:

- to test 20 EIR-fabricated pins of the mixed carbide type in an U.S. fast test facility
- to test special subassemblies in an LWR (Big Rock Point Reactor)
- during 1981 the irradiation programme in Prototype Fast Reactor (PFR) will achieve the hundred pin scale on material made in laboratory equipment. This will be followed by an endorsement

programme at the thousand pin scale on material fabricated in the UK gel pilot plant.

- to continue the Dutch irradiation programme in the HFR, in the Halden Reactor and in the Dodewaard Reactor.

Hybrid fuel, produced by compressing and sintering of microspheres to pellets:

- will be tested in German KNK II (FBR-fuel) and in KWO (Obrigheim power plant, LWR-fuel)
- will be irradiated in Belgium in the BR 2 test reactor. Possibilities for full-scale testing of Belgian fuel in fast reactors are limited by the fact that Belgium itself has no fast reactor and an access to fast reactors abroad is not easy.

## 10. SCIENTIFIC AND TECHNICAL PROBLEMS

Among the scientific and technical problems to be resolved are some which can be considered as being valid to all countries involved in the development of fuels, based upon spherical particles. These objectives include

- upgrading to production scale
- transition from batchwise to continuous operation
- changing to remote-control automatic plant operation
- fabrication of sufficient well characterized fuel for large scale irradiation testing, leading to licensing
- demonstration of the operating behaviour in material test reactors, prior to use in operating reactors
- demonstration of the fuel's ability to achieve irradiation conditions which are economically attractive when the whole fuel cycle is considered

There are also more individual problems, e.g.:

- optimization of the gelation process
- development of a method to produce "fines" with a more narrow size spectrum
- improvement of quality control techniques, applicable to the fines

- recycling of chemicals and decreasing waste quantities. Development of waste calcination processes
- increasing spherepac rod density
- influence of fabrication parameters on the deformability of the particles during pellet formation to avoid excessive roughness, surface irregularities and internal cracks
- irradiation of spherepac fuels in a fast reactor core with alternate cladding alloys
- study of reprocessing spherepac fuel.

## 11. POTENTIAL USE WITHIN THE NATIONAL NUCLEAR PROGRAMMES

BELGIUM: The COGEPEL process will be perfected within the framework of Belgium's fuel fabrication programme. Certain irradiation tests are to be carried out in Belgium, but others will have to be performed abroad.

FRANCE: No development is planned for the near future within the national nuclear programme.

FED.REP.GERMANY: as far as LWR and FBR are concerned possible applications emerge with regard to plutonium recycle; this also applies to LWR with thorium cycle in cooperation with Brazil.

ITALY: Starting from 1966, CNEN and Agip Nucleare decided to work jointly on the application of GSP route for the production of nuclear fuel in particular for the fabrication of plutonium-uranium mixed oxid-fuel. Therefore the activities in this field are carried out in the framework of a research and development programme carried out in common by these organizations.

NETHERLANDS: Further experimental fuel elements will be produced, irradiated and examined after irradiation. Potential for further use depends on the National Programme which is as yet undecided.

SWITZERLAND: EIR has several formal and informal collaborations with fuel developers in other countries. Currently the main objective is to contribute to advanced fuel development, to maintain contacts with newest fuel cycle and fabrication technologies and advanced reactor programmes. An important feature of EIR's facility is the ability to handle plutonium containing fuels.

UNITED KINGDOM: The gel vipak fuel concept is being developed in the UK as an alternative to conventional pellet fuel for use in the FBR. The present reference fuel design for CDFR is based on annular pellet fuel but this will be reviewed in the light of experience with the Gel Pilot Plant, the results of the irradiation programme in PFR and reprocessing studies.

USA: The potential use mainly will depend upon the relative performance and cost of the demonstraton subassemblies in LWR's and FBR's. Continuing research, development and production for HTGR.

USSR: Respective for utilization of particle fuels depends on the result of research work and economic studies.

## 12. INDUSTRIAL POSSIBILITIES

To provide improved FBR fuel fabrication processes and to assure meeting increasingly stringent health and safety criteria an advanced automated fabrication system must be developed. There exists common conviction that the gel particle processes can meet the requirements for an advanced plutonium fuel fabrication line.

The potential of these processes relates to the fact that for the complete route from nitrate solution to pin filling or compressing to pellets the plutonium can be transported in liquid or as hard free flowing granules, so that all operations are conducive to the application of remote handling and automation. The wet fabrication route is especially appropriate to refabricate multicycled or spiked fuel. In some countries commercial developments are already underway. The further expansion of these industrially implemented programmes will depend upon the relative performance and economics of the sphere-pac process and the "hybrid pellet" process during the next few years.

## 13. RECOMMENDATIONS ON INTERNATIONAL CO - OPERATIONS

Most of the countries which are engaged in the development of fuels based upon spherical particles plead for more international cooperation in this field. There have been made even suggestions of special topics, e.g.

- pooling of all irradiation test results
- programme to investigate behaviour of sphere-pac fuel beds
- international development of a sphere-pac pin behavioural code
- international study to produce a realistic concept of a reprocessing and refabrication center using gel technologies. Problems of access to commercial information would need to be resolved since it would be necessary to base the analysis on a concrete plant concept.
- IAEA should contribute to enlarging the opportunities for full-scale testing of such fuels. On the other side CEA of France is offering interested countries the use of existing irradiation facilities (capsules and loops in the Siloe and Osiris Reactors) and Studsvik is interested on a commercial basis in any international coopperation that can be combined with its experimental facilities.
- in context with the desire for more international cooperation it has been pointed out that developments are costly and have considerable commercial significance which would need to be recognized and properly dealt within any arrangements made.

More cooperation in this field may be accomplished most effectively by arranging an international conference, preferably under the auspices of the IAEA. This international conference should be held in due time to clarify the potential and the present status of fuel based upon spherical particles.

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APPENDIX A

# World Survey on Experience With Particle Fuels in different Reactor Concepts

## Questionnaire and List of Responding Countries

The following information was sent to 14 IAEA Member States in June 1980. The purpose of the survey is explained first, then follows the questionnaire.

### Introduction

To date particle fuel is only used in high temperature reactors (HTR). In this reactor type the particles exist of oxide fuel with a diameter of about 0.5 mm and are surrounded by various coatings in order to safely enclose fission products and decrease the radioactive release into the primary circuit. These spherical coated particles with a total diameter of less than 1mm are the basis for the graphite fuel elements in HTRs.

However, it is felt that particle fuels could have advantages compared with pellets both on fabrication and in-core behaviour in several reactor concepts. This fuel is now of general interest and there is a high level of research and development activity in some countries.

Particle fuel can be fabricated by dry and wet chemical processes. Using the dry process, the fused or sintered materials are usually crushed into the particles or suitable powder is agglomerated and then sintered.

There are numerous wet processes, all of them using solutions or sols of fissile or fertile materials which are dispersed into uniform droplets. The spherical shape of these droplets is established by gelation which can be achieved by either a precipitation or dehydration reaction. After washing the gel spheres they are subsequently dried and sintered.

A potential attractive alternative to the use of pellets as metal-clad fuel for nuclear reactors is the sphere-pac process. It consists of the production of dense ceramic spheres of the desired fissile and/or fertile materials which are loaded into metal tubes to form the fuel rods. This process and operating equipment offers the advantage of being comparatively simple with lower radiological or exposure risks. Its basic simplicity may make it more attractive for the remote fabrication of advanced or proliferation-resistant fuels. There is also some evidence that sphere-pac fuel could have some performance advantages over traditional fuels.

A hybrid fuel pellet fabrication concept in which microspheres are pressed into pellets and sintered is also being developed. This concept may be attractive for some fabrication concepts as well as having some in-core behaviour advantages.

Because of the necessity to improve economics, safety and environmental protection, to increase the proliferation resistance in all steps of fuel production and utilization research and development work on fuel based upon spherical particles has been carried out in the last years. In order to collect, organize additional information and summarize experience by Member States on fuel based upon spherical particles, the attached questionnaire has been prepared.

The questionnaire is structured to cover the following items:

- Assessment of different applications of spherical fuel particles relevant to national programmes;
- Prospects for further test application in operating reactors;
- Potential contributions to some fuel cycle safeguards issues;
- Economic considerations;
- Industrial implementation possibilities;
- Summary of current activites and future plans.

The questionnaire does not intend to request information which is either sensitive or proprietory. Any information supplied will be assumed to be publishable in a summary report unless otherwise indicated.

QUESTIONNAIRE

1. Indicate work completed within your country in the field of fabrication, testing, irradiation and post-irradiation examination of spherical particles or fuel elements based upon spherical particles.

2. Summarize the experience made in your country within the last years.

3. What are the current activities (as per following scheme)?

   3.1 Chemical type of fuel (oxide, carbide, others).

   3.2 Reactor type.

   3.3 Enrichment of fuel.

   3.4 Fabrication process of spherical particles (dry process, sol-gel process with internal or external gelation, gel precipitation process, others).

   3.5 Coating of particles.

   3.6 Processing of spherical particles to fuel elements.

   3.7 Quality control testing of fuels.

   3.8 Irradiation conditions (thermal, epithermal, fast flux, irradiation time, temperature, burn-up, linear ratings).

   3.9 Post-irradiation examination of fuels.

4. Has work been performed at laboratory scale, at pilot plant scale, or an industrial scale? Is the fabrication process a continuous one?

5. Estimate of the potential use of the development of fuels based upon spherical particles within the national nuclear programme.

6. Any economic and safety studies on fuels based upon spherical particles?

7. Estimation of possibilities of remote fabrication and automatic handling of particle fuels?

8. Potential contribution of utilization of fuels based upon spherical particles to some fuel cycle safeguards issues?

9. Prospects for further test application in operating reactors?

10. Industrial possibilities?

11. What scientific and technical problems to be resolved can be mentioned?

12. What are future plans?

13. Any recommendations on international co-operation in this field?

List of Responding Countries

Belgium

France

Germany, Federal Republic of

Italy

Netherlands

Sweden

Switzerland

United Kingdom of Great Britan and Northern Ireland

United States of America

Union of Soviet Socialist Republics

## APPENDIX 3

### Consultant Group on World Experience
### With Particle Fuels in Different Reactor Concepts

| | |
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