

## Reactor Boiler and Auxiliaries - Course 133

## PREPARATION AND MANUFACTURE

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The production of uranium, in the form and purity needed for fuel manufacture, entails a series of carefully designed refining operations. In addition to the pure metal, alloys and several chemical compounds of uranium are produced as basic fuel materials. The bulk of the work done in Canada in this line concerns natural uranium products, but enriched and depleted materials are also being processed in increasing quantities.

Uranium concentrate from the mines is shipped to the refinery as "yellowcake", a precipitate of di-uranates, and this is the starting material for all the refinery processes.

The impurities in yellowcake vary according to the source of the material, so that the preliminary refining operation is the blending of batches of yellowcake from several sources. Although the total concentration of impurities is unaffected by the blending process, it has the advantage of providing a means of control of individual impurities.

Chemical operations commence with the digestion of the yellowcake with nitric acid to produce an acid slurry of uranyl nitrate. This is purified in a series of extraction columns, where the slurry is contacted with an organic solvent which dissolves the uranyl nitrate, leaving most of the impurities in the aqueous phase. The organic solution is scrubbed with a small amount of water which removes most of the remaining impurities, and is then contacted with large amounts of pure water, causing the uranyl nitrate to transfer back to the aqueous phase and leaving impurities in the organic solvent. The final aqueous solution of uranyl nitrate has a purity which meets the high standard required for nuclear fuel production, and after concentration in a two-stage evaporator, contains about 1400 grams of uranium per litre.

Having produced a uranium compound of the required purity, it remains to convert this compound into a derivative which is suitable for nuclear fuel manufacture. In one series of operations, the uranyl nitrate is heated to give uranium trioxide by thermal decomposition. The trioxide, also known as "orange oxide", is cast into pellets which are treated by a moving bed process in two stages. The pellets are heated, and pass down a reaction vessel where they are met by a stream of hydrogen, causing reduction to the dioxide (brown oxide). In a second heated vessel, the pellets are changed, by a stream of hydrogen fluoride, into uranium tetrafluoride or "green salt". Excess

acid is removed by a scrubber and the green salt is collected on filters.

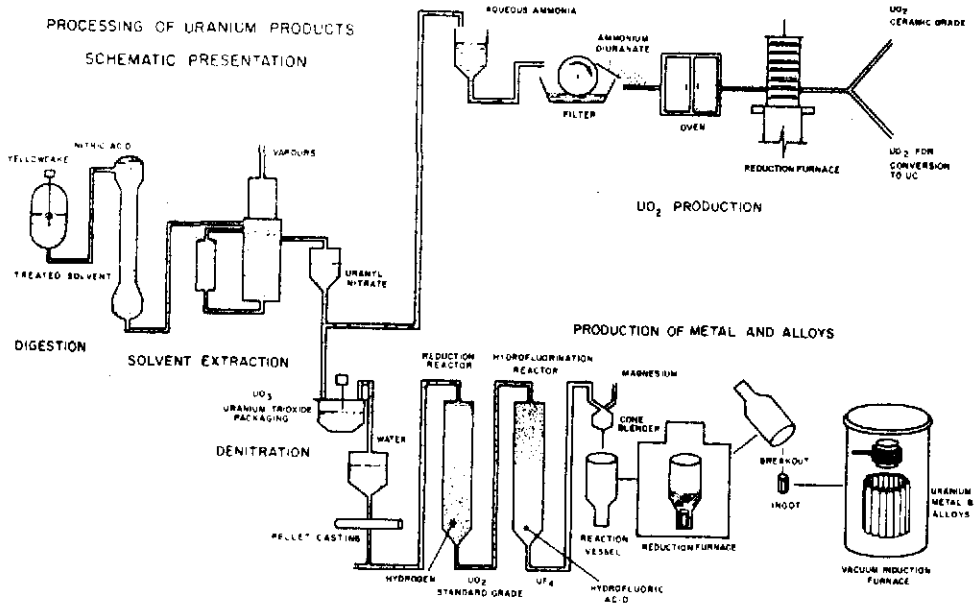


Fig. 1

### Uranium Metal

Uranium metal is made from the tetrafluoride by a thermite-like process in which the salt is reduced by magnesium metal. The reaction takes place in a vessel with a refractory lining which is made from magnesium fluoride - a by-product slag recovered from previous production. Weighed quantities of powdered uranium tetrafluoride and granulated magnesium are blended and packed uniformly in the reaction vessel which is then capped with refractory slag. The vessel is placed in a furnace and heated slowly over a 12 - 16 hour period to 650°C; the thermite reaction starts spontaneously and is complete in two or three minutes during which time the temperature rises to 1900°C. The metal and slag are both molten when produced, and the uranium collects at the bottom of the vessel. After two days of cooling, the metal is removed from the vessel as a cylindrical ingot which is about 50 cm in both diameter and height, and weighs about 2 tonnes.

The uranium ingots may be directly fashioned to the required shape, by forging and rolling, or they may be remelted under vacuum and precision cast to shape.

### Uranium Dioxide

Uranium dioxide has already been described as one of the stages in manufacturing green salt. The dioxide produced in this

way, when pulverized or fused, can be fabricated into fuel elements by swaging, vibratory compacting and other methods.

The high-stability fuel for power reactors, however, uses a different quality of uranium dioxide (known as "ceramic grade" dioxide) which is produced by a somewhat different process resulting in a powder with a high specific surface area. This ceramic grade of dioxide can be sintered, without the addition of binding materials, to form high-density pellets. In this process, high purity uranyl nitrate solution is mixed with ammonia to precipitate ammonium di-uranate (ADU). The precipitate is dried and pulverized and then placed in trays stacked in a closed retort containing an atmosphere of inert gas. The retort is heated, causing the ADU to decompose into uranium trioxide; this is reduced to the dioxide by passing hydrogen into the retort to displace the inert gas. After the process is completed, the retort is allowed to cool and the dioxide is removed. The product is pulverized to remove agglomerates and batches are blended to insure consistency in the quality of the uranium dioxide powder.

### Uranium Carbide

Uranium carbide has a higher density and thermal conductivity than uranium dioxide and is a fuel material with considerable promise, particularly for use in organic-cooled reactors. To produce the carbide, uranium dioxide and graphite are pulverized and thoroughly blended. Sintering the mixture in an induction furnace produces a low density uranium carbide, which is transferred to a skull-arc furnace. Here, the carbide is melted and is cast into high-density slugs of the required shape which, after surface grinding, are ready for incorporation in fuel rods.

### Manufacturing Technique

The procedure for manufacturing a fuel bundle depends on the type of bundle to be produced. The description which follows relates to the production of Douglas Point fuel; although there are many differences of detail, the manufacture of NPD and/or Pickering fuel follows the same general pattern.

Fig. 2 shows details of a Douglas Point first charge fuel bundle. The zircaloy tube for making the sheaths is received in batches and samples from each batch are checked for straightness, mean internal diameter, wall thickness and variation of internal diameter. This inspection is followed by an ultrasonic test for flaws which is applied to every tube. Acceptable tubes are cut to length, degreased, and loaded into the magazine of a special type of welding machine for the attachment of end plugs. Twenty tubes are loaded at one time, nineteen of them being a batch of tubes which will be made into one fuel bundle, plus one tube which is a dummy and is used for checking the welding operation.

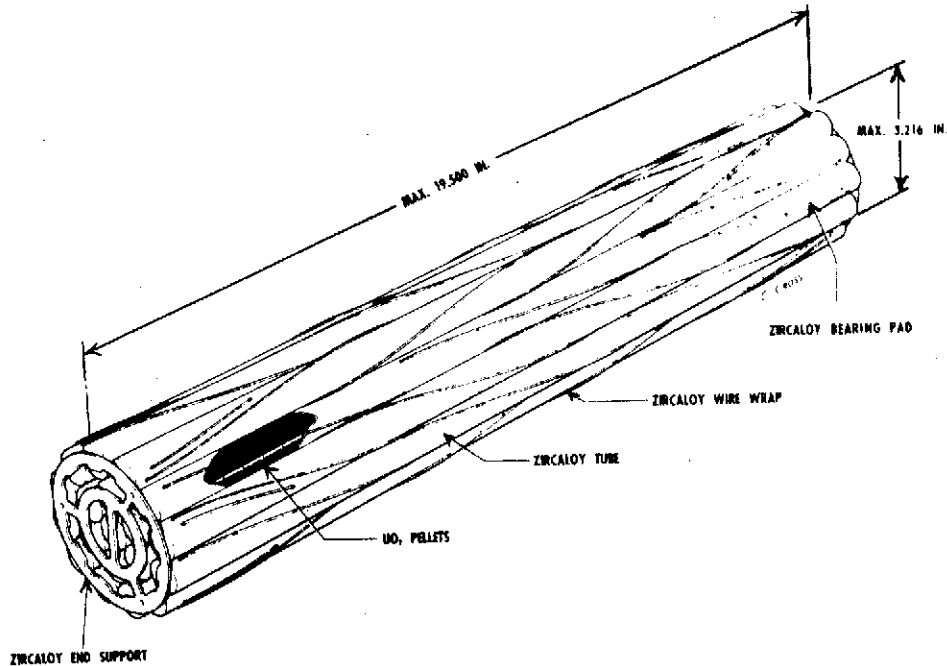


Fig. 2

End plugs, machined from zirconium bar stock are also loaded into the welding machine, which is a resistance welder using an electro-magnetic follower to apply pressure to the end plug while the weld is produced. Each weld is performed individually. The welded sheaths are placed in a 19-element jig with open ends upward. The  $UO_2$  pellets are now loaded into the tubes by hand; as each sheath is filled with 24 pellets, the space remaining is checked to ensure that there will be sufficient clearance for the second end plug.

The filled sheaths are returned to the welding-machine magazine, which is evacuated and then filled with inert gas consisting of argon with 10% helium. The second end-plugs are welded into place to seal the sheaths and the elements are completed by a machine in which the flask from the welding operations is removed and the end plugs are profiled to their final shape. The completed elements are tested for leakage by means of a sensitive helium leak-detector.

The elements are cleaned by pickling and the appendages required for maintaining inter-element spacing attached (wire-wrap spot-welded every inch in the case of the Douglas Point first charge). The welds on a test sample are given a shear test to determine the weld strength. The elements which will form the outer ring of the bundle now have bearing pads added. These are also zirconium wire of thicker diameter than the wire wrap to ensure bundle-to-pressure tube spacing.

The end-plate to end-plug welds are now effected in a jig which rigidly holds the completed elements in their "bundle" positions. These welds and their structure are an improvement over the original NPD design, and will likely be used for Douglas Point replacement fuel, and Pickering. The end-plug has a shallow, conical protrusion on it with a small platform at the base of the cone. When pressed against the end plate, a current impulse is passed between the two. The high current density at the tip of the cone causes the zircaloy to melt, and the end plate settles down onto the platform, producing a strong, reliable weld.

### Other Fuel Designs

There is an extremely broad variation of nuclear fuel in use at the different research institutions and power stations operating today. A few examples of these fuels are given in Fig. 3. These sketches are each drawn to scale, but not all on the same scale.

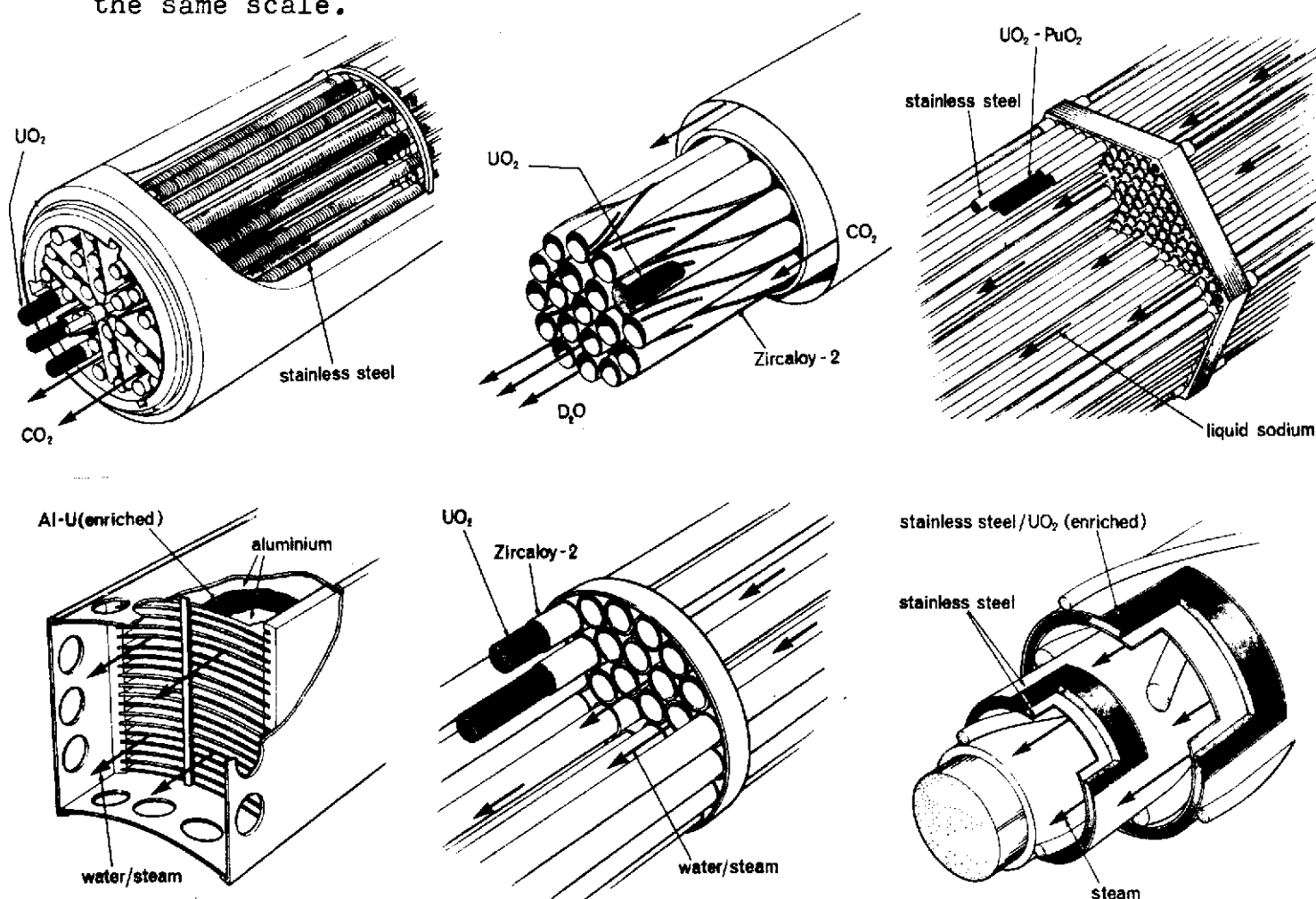


Fig. 3

In the upper row, left to right, the fuels are that of the Advanced Gas Cooled Reactor (AGR); the Douglas Point Reactor and the Prototype Fast Reactor (PFR) in the United Kingdom. In the lower row, left to right, the fuels shown are that of the Materials Test Reactor (MTR), similar in design to that at CRNL in the Pool Test Reactor (PTR); the British Steam Generating Heavy Water Reactor (SGHWR) and the Pathfinder Boiling Water Reactor (BWR), utilizing the superheat element shown.

Ease and cheapness of reprocessing spent fuel in order to recover any remaining fissile material must be kept in mind in designing fuel elements, but cheap initial fabrication is usually more important. In this context, because of the toxicity of plutonium and the radioactivity usually associated with contaminants of U-233, the cost of fabricating fuel elements containing these fissile materials is greater than the cost for fuels enriched in U-235. The standard of reliability of nuclear fuel elements is extremely high, but manufacturing techniques have been developed to meet this high standard at an acceptable cost.

Many of the requirements for solid fuel elements are conflicting - for example, greater reliability can be achieved by making the sheath thicker but only at the expense of increased parasitic neutron absorption - and the weight which must be attached to each factor depends on the type of reactor. The great majority of fuels for power reactors which have achieved competitive performance or which are currently in an advanced stage of development are in the form of metal, an oxide or a carbide, arranged in either a tubular or cylindrical shape.

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