Radiochemistry Group of the Royal Society of Chemistry

The Nuclear Fuel Cycle

(No. 7 in a series of essays on Radioactivity produced by the Royal Society of Chemistry Radiochemistry Group)

Introduction – The Nuclear Fuel Cycle

The nuclear fuel cycle includes all the stages through which uranium passes in its use to produce heat that is then used for the generation of electricity. Mining, fuel manufacture, electricity generation, transportation of used fuel, reprocessing, recycled fuel and waste management are all a part of this cycle.

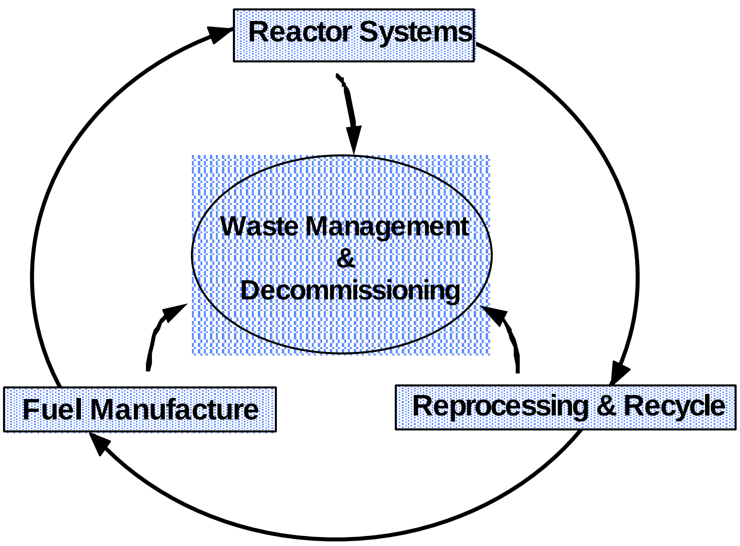


Diagram of the fuel cycle

So let us now consider and find out what each of these stages involve.

Mining Uranium Ore

The major sources of uranium ore are found in Africa, Australia, North America and Russia and are taken from the ground in open cast mines. Only a few of the uranium ores known contain sufficient uranium (greater than 0.1%) to extract commercially. The most important are Pitchblende and Coffinite (silicate).

The ore is usually processed by crushing and grinding to a size of less than 1mm. The uranium is then leached using either acid or alkali and an oxidant to convert U (IV) to U (VI).

Oxidation of U(IV) to U (VI)

$$UO_2 + 2Fe^{3+} \rightarrow UO_2^{2+} + 2Fe^{2+}$$

Acid leaching of U(VI)

$$UO_3 + 2H^+ \rightarrow UO_2^{2+} + H_2O$$

$$UO_2^{2+} + 3SO_3^{2-} \rightarrow UO_2(SO_4)_3^{4-}$$

$$UO_2^{2+} + 3CO_3^{2-} \rightarrow UO_2(CO_3)_3^{4-}$$

Alkaline leaching of U(VI)

$$UO_3 + 3NaCO_3 + H_2O \rightarrow Na_4[UO_2(CO_3)_3] + 2NaOH$$

$$2\ Na_4[UO_2(CO_3)_3] + 6NaOH \rightarrow Na_2U_2O_7 + 2NaCO_3 + 3\ H_2O$$

$$UO3 + Na_2CO_3 + 2NaHCO_3 \rightarrow Na_4[UO_2(CO_3)_3] + H_2O$$

The level of uranium is then concentrated using ion exchange and solvent extraction. The final product is precipitated and dried.

This 'natural' uranium mainly consists of two isotopes, uranium – 235 (0.7%) and uranium – 238 (99.3%). It is the uranium – 235 which is the isotope of interest as it more easily undergoes fission in a nuclear reactor.

First, however, the concentrated ore has to be converted to a form suitable for use within a nuclear reactor.

Fuel Manufacture

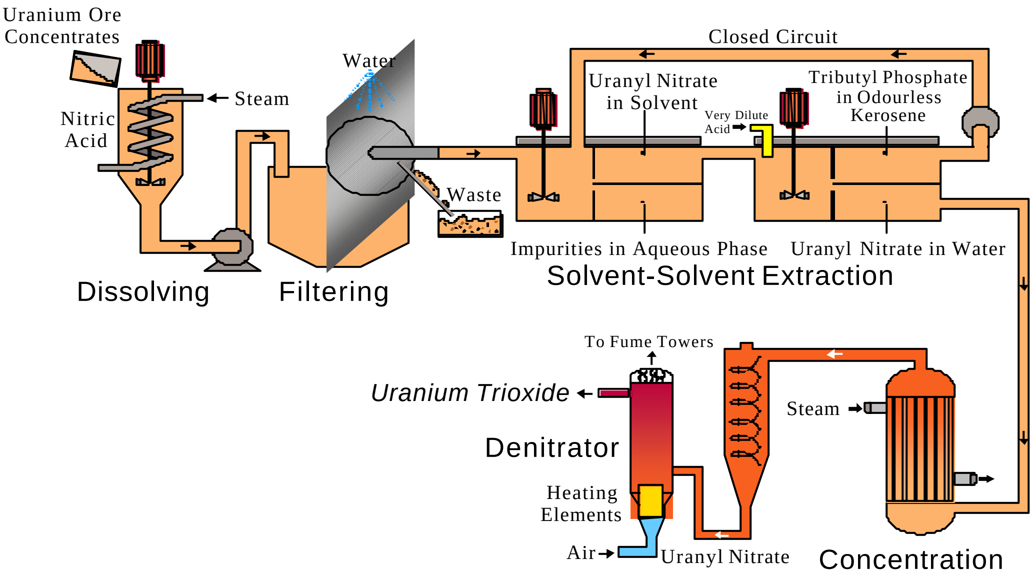
After arrival at the fuel manufacturing plant, the concentrated uranium ore requires further purification to reach nuclear grade. This further purification is to remove neutron poisons, such as boron and cadmium, which would reduce the performance of the fuel in an nuclear reactor, and other impurities which impair the chemical and physical processes needed to produce finished fuel.

Solvent extraction using tri-n-butyl phosphate (TBP) is used to separate uranium from these impurities.

UO_2^{2+} (aqueous) + Impurities (aqueous) + $2N0_3$ (aqueous) + 2TBP (organic)

$\rightarrow UO_2(N0_3)_2$ TBP (organic) + Impurities (aqueous)

The pure uranium in the organic phase is extracted into an aqueous solution using dilute nitric acid.



Uranium Purification Process

Magnox reactors require natural uranium metal fuel and an oxide fuel (UO_2) enriched in the uranium – 235 isotope is used in advanced gas-cooled reactors (AGRs), pressurised water reactors (PWRs) and boiling water reactors (BWRs).

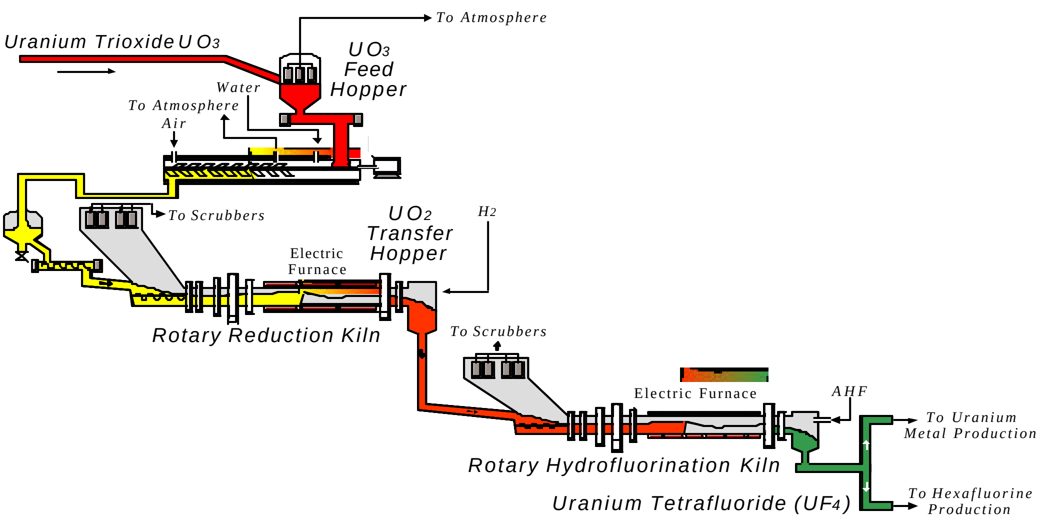
Magnox Fuel

The purified uranium trioxide (UO_3) is converted, firstly to uranium dioxide (UO_2) by hydrogen reduction and then to uranium tetrafluoride (UF_4) by hydrofluorination.

$UO_3 + H_2 \rightarrow UO_2 + H_2O$

$UO_2 + 2HF \rightarrow UF_4 + 2H_2O$

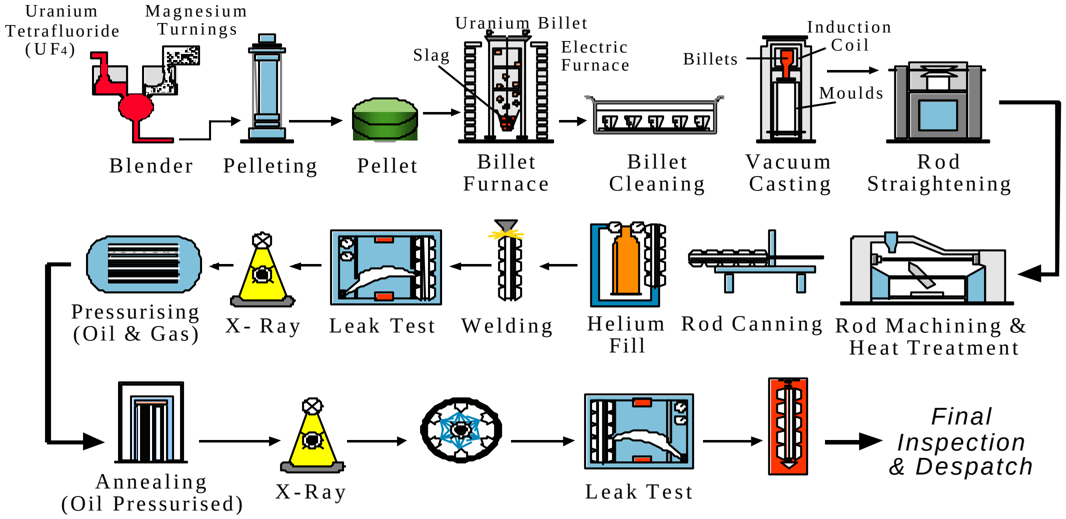
Uranium Tetrafluoride Production



The UF_4 is mixed with magnesium metal, heated to 650°c and the pure uranium metal is formed into a fuel rod.

$$UF_4 + 2Mg \rightarrow U + 2MgF_2$$

The uranium metal rod is encased in a magnesium alloy can from which it gets its name. This type of fuel was the first to generate electricity commercially in the UK.



Magnox Fuel Production

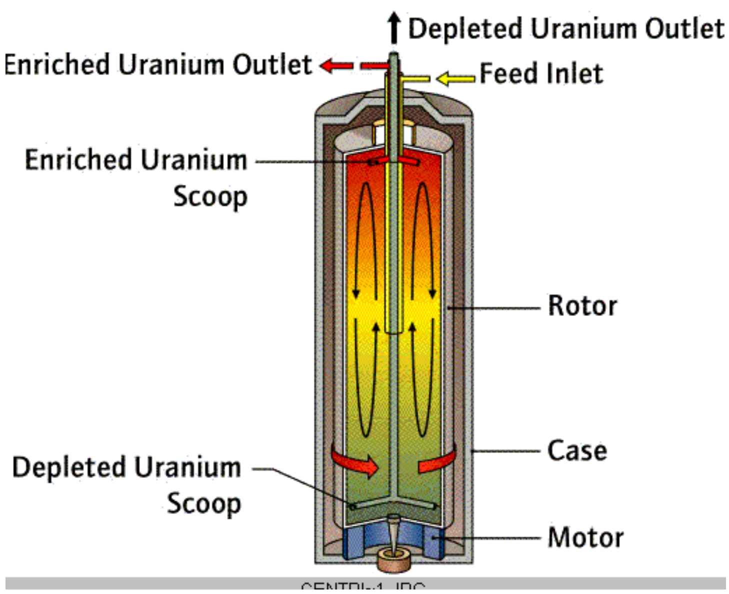
Oxide Fuel

Oxide fuel needs to be enriched in the uranium – 235 isotope to obtain optimum performance from the reactor.

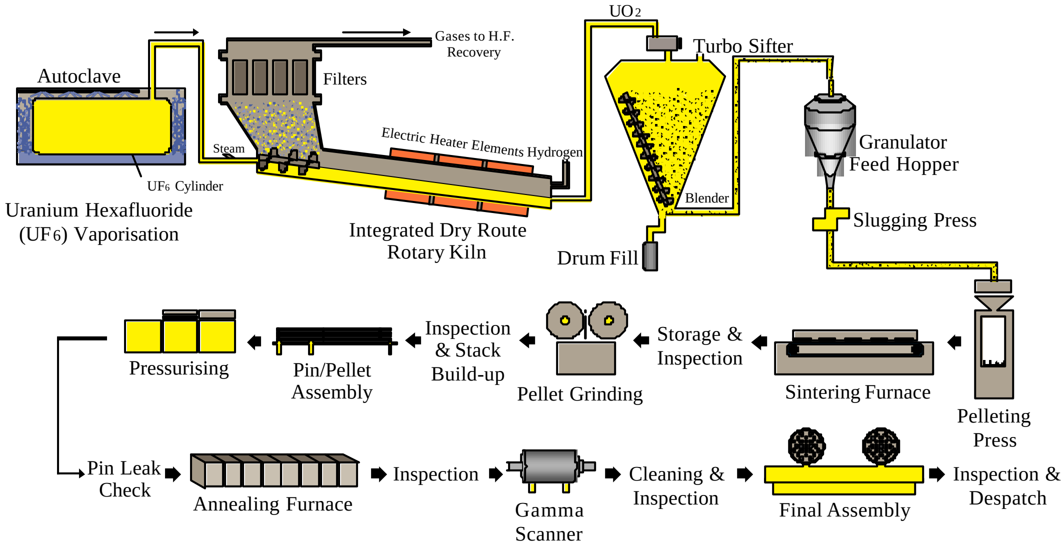
Pure UF_4 is converted to uranium hexafluoride (UF_6) using elemental fluorine. It is this UF_6 which is then used as feed material to the enrichment process which increases the fraction of the uranium – 235 isotope to between 2.5% (AGR) and 4.5% (PWR).

In the United Kingdom, enrichment is carried out using a gas centrifuge which utilises the fact that the heavier uranium – 238 isotope moves towards the periphery whilst the lighter isotope remains towards the centre of the centrifuge.

Gas Centrifuge



This enriched UF_6 is then converted to uranium dioxide, pressed and sintered into pellets and encased in metal tubes to form the fuel assembly.



UO_2 production

Fuel Recycling and Reprocessing

During the time the fuel is in the reactor the amount of the uranium – 235 isotope reduces and fission products and activation products increase. The fuel becomes ‘burnt up’ or ‘spent’.

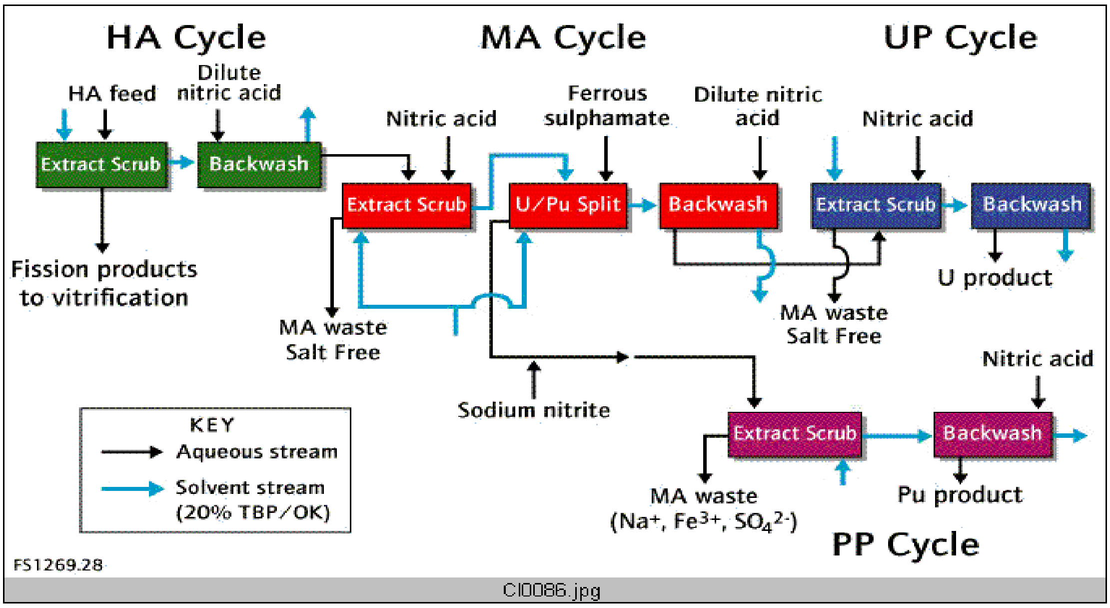
At this point it is removed from the reactor and transported in spent nuclear fuel transport flasks for reprocessing and recycling.

For Magnox fuel, the magnesium alloy can is stripped off the fuel rod before the uranium fuel rod is dissolved in nitric acid. For oxide fuel the fuel pins are cut (sheared) into small lengths and the uranium dioxide is then dissolved by nitric acid leaving the empty pieces of metal cladding undissolved.

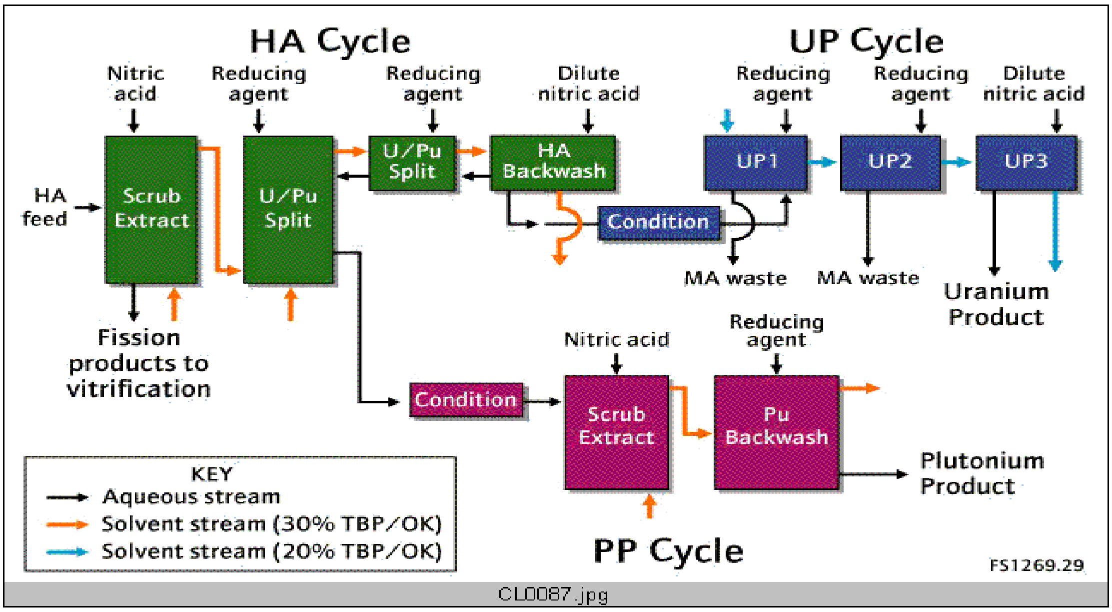
This solution of spent fuel liquor is then purified using tri-n-butyl phosphate (TBP). Both in uranium and plutonium are extracted into the TBP. The aqueous phase containing fission products is transferred to be concentrated and vitrified in glass.

The plutonium is separated from the uranium by selectively reducing the Pu (IV) to Pu (III) which is not extractable by TBP. The uranium is then back extracted from the TBP using dilute nitric acid.

Magnox Reprocessing



Oxide Reprocessing



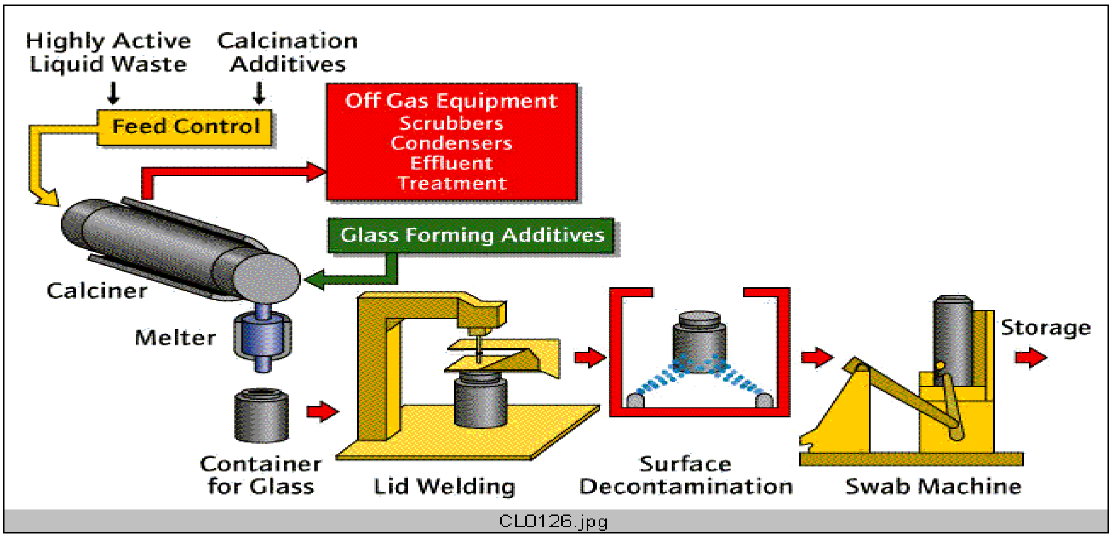
At least 96% of the weight of fuel entering the reprocessing plant is recovered as uranium which can then be recycled and used as new fuel. Up to 1% is recovered as plutonium which can then be used as mixed oxide fuel. About 3% of the fuel is waste which is vitrified in glass. This waste contains 95% of the radioactivity.

All these processes are carried out in heavily shielded facilities using remote handling techniques.

Waste Treatment and Management

The active liquor produced during the recycling and reprocessing of used nuclear fuel is highly radioactive active and continues to generate heat from radioactive decay. Both the activity and the generation of heat define it as a high-level waste. The reprocessing of one tonne of nuclear fuel results in the production of about $0.1m^3$ (100 litres) of the high-level liquid waste. This liquid waste is stored in cooled, double walled stainless steel tanks for a period of time prior to the waste being vitrified. Vitrification is a process that turns the waste into solid glass like blocks that are the wastes final form for long-term storage. The process further reduces the volume of the waste by up to a factor of three.

Vitrification of high level liquid waste from fuel reprocessing



The reprocessing of the fuel also produces of intermediate-level waste, that is a material that is radioactive but not heat generating. Again, processing one tonne of nuclear fuel results in about $1m^3$ of this waste. It consists of items such as fuel cladding and sludges arising from the treatment processes. The waste is compacted, solidified and immobilised by mixing it with a cement based grout within a stainless steel drum giving a final waste form that is suitable for long-term storage.

Encapsulation of intermediate level waste