# DEVELOPING SAFETY IN THE NUCLEAR FUEL CYCLE

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## Summary

The nuclear fuel cycle had its origins in the new technology developed in the 1940s and 50s involving novel physical and chemical processes.

At the front end of the cycle, mining, milling and fuel fabrication all underwent development, especially fuel fabrication but in general the focus of process development and safety concerns was the reprocessing stage, with radiation, contamination and criticality the chief hazards.

Here, the potential for major accidents is not as great as in nuclear fission reactors because of the lower available energy; but energetic events such as criticality, chemical explosions or overheating are possible and early safety studies focused on understanding and protecting against them.

In later decades the research and development studies have given us understanding of the mechanisms and physical events that could lead to accidents and developed a high level of confidence in the protection against major incidents. Issues such as accidental criticality, "red oil" explosion and high level waste tank overheating are examples.

Safety research is not over however and there is still work to be done in advancing technical knowledge to new generation nuclear fuels such as Mixed Oxide Fuel and in refining knowledge of margins and of potential upset conditions. Some comments are made on potential areas for work. The NUCEF facility will provide many useful data to aid safety analysis and accident prevention.

The routine operations in such plants, basically chemical factories, requires industrial safety and in addition the protection of workers against radiation or contamination. The engineering and management measures for this were novel and the early operation of such plants pioneering.

Later commissioning and operating experience has improved routine operating safety, leading to a new generation of factories with highly developed worker protection, engineering safeguards and safety management systems. Ventilation of contamination control zones, remote operation and maintenance, and advanced neutron shielding are engineering examples. In safety management, dose control practices, formally controlled operating procedures and safety cases, and audit processes are comparable with, or lead, best industry practice in other hazardous industries.

Nonetheless it is still important that the knowledge and experience from operating plants continue to be gathered together to provide a common basis for improvement.

The NEA Working Group on Fuel Cycle Safety provides a forum for much of this interchange. Some activities in the Group are described in particular the FINAS incident reporting system.

## Introduction

The early development of reactor technology in the 1940s and 50s led to the establishment of first generation production reactors for generating electricity or for marine propulsion, the latter in a military context. Gas cooled and water cooled reactors were initially developed with a variety of fuels, coolants, and layouts. Over the years the prevalent thermal reactors have come to be water cooled reactors, the majority light water cooled, with uranium oxide fuel. Second generation gas cooled reactors are also oxide fuelled and so fuel cycle technology has focused on uranium extraction, fabrication and reprocessing with some plutonium fuel for fast reactors and plutonium recycle in mixed oxide fuelled thermal reactors. The development of this fuel cycle technology at industrial scale is what I will discuss.

Since this is the first NUCEF symposium I will try to mention safety research; I will also say something about the activities of the OECD/NEA Fuel Cycle Safety Working Group in promoting international exchange on fuel cycle safety. One can gain an interesting perspective on developments over the last fifteen years or so by comparing two state of the art reports on the safety of the nuclear fuel cycle produced by this group. One was published in 1981[1] and one in 1993[2].

Whilst glancing on all areas of the fuel cycle I will concentrate on reprocessing.

## Uranium Mining and Milling

Originally the safety of mining and milling was seen as chiefly industrial safety. Some worker radiological protection was necessary, especially underground, and led to relatively high ventilation rates to sweep away radon in confined spaces. But with many ore bodies at concentrations below .1% of uranium human working is still practicable[3].

Recently significant ore bodies are being developed in Canada at uranium concentrations up to 15%. This is economically attractive, particularly in the current state of over supply of uranium, but human working in such mines is impossible. Remote mining methods will be necessary and radiation protection will become more stringent. It will be a worker safety issue not a public risk, and there are no major research needs.

The environmental impact of mill tailings is potentially very significant if they are not adequately retained[4]. Early experience included both leaks and leaching problems, and the subsequent history has been one of gradual improvement with more forethought put into retention. There is still a legacy in some areas of old tailings.

## Conversion

The risks associated with conversion come from the use of corrosive and reactive agents such as hydrogen fluoride. This is not without its public safety aspects in the case of emergencies, but these are akin to emergencies at conventional chemical factories handling hazardous substances and there is wide experience of this.

## Uranium Enrichment

The principal processes are gaseous diffusion and centrifuge enrichment. Laser enrichment is a recent possibility but not used on an industrial scale.

Both diffusion and centrifuge plant now operate continuously in effect, centrifuge technology is more economic and now has proven reliability of well over 99% per year without significant incident[5].

Enrichment of recycled uranium leads to slightly increased activity from uranium 234 and 232. Industrial scale operations have also shown some neptunium[6]. Technetium 99 may also be significant eventually. But given the low inventory, sealed, reliable plant there are not many important safety issues arising from this part of the cycle. The protection of enrichment tailings from external events involving fire can be an issue.

## Fuel Fabrication

Oxide fuel production is well established at industrial scale in a number of countries (US, UK, France, Germany, Japan). Natural uranium is more a chemical than a radiological hazard, to combat this, the relatively primitive methods of dust control in the early plants have been improved on enormously in second generation facilities. Public risk is not significant.

Fast reactor fuel and mixed oxide fuel brings with it a range of more significant problems. Plutonium requires exacting safeguards. In addition there is significant neutron dose from plutonium 328 and 240 and gamma dose from ingrown Americium 241. Extremely secure containment of plutonium is essential to avoid inhalation. The methods developed are akin to those for plutonium handling at the back end of the fuel cycle and involve double containment, minimising glove box working and using remote operation where possible, and avoiding flammables, or build up of flammables such as hydrogen from radiolysis. The evolution of filtration technology and clean area philosophy has undoubtedly improved both the routine safety and accident resistance of plants.

In sintering furnaces hydrogen is used as a reducing agent and failure to control the furnace atmosphere could potentially lead to a hydrogen explosion. The technology is well understood however and has not resulted in incidents in practice; would be local only in any event.

Criticality probably remains the most serious potential accident, although it would affect workers rather than the public. With typical low enriched material criticality control is by mass and moderator control. More data at very low moderation could gain an increase in mass limits. For mixed oxide material the data on critical conditions are sparse. Research on critical quantities and dimensions for various MOX mixtures could enable margins better to be quantified.

The effects of a criticality incident at this stage are estimated in safety analyses, but there are a few data on moisture containing under moderated powders and research could improve knowledge of potential consequences.[7]

## Spent Fuel Storage

There have been problems in the storage of metal fuel from gas cooled reactors, notably at Sellafield arising from a cessation of reprocessing and the consequent need to store for longer than anticipated. This led to particular problems with caesium in the pond water and a need to remove that to avoid excessive discharges. That is a metal fuel problem. For the bulk of oxide fuel, zircaloy or stainless clad, there are both wet and dry storage alternatives available. Both are used on a commercial scale. Some countries, notably Germany, have developed transport casks for dry storage.

Surveys of experience and understanding[8] have concluded that feasible and proven technologies exist already for both short and long term storage of spent fuel. Further research needs are probably limited to refinements of procedures such as re-racking in wet storage ponds. There may still be some room for refinement of source terms quantifying the potential releases following accidental draining of a wet storage pool. But the engineering of such facilities makes drainage an extremely low probability event associated with such things as earthquakes beyond the design basis. The need for source term refinement cannot therefore be considered to be great.

Gaining data on long term degradation mechanisms is part of the IAEA's BEFAST programmes.

### Fuel Reprocessing

World wide reprocessing experience is already well over $10^5$ thm of fuel; the vast bulk of this has been reprocessed by liquid/liquid extraction between nitric acid in water and TBP in kerosene, in the so called PUREX process.

Reprocessing is in many ways the natural area for concern in fuel cycle safety. Radioactive inventories are high, and the material is at most stages in a readily dispersible form. But a key difference from reactors, which also have a high inventory, is the dispersive energy available. In a reactor at power this is extremely large and can lead to consequences on a Chernobyl scale. In a reprocessing plant energies are much smaller and associated with local disruptive events such as criticality, fire, and explosion.

External events which may breach containment such as aircraft crash or earthquake also require some thought and engineering.

I will discuss briefly some of the hazards involved in this process and areas where safety research may have a part to play in improving safety or the efficiency of what is already an established industrial activity.

### Criticality and Fissile Material

The nuclear fuel cycle is distinguished from other chemical processes handling toxic substances by two chief hazards: radiation and criticality. Radiation, a most important issue in worker protection is obviated by shielding. Well validated calculational methods are available.

Criticality is a complex phenomenon because of the interaction of the physical parameters such as absorption, fissile cross section, reflection. Not all are accurately known and calculation relies in practice on calibration from experimental measurements as close as possible to the system concerned. Simple calculational methods are based on handbooks that interpolate data points and are reliable and backed up by practical experience. But in industrial size plants quite complex computer codes are necessary to cope with large process quantities and all the operational circumstances. An example is that of uranium and plutonium nitrate solutions at relatively low enrichments. There are data on the relative components in solution but for mixed nitrate solutions process parameters established by calculation may still have over conservative safety margins and further measurements would support future MOX operations.

The problem areas are those where material location and quantity is unexpected or difficult to determine. An example could be the head shear pack under maintenance when moderator might mix with a quantity of retained fuel fines.

Many of the early criticality incidents (generally from the 1950s and early 60s) were caused by failures in the material control rather than gaps in the knowledge of critical systems. Typically mixing of undermoderated systems with moderator through stirring up solid residues in vessels or adding moderator. This has sometimes been associated (as in the Windscale incident in 1970) with an un-anticipated accumulation of material.

Whilst it is always the aim to avoid this by incorporating engineering safeguards it is normal practice to measure fissile inventory at key points. This is particularly difficult at the head end where the exact inventory of incoming fuel is subject to some uncertainty and the first accurate assay is in the accountancy tanks. In the dissolver it is hence common to use a neutron poison to ensure criticality safety. Poison is often expensive, such as gadolinium, and the amount added depends on the confidence one is prepared to put on knowing plant parameters such as the location of undissolved fuel, the burn up, reflection, and the nuclear cross sections.

It is an economic as much as a safety issue and since many of these plant parameters are very plant specific generic research is less likely to be helpful. Measurements of materials remaining in the hulls is normally by some technique such as neutron interrogation. Improved on-line assay methods here and in other process flows could improve operability and safety.

Another specific plant area where difficulties occur is in the waste area where material contaminated to a low level by transuranic waste must be taken and stored for later treatment. The assay of such low levels of fissile content is not particularly easy and leads to plant restrictions that are sometimes grossly conservative because one has to assume a fissile content at the level of resolution of the measuring equipment. (Similar issues apply in TRU waste treatment and disposal facilities). Development of measuring techniques is an area where research could bring improvements, as much in economics as safety.

Turning to the consequences of a criticality accident, these will in general consist of one or more pulses of radioactivity, perhaps accompanied by energetic effects and dispersion of material. Direct data are based on what can be deduced from early incidents together with experiments such as those conducted in the French CRAC facility. When using this information in safety analysis it is generally necessary to make assumptions that are conservative. In low enriched aqueous systems there may be scope to refine these by research and conservatism reduce. In heterogeneous systems where there is a possibility of a relatively rapid input of positive reactivity due to the criticality itself, it is not entirely clear that safety analysis assumptions are conservative.

### Fires and Explosions

Kerosene is not a highly flammable solvent and most reprocessing plant operations are at relatively low temperature. It is not a problem provided there is good process protection against hydrocarbons passing with process flows into high temperature equipment such as furnaces. But there has been some concern, arising from both experimental work and past plant incidents, because it is possible to get rapid exothermic reactions from the oxidation of zircaloy fines or from the decomposition of mixtures of irradiated solvents, heavy metals, and nitric acid (sometimes called red oil explosions). The latter in particular has a complicated chemistry and there is not a complete fundamental understanding of the processes involved. However there has been a significant amount of research and development on both systems directed at establishing the boundary conditions within which rapid decomposition can occur. That enables plant equipment such as evaporators to be operated in a safe regime. It may be that further work would establish greater confidence and allow margins to be reduced with possible improvements in the efficiency of plant operation. However the complexities of the system are great.

### External Events

External events such as wind, waves, aircrash and earthquake must be taken into account on an appropriate design basis, usually based upon the return period involved. This is the same approach as for reactors and other nuclear facilities and in general the predictive methods already developed can be used for plant siting. Engineering techniques already developed, particularly seismic engineering, can be carried into fuel cycle plant design to ensure that equipment failures caused by external events do not lead to release of material from the plant and that essential services such as coolant water are available at sufficiently high reliability. Some plant specific research or tests on equipment may be needed but further generic research is not.

**Waste Issues**

Some issues dealing with TRU have already been discussed. High level waste, or heat generating waste, has some special problems associated with it. The short term storage must have adequate heat rejection and is generally in liquid form. There is now relatively long term experience in this. Summaries drawing on the experience of the 70s and 80s[10] are still relevant. Most systems in use retain spare tank capacity to cope with an un-anticipated event such as corrosion failure, and also have highly redundant cooling systems to assure reliability. So whilst some years ago the boiling dry of a high level waste tank was considered an incident of potential concern, its probability is now recognised as being very low; in addition more recent studies[11] have shown that for beyond design basis accidents and boiling dry the bulk of activity release is delayed until the ultimate phase of approaching dryness when ruthenium in particular is volatilised. On the timescales involved emergency restoration of cooling by other means is almost certain and this accident sequence is hence of less concern.

**Routine Emissions**

Long lived isotopes, $^{85}$Kr, $^{129}$I, $^{14}$C, were for some time an issue in reprocessing wastes. Krypton is not easily trapped, but recent studies on its radiological impact have shown it to be less important than originally thought[13], especially in view of the limited world reprocessing capacity. Established scrubbing and encapsulation techniques are available for iodine and carbon[14]. Whether long lived isotopes are trapped or not is generally considered to be an issue of what is practicable at reasonable cost. They do not present a high level risk level.

**Encapsulation**

Vitrification has now become an established technology with industrial scale plants based on French design operating in France and the United Kingdom[12]. The process involves much higher temperatures than other stages but no differences in principle. There are currently no operating repositories for high level waste but the principle of geological barriers is widely researched.

Encapsulation of medium level waste is still a live safety issue for long lived isotopes since their chemical form is related to final disposal needs. But waste issues are not the central subject of this symposium.

**Learning from Experience**

Safety research has over the years underpinned design and improved protection against accidents of any size, certainly against those accidents with the potential for affecting anyone off site.

In the case of worker risks however, much if not all the improvement in safety has come from experience with plants and refinement either in management methods or in engineering protection. As examples of management practice, the use of contamination control zones is now highly developed, the practice of dose budgeting for intervention and maintenance operations as well as routine operations have led to reduction in worker doses As engineering examples, additional shielding and second generation glove boxes or remote operation have had an impact on dose from dusty material and plutonium in particular.

In learning from experience it is obviously valuable to draw on as wide an experience base as possible. The Fuel Cycle Safety Working Group, acting within OECD/NEA as a sub group of the CSNI takes the exchange of experience as a regular agenda item. The aim is to ensure that all member countries are aware of significant evolutions in practice or of individual incidents or events that can be learned from. Not necessarily those of any significant consequence but those with significant potential consequence.

Over the last two or three years we have introduced the FINAS system in an attempt to systematise the flow of information and aid reporting and awareness easier. It is analogous in some ways to the IRS system for reactors. After a relatively slow start the system now has upwards of fifty events on it. We are currently reviewing its effectiveness and we hope to be able to make it an additional important mechanism for international exchange.

Routine operational safety is likely to show continuing improvement through the proper utilisation of experience both national and international. For example, corrosion and degradation processes are often revealed through experience rather than experiment because of differences between plant and laboratory conditions.

### Advanced Reprocessing

A major change in the core technology away from the PUREX process would take a considerable time to evolve. This is not likely for the foreseeable future. But there are improvements that might be made, possibly in separation of TRU wastes or confinement of activity and trapping close to source. So the story is not yet over for safety research on process improvements.

### Conclusion

The nuclear fuel cycle is established as a safe industrial process. Safety research will now bring incremental improvements and will quantify margins rather than leading to a step change. It may be valuable to do work in areas such as criticality of low enriched systems and mixed systems; assay, treatment, and handling of TRU material; and refinement of safety analysis methods dealing with hazards such as fires.

Exchange of operational experience internationally provides a broader basis for steady improvements in operational safety.

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