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COEX™ process: cross-breeding between innovation and industrial experience

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Abstract

Recycling used nuclear fuel at an industrial scale has been a reality for over 40 years. Since it was founded in 1976, AREVA has designed and built two used fuel treatment plants in La Hague, France. These plants, named UP2-800 and UP3, use the PUREX process. UP3 began operations at the end of the '80s and UP2-800 in the mid '90s. The plutonium extracted in UP2-800 and UP3 is then processed in MELOX plant which started operation in 1995, to be recycled under the form of MOX fuel in LWR.
This technology has been selected by JNFL for its reprocessing and recycling plants. Rokkasho-Mura reprocessing plant incorporates also some Japanese technologies and is being commissioned soon.
Over 23,000 tons of LWR used fuels have been treated in La Hague plants and over 1200 tons of MOX fuels have been produced by MELOX plant.
Innovations have been constantly incorporated to these plants to improve process efficiency and to reduce the activity and volume of waste. During these years, AREVA has acquired an invaluable experience in industrializing processes and technologies developed in the laboratory.
In the frame of its continuous improvement policy, AREVA has developed jointly with CEA (French Atomic Energy Agency) a new process, COEX™ process, offering significant improvement in term of proliferation resistance, process performance and investment and operating cost.
The present paper recalls the process principles applied in French and Japanese recycling plants.
Then it describes the main steps of COEX™ process, the status of its development and the improvements compared to PUREX process.
The possible evolution of COEX™ process to cope with needs of future nuclear fuel cycles using fast reactors and possible recycling of minor actinides is presented.

Introduction

In the seventies, based on the assumption of a rapid growth of nuclear energy and uranium demand, closed fuel cycle was implemented at industrial scale using the PUREX process. The first generation of commercial plants dedicated to LWR oxide fuels appeared in Europe (France, Belgium and Britain) and in Japan. These plants have been designed and started up in the 60's and 70's, and were devoted at first to fuels issuing from gas-graphite reactors, and then to LWR fuels. They had limited capacity.
The requirements for process performances in commercial plants are stringent: very high purity of uranium and plutonium end products and high recovery ratio, authorized limits for release becoming more and more severe, necessity to condition all waste.
The risks of criticality, contamination and irradiation have to be mitigated and the requirements are also more and more stringent, especially concerning dose to operators.
The second generation of commercial reprocessing plants was designed in the 80's, taking advantages of lessons learned from first generation plants, and of a huge R&D effort aimed at:

- Increase plant availability through:
  - the use of corrosion free materials, especially for the equipment with boiling nitric acid solutions,
  - fully remote maintenance,
  - duplication of most sensitive equipment,
- Improve process performances by adopting fully continuous process and through stable performances of extraction process,
- Improve safety records: reduction of dose rate to operators and of release to the environment,
- Conditioning all process and technological waste.

Four reprocessing plants are currently under operation or final phase of commissioning: UP3 and UP2-800 at La Hague, France, THORP in UK and Rokkasho-Mura in Japan.
The plutonium extracted in UP2-800 and UP3 is then processed in MELOX plant which started operation in 1995, to be recycled under the form of MOX fuel in LWR.
Over 23,000 tons of LWR used fuels have been treated in La Hague plants and over 1200 tons of MOX fuels have been produced by MELOX plant.
Innovations have been constantly incorporated to these plants to improve process efficiency and to reduce the activity and volume of waste.
It is worth to notice that the strong co-operation between R&D, engineering and operators has played a key role.

COEX™ process

After the successful start-up of UP3, UP2-800 and MELOX plants, R&D effort has been pursued and numerous advances have been made possible thanks to it.
A major step has been set up with the development of a new reprocessing-recycling concept aimed at:

- Further enhance proliferation resistance,
- Maintain high level of process performances
- Minimize both investment and operation cost
- Take advantage of present industrial experience
- Keep open possible evolutions to take into account new type of reactors or change in trans-uranic elements management

The COEX™ process results from several developments started up more than 10 years ago, and gathered through an original engineering approach.
One of the main drivers was to benefit from the industrial experience of existing plants in order to avoid long and costly technological development, to be able to propose this new concept for design of new recycling plants right away.

Process description

The main characteristics of COEX™ are:

- No plutonium separation at any point of the process
- Use of co-conversion process to produce a solid solution of (U, Pu)O_2

Various implementations are possible for COEX™. The implementation presented in this document is aimed at:

- Minimization of plutonium under solid form, even while mixed with uranium
- Integration of reprocessing and recycling facilities on a single site

To better illustrate the differences between existing facilities and COEX™, the following figures give the block flow diagram of La Hague/MELOX, Rokkasho-Mura and COEX™:

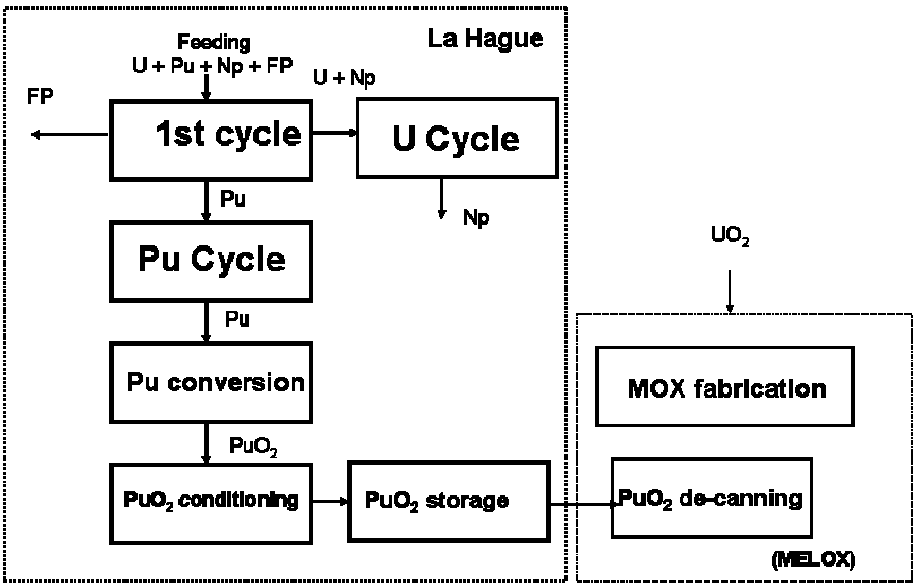

Figure 1: reprocessing/recycling process as implemented in La Hague and MELOX

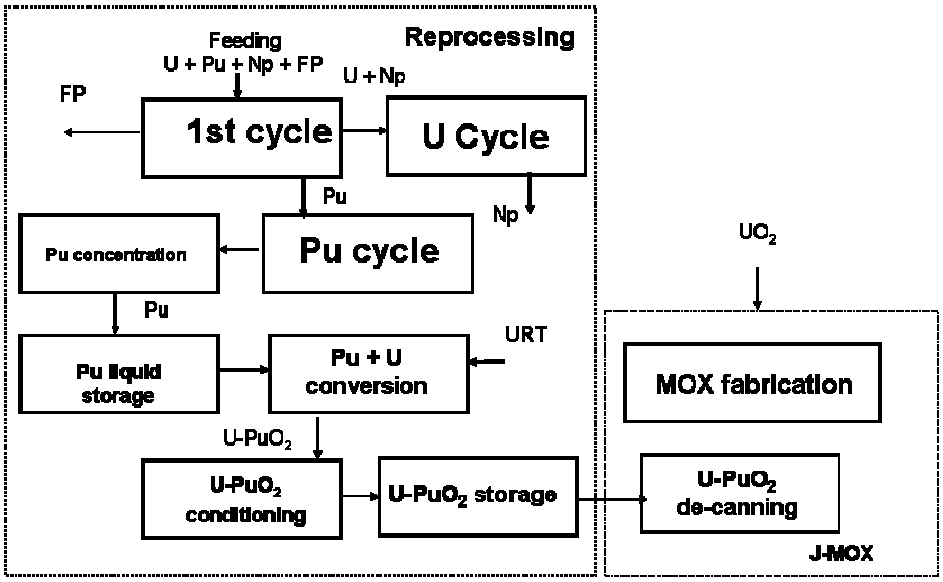

Figure 2: reprocessing/recycling process as implemented in Rokkasho-Mura

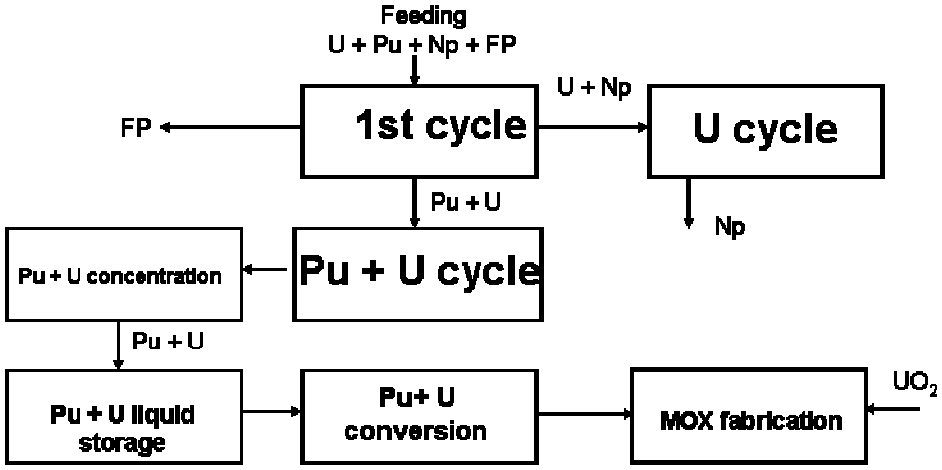

Figure 3: COEX™ process

The main evolution between PUREX and COEX™ are:

- no separation of plutonium in separation stage of first cycle
- concentration of (U,Pu) solution before conversion
- storage of concentrated (U,Pu) solutions
- use of co-conversion process, to produce (U, Pu)O_2 solid-solid solution
- direct feed of mixed oxide from conversion to MOX fabrication

This scheme implies simultaneous construction and operation of reprocessing and recycling facilities, which was not the case in both La Hague/MELOX and Rokkasho.

Extraction principles

The following figures explain the COEX™ chemical flowsheet.

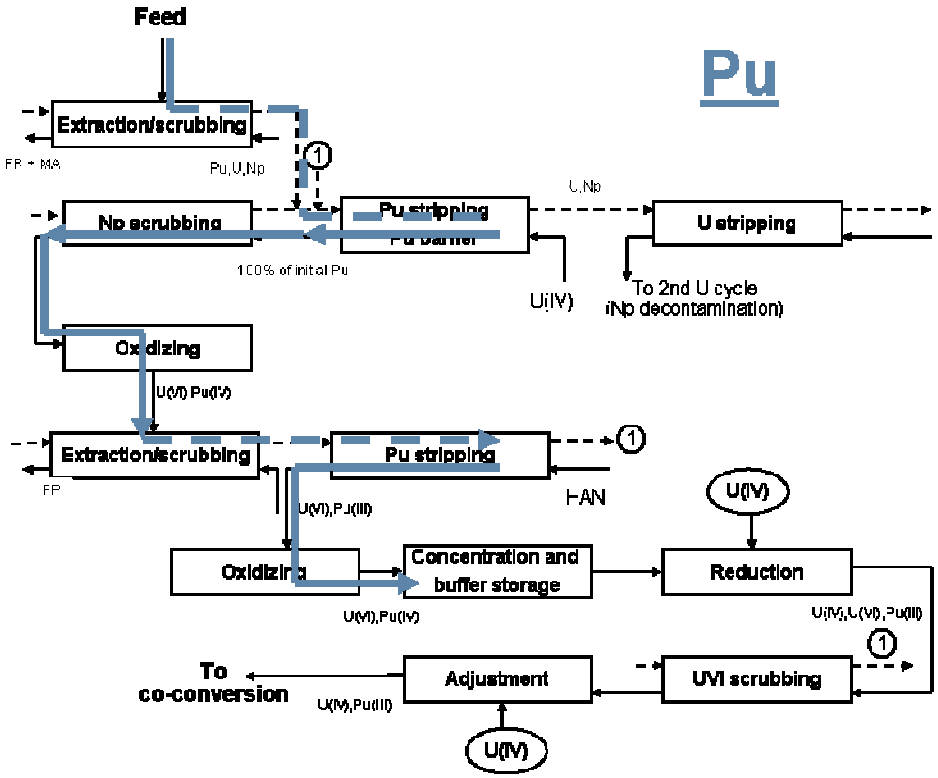

Figure 4: plutonium routing

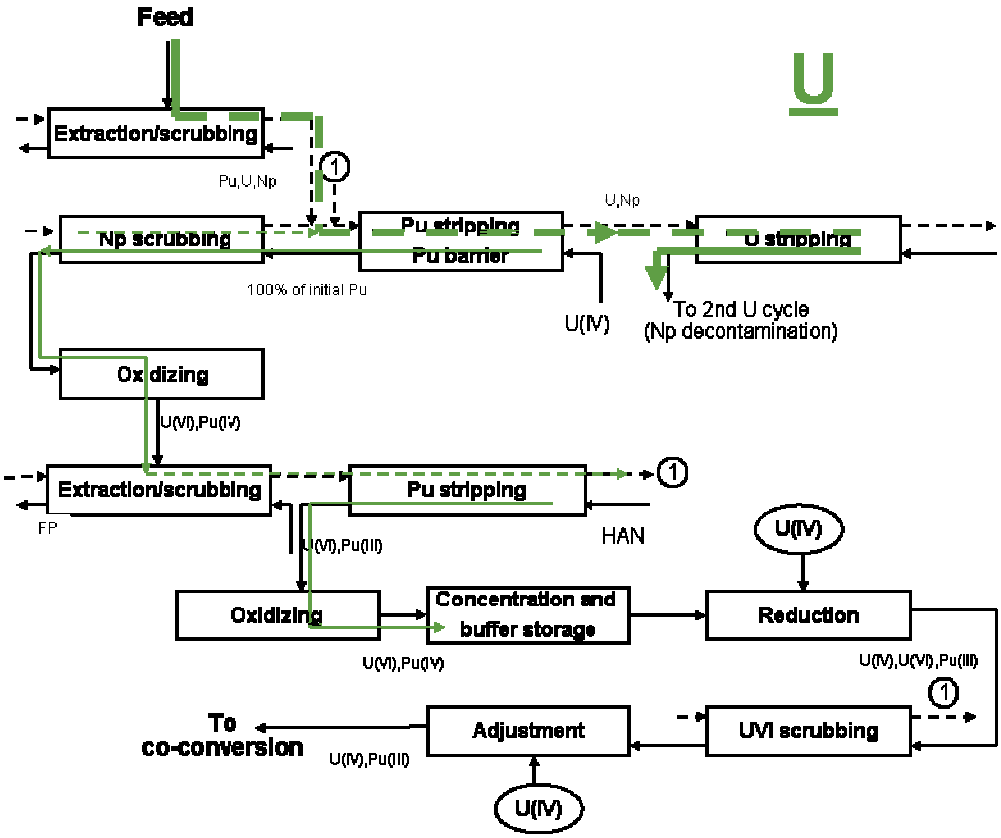

Figure 5: uranium routing

The figures 4 and 5 explain the routing of (U,Pu) and uranium: the operating parameters of Np scrubbing in (U,Pu) separation stage of first extraction cycle is modified to avoid Np to follow (U,Pu) stream, and to allow some uranium to be stripped with plutonium.
Most of the uranium is sent to U stripping operation and later on to 2nd uranium cycle for Np and FP decontamination in order to meet U product specification.
In (U,Pu) purification cycle, U scrubbing stage is deleted, thus allowing uranium to follow plutonium stream up to the concentration step.
To ensure safe operation of concentration, uranium and plutonium are oxidised to valences VI and IV. After storage, valence and uranium content are adjusted in order to get suitable conditions for conversion.

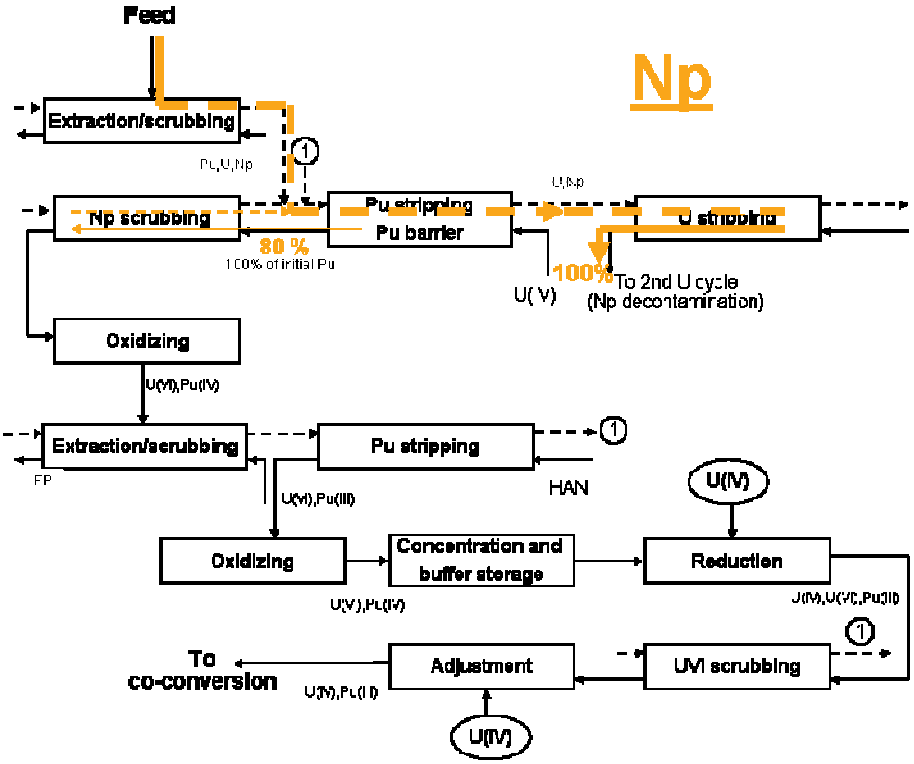

Figure 6: neptunium routing

Neptunium routing is same as for PUREX process: neptunium follows main uranium stream in first extraction cycle.

Conversion process

R&D has been conducted for over ten years to develop an oxalic co-conversion process allowing obtaining a homogeneous blend (solid solution). The figure 7 shows the mesh parameter (or lattice parameter) for a $(U,Pu)O_2$ powder obtained by oxalic conversion, as a function of U/Pu ratio. According to Vegard law, a solid solution correspond to a mesh parameter located is on the blue line, It has been found, that plutonium content should not exceed 50 % to reach this goal. Over this ratio, two phases co-exist: for instance at 70 % of Pu, two values appear, one corresponding to pure PuO_2, and another corresponding to a mixture 50/50 between U and Pu.
Therefore a target of 45 % of plutonium has been set to keep operating margin.

The choice of oxalic conversion process, already used today in La Hague plants for years, allows using same industrial equipment as in existing plants.

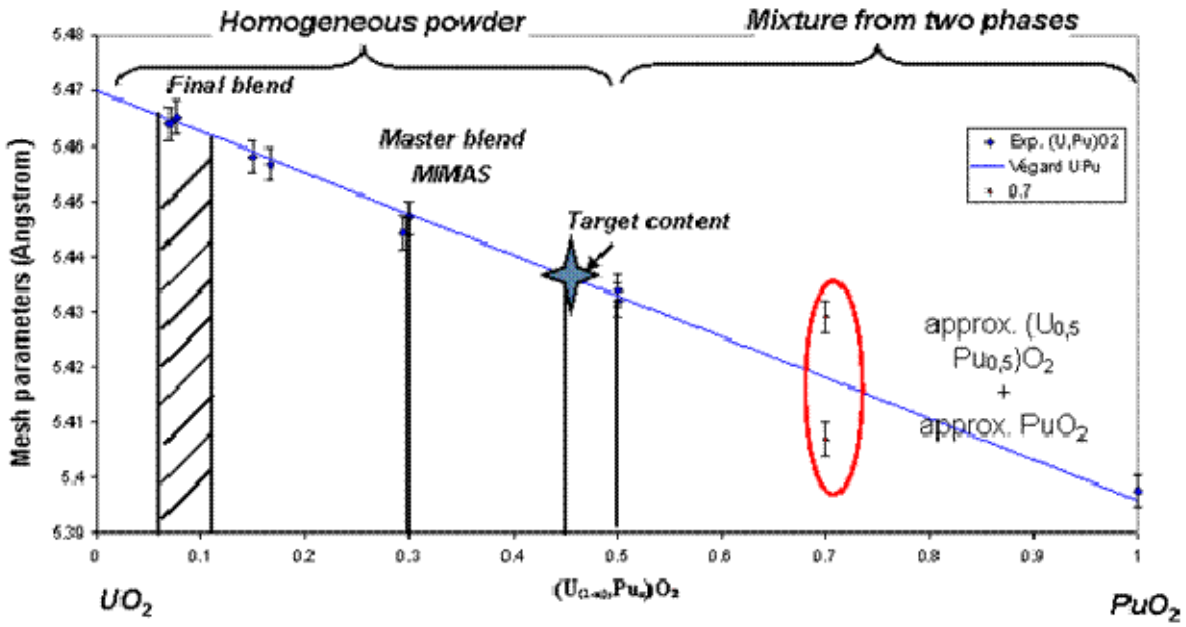

Figure 7: powder quality versus U/Pu ratio

Future development

COEX™ is an evolutionary process.
A first possibility is to produce (Np,U, Pu) mixed oxide, for instance to fabricate mixed oxide fuel for fast breeder reactor.
A quite simple modification of (U,Pu) stripping flowsheet in first extraction cycle allows neptunium to follow (U,Pu) stream. Then in (U,Pu) purification cycle, providing some minor flowsheet modification, neptunium will follow (U,Pu) stream up to concentration stage.
The feasibility of co-conversion for a mixture of uranium, plutonium and neptunium has been demonstrated at laboratory scale.
Later on, it could be envisaged also to separate others minor actinides, mainly americium and curium, for specific management. This could be achieved by implementing DIAMEX and SANEX process on the raffinates of first extraction cycle as shown on figure 8.

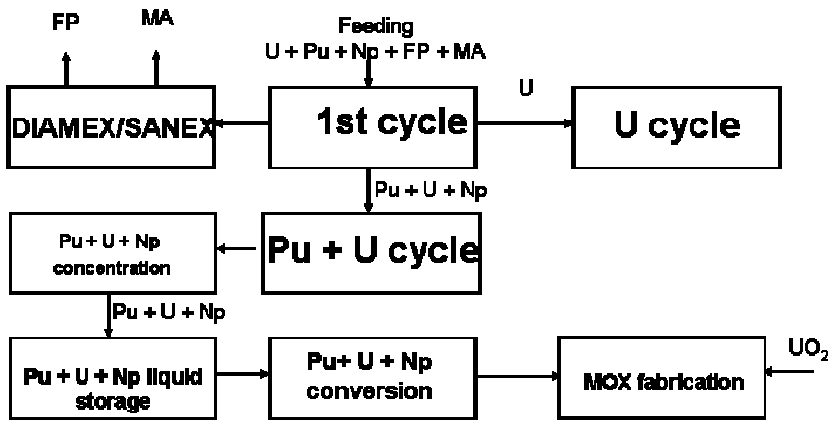

Figure 8: COEX™ process with separation of minor actinides

CONCLUSION

The development of COEX™ process (patent pending) has reached the goals presented previously:

- Resistance to proliferation is enhanced by:
  - no separation of plutonium
  - no storage of plutonium under solid form
  - minimization of plutonium hold-up between reprocessing and fuel fabrication
- High level of process performances: COEX™ is an evolutionary process based on technologies which have proven their efficiency at industrial scale in existing commercial reprocessing plants. The co-conversion process feasibility is acquired.
- Comprehensive liquid waste management, for both aqueous and organic waste,
- Investment and operating cost are minimized thanks to reduction of main process functions (refer to figures 1 to 3)
- Evolution of COEX™ process are possible to deal with new requirements of radio nuclides management

Other processes are under development worldwide, at least at laboratory scale. However, compared to existing commercial reprocessing plants, most of them imply significant modification upstream of extraction process at dissolution stage, for instance, of downstream in liquid waste management, due to significant evolution of composition or flow rates. Most of theses processes use new solvent for which a specific management need to be developed, COEX™ process will be able to take advantage of solvent management developed for La Hague and Rokkasho plants, which has proven its efficiency both for process performances and for final conditioning of waste.

The chemistry of COEX™ process is well understood and builds on the over 50 years of industrial scale experience of reprocessing. The COEX™ process will be demonstrated at laboratory scale using active solutions. The demonstrated success of Purex process shows that COEX™ should be scale-up readily and without large scale tests.