

WASTE TO STABLE SALT (WATSS) SPENT FUEL RECYCLING

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Abstract

The Waste To Stable Salt (WATSS) process is a groundbreaking nuclear waste recycling technology designed with versatility and efficiency in mind. The WATSS process reduces waste liabilities while unlocking unmatched value from existing fuel resources. When combined with a fast spectrum reactor such as the Stable Salt – Wasteburner (SSR-W), it enables the closing of the fuel cycle with no need for uranium enrichment.

As the core stage of the process, the extraction of transuranic elements from the uranium oxide matrix can be obtained by the direct conversion of transuranic oxides in molten salt to chloride or oxychloride species, while uranium oxide ultimately settles as insoluble dioxide solid material. This is based on a pyrochemical processing technique relying exclusively on chemical redox potential for the extraction of components of spent fuel in molten salt. This is significantly less complex than pyro-processing based on electrochemical separation.

Computational tools and experiments with simulant spent fuel have been combined to determine commercially viable extraction conditions, and a set of options have been tested with real spent fuel at the Canadian Nuclear Laboratories (CNL) facilities in Chalk River. The results validate the viability of the selective extraction, which is the main part of the broader Waste-to-Stable-Salt (WATSS) fuel recycling process.

1. Introduction

As the demand for electricity increases in daily life, from electric vehicles to data centers, the urgency to reduce our reliance on fossil fuels has never been greater. It also becomes evident that intermittent and seasonally driven sources need to be backed by a stable provision of base load power, renewing interest in new nuclear capacity as a reliable and sustainable solution. Two fundamental considerations need to be addressed to ensure the long-term sustainability of this option: the continued availability of fissile fuel and the suitable management of waste. Both factors may be addressed by realizing that the discharged fuel from the current fleet of reactors, considered as “spent fuel” still contains a significant amount of fissile material.

Whether it is for fuel availability or for waste management considerations, it becomes more and more important to view spent fuel recycling as a sustainable way forward, especially the grouped recycling of transuranics (TRU - unseparated Pu and minor actinides) for their consumption in fast-spectrum molten salt reactors. This is the purpose of the Waste to Stable Salt (WATSS) process developed by Moltex Energy. The environmental merits of spent fuel recycling, and especially the combination of multi-recycling with fast reactors, are highlighted in several recent reports [1,2]. Specific considerations of the WATSS process on waste categorization and waste repository footprint are discussed in [3].

An important consideration in the choice of a nuclear fuel cycle strategy is the acknowledgement that irradiated fuel contains extremely valuable components, often difficult or impossible to acquire elsewhere. This obviously relates to actinides that could be re-used as fuel for further energy production, which is already a compelling reason to consider spent fuel recycling as a means to mitigate fuel supply security issues, limited natural resources and access to uranium enrichment. But the potential value of spent fuel is broader and also relates to a wide range of increasingly valuable elements and isotopes for other applications, including in the medical field [2].

Reprocessing and recycling of used fuel are traditionally considered sensitive activities in terms of proliferation since the Plutonium-Uranium Redox EXtraction (PUREX), the process currently used at an industrial scale to recover plutonium from spent fuel for the production of Pu-U mixed oxides (MOX), may also be applied to - and has initially been developed for - the production of weapons-grade material. Besides its proliferation concerns, this legacy method has proved to be expensive and involves huge plants due to the large volumes of solvents required. However, if the recycled material is intended to be consumed in a fast spectrum reactor, such as the Stable Salt Reactor – Wasteburner (SSR-W) developed by Moltex, plutonium doesn't need to be separated from other actinides [4]. This opens the door to different methods of recycling, with considerably increased potential for proliferation resistance [5-7]. Among these methods are a set of processes broadly referred to as pyro-processing, where molten salts are used as the solvent for the chemical species instead of water and organic solvents.

2. Prior Art

The most developed pyro-process was developed at the Argonne and Idaho National Laboratories in the USA [8]. This process was originally developed to separate TRUs from uranium in metallic nuclear fuel from the EBR-II reactor. It involves selective electrochemical transporting of uranium from the metallic fuel to an iron electrode, and then electrochemically transporting TRUs to a molten metal electrode. The process has been extended to uranium oxide spent fuel by adding an initial stage of electrochemically reducing the uranium oxide to metal using a molten salt electrolyte and a consumable carbon anode [9]. While this method has clear advantages over the PUREX process it requires two consecutive electrochemical treatments in separate apparatus and is thus relatively complex and expensive.

A second pyrochemical process has been less investigated but showed significant promise of being able to simply chemically extract TRUs from spent fuel leaving the uranium as an oxide. This process was first considered by Wenz in the late 1960's [10] who showed that oxidation of UO₂ pellets to U₃O₈ by a process known as voloxidation (which at the same time breaks down pellets into a powder), followed by extraction in molten MgCl₂ based salts, with a reducing metal added to reconvert the uranium to UO₂, resulted in as much as 95% of the TRUs dissolving in the salt while most of the uranium remained as solid oxide. Further work on this process did not take place however as the only metals found to be effective as reducing agents were tantalum and zinc. Since stoichiometric amounts of reducing metal are required to reduce U₃O₈ to UO₂, tantalum was impractical for commercial use due to its high cost. Zinc has a much lower cost but has problems of being volatile at the extraction temperatures and leaving large amounts of ZnCl₂ in the molten salt which can interfere with subsequent purification of the TRUs and make reuse of the salt challenging which results in very high volumes of highly radioactive and water-soluble waste.

A related process to that described by Wenz has been demonstrated at the Idaho National Laboratory [11]. In this process, uranium oxide fuel, again following oxidation to U₃O₈, was extracted in a

LiCl/KCl molten salt containing UCl_3 . Uranium metal was added as a reducing agent either directly or by an electro-reduction process to convert the U_3O_8 to UO_2 and 90% extraction of transuranic elements was achieved, though the molten salt also contained substantial amounts of uranium from the UCl_3 initially added. Effective separation of TRUs from uranium was not therefore achieved in these tests.

Thus, while these direct extraction processes show promise, a practical application of them has not to date been achieved. The key challenge is to identify a molten salt extraction system that will reduce transuranic metal oxides to salt soluble chlorides and/or oxychlorides, that does not require large amounts of expensive reducing agents and which does not result in the production of large amounts of salt soluble species derived from the metallic reducing agent or from the large amount of uranium in the spent fuel. These challenges have been addressed with the WATSS process, developed in the framework of a First-of-a-kind deployment at the Point-Lepreau nuclear site operated by New Brunswick Power for the recycling of spent fuel generated by the existing CANDU reactor.

3. The WATSS process

The combined WATSS and waste-burning fuel cycle can be divided into two recycling phases, corresponding to the so-called WATSS-C and WATSS-S processes (dependent on the feed material being CANDU or SSR-W spent fuel). Although WATSS is initially being developed to recycle spent CANDU fuel, it may also be adapted to recycle light-water reactor (LWR) spent fuel based on low-enriched uranium oxide or even irradiated fuel from other types of reactors in the form of nitride, carbide, silicide or metal.

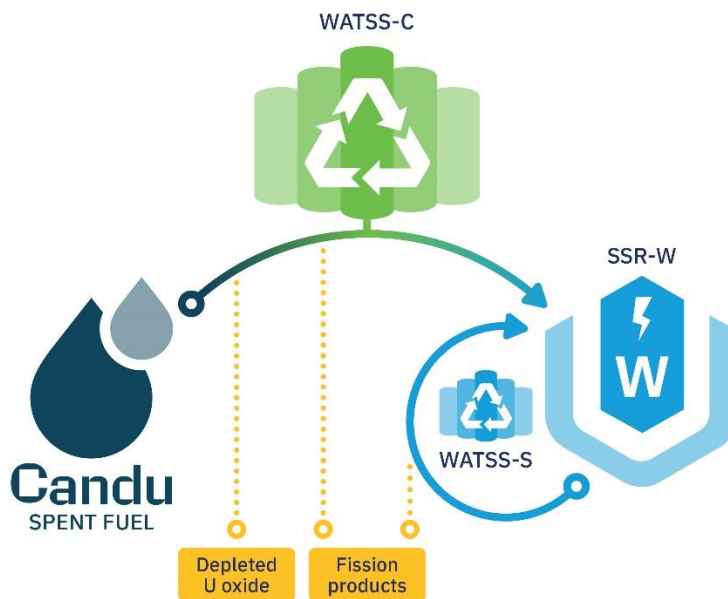


Figure 1 Schematic representation of WATSS / SSR-W fuel cycle (Adapted from [4]).

WATSS-C includes the de-cladding of spent fuel bundles and pre-treatment of the oxide matrix, the extraction of TRU species from uranium oxide, their concentration in precursors of spent fuel and the blending of spent fuel precursors to reach the desired composition.

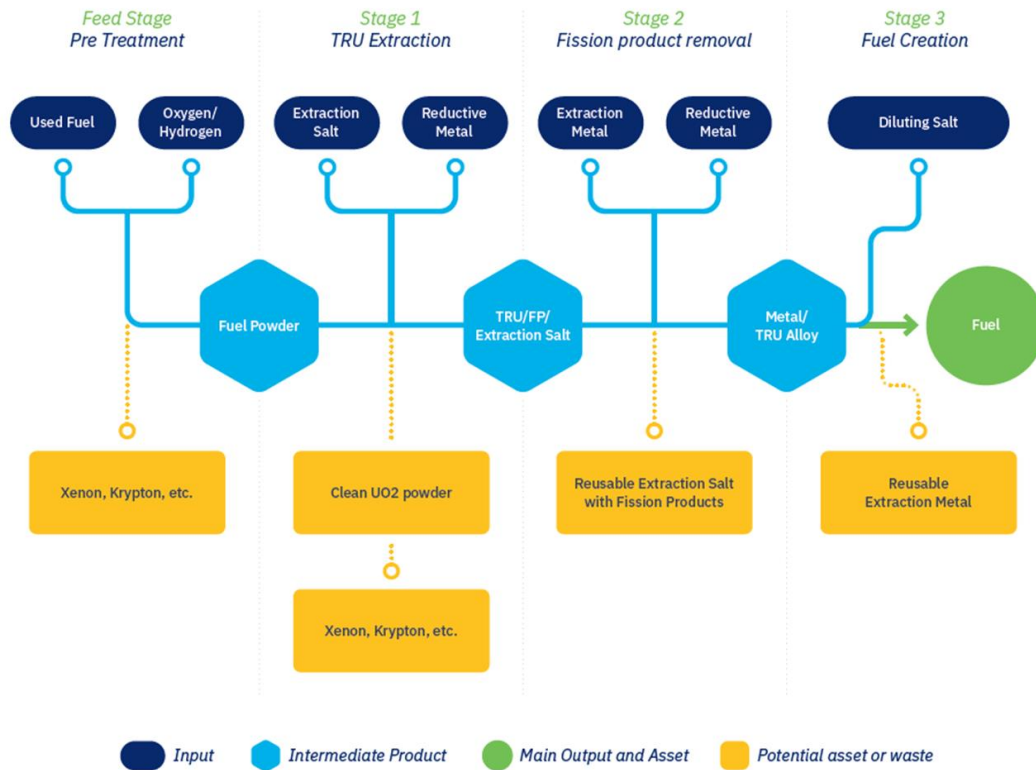


Figure 2 Schematic representation of the different process stages of WATSS.

WATSS-S is a simplified version of WATSS-C, in which pre-treatment and extraction from the oxide matrix are not required since the feed material is already in a salt form.

The different steps of the complete WATSS-C process, reported in Figure 2, are described in the following sections. The combined fuel cycle with SSR-W, as represented in Figure 1, is an example of the employment of WATSS. The process stages in Figure 2 can also be adapted to the production of fuel for other types of reactors. Using CANDU spent fuel as feed material is also an example, the process being compatible with spent oxide fuel from other types of reactors and even other spent fuel forms such as metal, nitride, carbide or silicide, as discussed in section 3.1.

3.1 Pre-treatment

After de-cladding of the irradiated spent fuel, the fuel pellets undergo voloxidation in air or oxygen. This process leaves the uranium in the U₃O₈ form and breaks the pellets into powder. TRU compounds, already at their highest oxidation state of +4, remain as dioxides. It has also been highlighted by computational modeling and a preliminary literature review [12-14] that a voloxidation treatment of uranium metal or different forms of nitride, carbide and silicide also produced U₃O₈, opening the route to the application of WATSS for these fuel types.

This pre-treatment can optionally be further extended by cyclic oxidation and reduction in a process known as OREOX (Oxidation-Reduction of Oxides) [15]. The application of multiple cycles of oxidation and reduction further reduces the size of the particles and leads to a finer powder in which the area-to-volume ratio is increased.

3.2 TRU extraction from the oxide matrix

The primary objective of the overall process is to effectively separate TRU species from uranium, which is present in overwhelming proportion in the oxide matrix. This stage, referred to as “Stage 1” in the overall WATSS process, is based on the direct extraction from powdered spent fuel into a molten salt in the presence of a reducing metal that would convert transuranic elements into salt-soluble species (trichloride or oxychloride), while leaving uranium as an insoluble dioxide. This is essentially based on the process identified by Wenz (see section 2), which Moltex has now revisited with alternative, cheaper, reducing reagents identified using modern chemical computational tools.

Outotek HSC v10 GEM module was used for all chemical thermodynamic calculations. The thermodynamic data used was from the built-in database. The input parameters were U_3O_8 (100 mol), PuO_2 (0.316 mol, the average proportion of Pu in CANDU spent fuel), a eutectic salt composition of NaCl (466 mol), KCl (580 mol) and $MgCl_2$ (656 mol), as well as the incremental addition of different reducing metals.

Simulations have also been performed with CeO_2 as a surrogate for PuO_2 , with varying ratios of salt to powder, the inclusion of fission product oxides and the combined addition of more than one reducing metal. The results leading to the most compelling separation between Pu and U species (Pu as trichloride or oxychloride, with U as dioxide) have been tested with simulated spent fuel (SimFuel), consisting of sintered UO_2 pellets containing cerium dioxide as a surrogate for PuO_2 .

A variety of extraction conditions have been investigated, with the most representative results reported in Figure 3, highlighting an extraction efficiency as salt-soluble species of about 90% of the cerium oxide originally present as dioxide, while the concentration of salt-soluble uranium species drops sharply as the reaction progresses. The chemical processes involved have been discussed previously [16,17].

To validate the extraction of plutonium species from real spent fuel, experiments have been conducted at the Chalk River facilities of the Canadian Nuclear Laboratories (CNL). Experimental procedures and analytical methods are described elsewhere [16,17].

The different experimental conditions, the extraction of plutonium species (as percentage of Pu converted to salt-soluble species) and of uranium are reported in Table 1. Preliminary extraction results obtained from CNL for the cases of magnesium and niobium as reducing metals have been reported in [16] and [17] respectively. The values in Table 1 are slightly corrected values from the CNL final report [18].

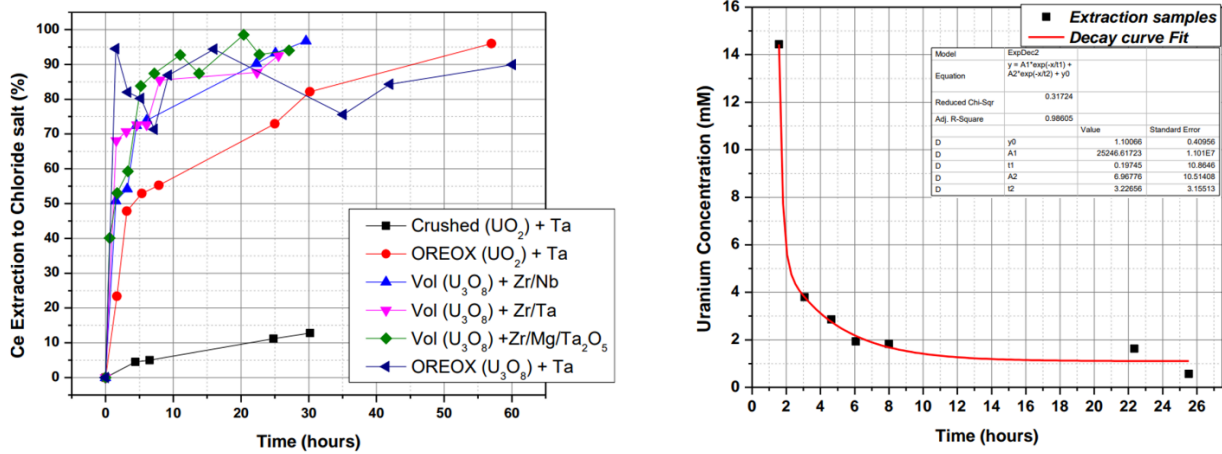


Figure 3 Left: Extraction efficiency from SimFuel, featuring different pre-treatment and addition of different identified reducing metals. Right: Evolution of the carry-over of salt-soluble uranium species during the extraction stage (data points correspond to Vol(U₃O₈) + Zr/Ta dataset). The sharp drop in uranium carry-over in the salt phase relates to the initial presence of the salt-soluble UO₂Cl when U₃O₈ and MgCl₂ are mixed, and its reduction as UO₂ in presence of a reducing metal [16,17].

Table 1 Extraction results from experiments conducted at CNL, with spent CANDU fuel unless specified otherwise. Reported uncertainties are for 2 standard deviation (95% confidence interval).

Entry	Pre-treatment / uranium oxide form	Reducing metal	Reaction time (h)	Pu extr. (%)	U extr. (%)	Pu/U ratio
1	Voloxidation / U ₃ O ₈	Ta + Zr	24	60.0 +/- 2.9	0.048	5.16
2	OREOX / U ₃ O ₈	Mg	24	76.4 +/- 3.7	0.95	0.33
3	OREOX / U ₃ O ₈	Zr	24	61.1 +/- 3.0	4.44	0.06
4	OREOX / U ₃ O ₈	Nb	24	87.4 +/- 4.2	0.050	7.27
5	OREOX / U ₃ O ₈	Nb	60	92.1 +/- 4.5	0.018	21.5
6	OREOX / U ₃ O ₈	Ta + Zr	24	47.7 +/- 2.3	0.34	0.58
7	OREOX / U ₃ O ₈	Ta + Zr	60	44.7 +/- 2.2	0.54	0.35
8	OREOX / U ₃ O ₈	Nb + Zr	24	35.8 +/- 1.7	0.017	8.99
9	OREOX / UO ₂	Ta	60	34.1 +/- 1.6	0.015	9.76
10	OREOX / (SimFuel) U ₃ O ₈	Ta + Zr	24	71.1 +/- 3.9 (Ce)	0.023	N/A

The Pu/U ratio in the extraction salt (last column of Table 1) relates to the relative extraction efficiencies of the elements analyzed and their initial abundance in the feed material.

Previous non-active work (Figure 3) and modelling had suggested that zirconium would be a suitable reducing metal and would enable use of the Zr cladding of the recycled fuel as a reagent in the recycling process. However, it appears that Zr struggled to extract above 60%. This was seen when used alone or in combination with another metal. The issue is thought to stem from the oxidation of the metal surface, which likely creates a kinetic barrier and limits access to fresh Zr for effective extraction. This phenomenon had been observed in some experiments with SimFuel, although the impact on extraction efficiencies was not as pronounced. The higher uranium carryover, especially when zirconium is used alone, suggests that not all of the higher-valent uranium was reduced, likely due to insufficient reductive power.

When the uranium matrix is in the UO₂ form, the PuO₂ extraction efficiency is significantly lower, highly likely due to the inability of uranium oxide to convert to an intermediary salt-soluble form (oxychloride), therefore limiting the molten salt access to Pu in the uranium oxide grains. This absence of a salt-soluble uranium oxychloride intermediary, as discussed in [16], also leads to a significantly low carry-over of uranium at the end of the extraction process.

Overall, the most suitable reducing metal candidate is niobium as shown by Entries 4 & 5 in Table 1. After 24 hours, there was an extraction of almost 90% of the Pu in the oxide matrix, 10% higher than the next nearest result (Entry 2). Allowing the reaction to continue for an additional 36 hours, gave an extraction exceeding 90%. At both time points, minimal carryover of U was found.

As mentioned above, magnesium (Entry 2) showed 76% extraction of Pu. This reducing metal produces no by-product as the molten salt already contains MgCl₂. Further optimisation would be required to keep the carryover of uranium in the salt relatively low, but this option also represents a promising route.

3.3 TRU concentration

The following step in the process is to concentrate the extracted TRUs. This is done in two successive stages, named Stages 2 and 3 respectively.

In Stage 2, the molten chloride salt, containing the TRUs and the fission products that have formed chloride and oxychloride forms, is brought into contact with a liquid metal carrier and a stronger reducing metal than in stage 1, to form a liquid alloy of actinides with the carrier metal, while largely leaving fission products in the salt. The process has been used since the 1960s in the nuclear industry for spent fuel analysis and processing. It has been used for both molten fluoride and chloride salts using an array of different reducing metals [19-21]. Work by Kinoshita [22] has shown the use of Bi-Li alloy to introduce their reducing metal to the reaction mixture. Li would not be appropriate for the WATSS process as the Li metal would preferentially react with MgCl₂ in the eutectic salt and change the composition. As a result, Mg has been identified as a more promising reducing metal for this stage of the process. Molten bismuth is considered as a suitable alloying metal for this purpose, although other alternatives can be considered [21].

The purpose of Stage 3 is to halogenate the TRUs from the molten metal alloy to a molten salt phase, leading to a salt more concentrated in actinides than the output of Stage 1, and largely exempt of fission products. This is achieved through an exchange reaction by addition of an oxidising salt, for

instance the chloride salt of the carrier metal (BiCl_3 in the case of the use of bismuth as alloying metal). A dilutant salt can be added to the system without affecting the chemistry as long as it is consistent with the type of halide used. The selection of the preferred salt will be based on the SSR-W reactor and fuel salt design, as discussed in the following section.

The isotopic composition of the actinides is determined by the composition of the spent fuel used as feed material, and the ratio of U/TRU largely depends on the extraction efficiency from Stage 1. To control the reactivity of the fuel salt, different compositions of the product of Stage 3 (with specific compositions from batch to batch) will be combined and the dilutant salt will be adjusted to reach a eutectic composition. This is described in the “Production of Fuel Salt” part of the process. In preparation for this last stage, the composition of the product of each batch following Stage 3 will be analysed and the product, considered as a “fuel salt precursor” kept in storage until this specific precursor is determined to be adequate for blending with another identified precursor.

Alternative options for stages 2 and 3 may also be considered for the production of fuel in chemical forms different than chloride salts, depending on the type of reactor in which the recycled TRUs are expected to be exploited.

3.4 Production of Fuel Salt

The general fuel salt composition for SSR-W is $\text{NaCl-MgCl}_2\text{-XCl}_3$, where X is formed of uranium (U), transuranics (TRU) and potentially some remaining lanthanides (Ln), as highlighted in Figure 4 [4]. The use of MgCl_2 allows the fuel reactivity to be controlled via either the ratio of MgCl_2 to XCl_3 or by the ratio of U/TRU, either of which can be varied as part of the blending stage at the end of WATSS. This gives a high degree of flexibility.

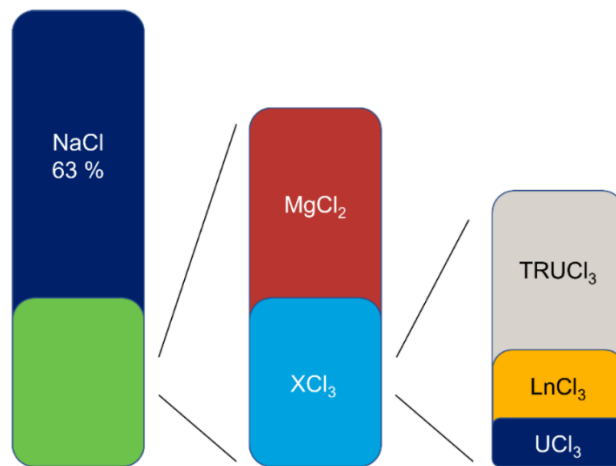


Figure 4 Components of the fuel salt for the SSR-W reactor (Reproduced from [4])

The phase diagrams of the $\text{NaCl-MgCl}_2\text{-UCl}_3\text{-PuCl}_3$ system indicate that the eutectic region determines the proportion of the monovalent component, with the freedom to vary the proportions of MgCl_2 and trichloride components over wide ranges without significant changes in properties. After blending the chosen salt precursors to obtain the expected composition of actinides, the eutectic point will be reached by the addition of adequate amounts of inactive salt components.

4. Economic considerations

An economic feasibility assessment was carried out for the First Of A Kind (FOAK) demonstration WATSS facility [23]. Using an independent cost estimate carried out by DBD International Ltd [24], a UK firm with specialist expertise in nuclear process facilities, and the publicly available cost estimate for pyroprocessing facilities produced by Argonne National Laboratories [25], the feasibility assessment produced a capital cost of C\$870m, based on a WATSS facility scaled to produce the first core load for one 300 MWe SSR-W in 17 months of operation. The associated annual operating costs at this capacity are estimated at C\$84m. When combined with the SSR-W capital and operating costs, the overall Levelized Cost Of Electricity (LCOE) is estimated at C\$143/MWh for the FOAK. Various scenarios were modelled with double construction cost, time, lower efficiencies, cost of capital etc, the highest of which was \$157/MWh. If the FOAK reactor is uprated to 500 MWe after 10 years of operation (as it is designed to do), and the WATSS facility runs at full capacity recycling used fuel from different sites, a very competitive LCOE of C\$87/MWh is forecast.

The economic feasibility study demonstrated that these numbers were viable if an extraction rate of over 75% was achieved. Given the significantly higher extraction rates highlighted, the economics are expected to improve.

As the prices of uranium and rare earth elements continue to rise, the economic rationale for recycling used nuclear fuel strengthens proportionally. When combined with the strategic benefits of enhanced energy security and the capability to destroy virtually all long-lived actinides, such as plutonium, the case for adopting fuel recycling becomes increasingly compelling.

Further details on these costs are available in the white paper on the economics of the Moltex WATSS process [26].

5. Conclusion

The WATSS process consists of a series of treatment stages turning spent fuel, previously considered as a liability, into fresh fuel for a fast spectrum wasteburner reactor in a comprehensive, commercially viable, recycling process leading towards the closure of the fuel cycle.

The central stage of the process has recently been validated through extraction experiments carried out at the CNL facilities, while the other stages of WATSS largely rely on principles that have already been demonstrated in different applications.

6. Glossary of acronyms

CANDU	CANadian Deuterium Uranium reactor
CNL	Canadian Nuclear Laboratories
FOAK	First Of A Kind
LCOE	Levelized Cost Of Electricity
LWR	Light Water Reactor
OREOX	Oxidation-Reduction of Oxides
PUREX	Plutonium-Uranium Redox EXtraction
SSR-W	Stable Salt Reactor - Wasteburner
TRU	TRansUranic elements
WATSS	WAsTe To Stable Salt process

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