The CNRS Research Program on the Thorium cycle and the Molten Salt Reactors

Contributors[1]
Thierry Auger[1], Gérard Barreau[2], Jean-Pierre Chevalier[3], Xavier Doligez[4], Sylvie Delpech[5], Hubert Flocard[6], Bernard Haas[2], Daniel Heuer[4], Elsa Merle-Lucotte[4].

1) CNRS, Ecole Centrale Paris, laboratoire MSSMAT, UMR 8579
2) CNRS/IN2P3, Université de Bordeaux, CENBG, UMR 5797
3) Conservatoire National des Arts et Métiers
4) CNRS/IN2P3, Université Joseph Fourier, Institut Polytechnique de Grenoble, LPSC, UMR 5821
5) CNRS, Ecole Nationale Supérieure de Chimie de Paris, LECA, UMR7575
6) CNRS/IN2P3, Université Paris-Sud, CSNSM, UMR 8609

[1] The file of this document is available at: flocard@csnsm.in2p3.fr

Summary

In order to build a sustainable future for nuclear energy production, the international community assembled within GEN-IV has identified six classes of innovative systems deemed worth a thorough investigation with a view to a future implementation. The molten salt reactor (MSR) has been selected among them. France is one of the countries which have expressed an interest for this option.

Sustainable nuclear fission is established on two pillars: breeding and recycling. Together they ensure a long-term stable (or if needed, expanding) energy production compatible with the resources available on the planet. In addition, another major goal of recycling (also referred to, as "closed cycle" in contrast with the "open cycle" of present LWR[2]s), is to achieve a substantial decrease of the radiotoxicity of the waste stream via incineration. The physical phenomenon on which incineration is based is neutron induced fission. For a large class of radio-nuclides this specific type of nuclear reaction requires fast neutrons[3].

As a consequence, all closed cycle GEN-IV systems are "fast" reactors. Official and technical presentations give SFR, GFR, LFR and (sometimes) SWR as GEN-IV examples of fast systems[4]. Beyond the requirements of the incineration mission, a major reason why, in order to be considered "sustainable", these four systems have to be fast is well known: they operate with the uranium-plutonium (U-Pu) cycle whose neutron economy only allows breeding when the neutron spectrum is not moderated.

Customarily, public presentations do not include Molten Salt Reactors in the list of GEN-IV closed cycle breeders. It appears that this omission is more grounded in technical history than justified by Physics. In general, MSRs are studied in relation with the thorium-uranium (Th-U) cycle whose neutron economy allows breeding with slow neutrons. All major MSR realizations (ARE, MSRE[5]) and projects (MSBR[6]) of the past, especially those conducted by ORNL[7], have been reactors working in the thermal regime. As said above, this characteristic forbids the incineration of several actinides present in the spent fuel of LWRs. Thus, these reactors do not fulfil the transmutation mission of the present sustainability agenda. Of course, in the years 1960-80, this was not considered to be a liability nor even a weakness.

As a matter of fact, the physics of the Th-U cycle is more flexible than that of the U-Pu cycle: it certainly allows breeding in the thermal regime but also in the fast regime. This opens the possibility to an investigation of non moderated MSR reactors which can accomplish the two missions which define sustainability, namely breeding and incineration of all actinide isotopes. This is "the road not taken" that the CNRS[8] teams have chosen to explore and which has led them to a novel concept: the TMSR-NM[9].

This document presents an overview of the activities and results obtained so far by CNRS groups on the TMSR-NM. It aims to show that this concept is both promising and versatile. TMSR-NM can be started with the plutonium and minor actinides from the waste of GEN-III reactors. It will incinerate them while effecting a gradual transition to the Th-U cycle opening a usage of the vast natural thorium resources. In all modes of operation, it allows breeding at a rate compatible with the growth of the energy needs foreseen for many developed and developing economies. The radiotoxicity of its waste stream can be made as small if not smaller than that all other closed cycle GEN-IV systems. Moreover, the same concept with the same basic design can be adapted to destroy its own radiotoxic inventory should a decision to stop nuclear fission energy production be taken in a still undecided future.

Of course to go from a concept such as TMSR-NM to a practical reactor design, much research is needed at both basic and applied level on a vast array of questions pertaining to physical, chemical and material sciences. Yet, the present situation on these issues already looks promising. The modest on-line recycling fluxes required by the TMSR-NM nuclear fuel as well as the chemistry needed for this operation appear to be within reach of present technology. For the fuel reprocessing

[2] Light Water Reactor

[3] In this context, fast corresponds to neutrons whose energy lies between few hundred keV and few MeV.

[4] SFR stands for Sodium Fast Reactor, GFR for Gas Fast Reactor, LFR for Lead Fast Reactor and SWR for Supercritical Water Reactor.

[5] ARE stands for Aircraft Reactor Experiment, and MSRE for Molten Salt Reactor Experiment

[6] Molten Salt Breeder Reactor

[7] Oak Ridge National Laboratory

[8] Centre National de la Recherche Scientifique. In the present document, for the sake of concision, everywhere "CNRS" stands for "CNRS and associated French Universities"

[9] Non Moderated Thorium Molten Salt Reactor.

which, more than for any other GEN-IV reactor, is integrated into the very operation of MSRs, we are proposing a preliminary flow-sheet. In turn, this flow-sheet defines specific research items in each of its sub-blocks. These are progressively addressed within the CNRS programme.

As for material science, here also one does not start from a blank sheet. When ORNL closed its research on the subject, it had defined many credible solutions (performant Ni-based alloys for instance). It had tested some of them in representative temperature and irradiation conditions. Since then, major advances in material sciences have further improved the situation. CNRS teams collaborating with industry are presently building on this core knowledge and have launched work on innovative alloys.

In order to define a work agenda compatible with material and human resources available in basic and applied chemical and material sciences, it is necessary to circumscribe the domain encompassed by intensive physical parameters (temperatures, flow velocities, pressure gradients, …) which potentially govern the operation of a TMSR-NM. This can only be achieved on the basis of a reactor design which guides the determination of the boundaries of the relevant multi-parametric subspace. For CNRS, this reactor design stands therefore presently mostly as a tool to plan and organize its basic research. In addition, it introduces a much needed thermal-hydraulics component into the work program. CNRS teams are presently realizing numerical simulations preparatory to the making of a design. Simultaneously, they have started the construction of a forced convection loop which, along tests of material resistance in presence of molten salt, will investigate the physics of transport, corrosion, abrasion and thermal exchanges with a liquid salt.

Within the framework defined by a law voted by the French parliament on June 28 2006, the CNRS has setup a global research programme PACEN (Programme sur l'Aval du Cycle et l'Energie Nucléaire). PACEN organizes and support the activities of CNRS and French University groups on all questions related to the backend of the nuclear electricity production and on the transition towards sustainable nuclear energy. This document presents the status as of June 2008 of the research conducted on a) the thorium cycle[10] and b) the molten salt reactors by the CNRS teams. It outlines the main lines of work identified by the academic community for the coming years within the overall exploratory mission assigned to this research.

The CNRS activities on the thorium cycle and molten salt reactors have been financed by the "Programme Concerté de Recherches sur les Réacteurs à Sels Fondus" (PCR-RSF) of PACEN. An additional contribution has come from the PACEN research group GEDEPEON[11] which is jointly operated by CNRS, CEA, EDF and AREVA-NP. The FP6 Euratom programme ALISIA has provided support for international contacts and collaborations.

[10] CNRS also supports other Th-cycle related activities in particular involving solid fuel (ThO2). They concern dedicated systems and scenarios as well as basic data relevant for aqueous roprcessing. A note describing these activities is forthcoming.

[11] GEDEPEON is the acronym of « GEstion des DEchets et Production d'Energie par des Options Nouvelles »

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I Introduction

In the third quarter of the last century, in parallel with the Sodium Fast Reactor (SFR), the Molten Salt Reactor (MSR) was for some time considered as an alternative route towards breeding. Following the encouraging results obtained by the MSR-Experiment (MSRE) [Hen70], the Oak Ridge National Laboratory (ORNL) launched the design of a 1GWe system: the Molten Salt Breeder Reactor (MSBR) [Bro70, WHa70].

This particular project relied on the Thorium-Uranium (Th-U) fuel cycle which in principle allows nuclear energy production, breeding and, as compared to SFR working in the Uranium-Plutonium (U-Pu) cycle a reduced production of the most heavily radiotoxic minor actinides. On the other hand, MSRs have some additional assets which are not tied to the Th-U cycle. The liquid fuel does not have to be kept under high pressure even at high temperature. It is also rather insensitive to irradiation. With a MSR, one circumvents the sometimes arduous problem of solid fuel fabrication (whether in heterogeneous or homogeneous mode) when minor actinides are present. Moreover, the liquid fuel allows a continuous tuning of core reactivity. The amounts of fissile and fertile matter can be adjusted without unloading the core, doing away with the need for any initial reactivity reserve.

Here we discuss a MSR design which can be adapted to various actinide fuels and which allows a smooth transition from the U-Pu cycle to the Th-U cycle while destroying most of the actinide waste of the GEN-III reactors. MSRs also offer interesting options for reducing their own inventory. Finally, as compared to solid fuel reactors, MSRs whose thermal and void coefficients are negative, whose fuel is transparent, allowing in service inspection and whose processing is performed in the near vicinity of the reactor also have distinctive features with respect to both safety and proliferation.

Although the smaller potential breeding rate of the Th-U cycle, as compared to that of the U-Pu cycle in a SFR, most certainly played an important role in the decision to abandon MSBR, at that time, it was also recognized that MSRs, when operated in the thermal regime, had still not reached sufficient maturity on crucial points such as safety procedures, material resistance and fuel processing. This forbade envisioning the rapid transition towards industrialization which was then considered necessary. Following the decision to stop the MSBR project at Oak-Ridge, all major R&D programmes on this type of reactor came more or less to a halt.

However, despite the weaknesses of earlier concepts, in view of MSR intrinsic interest as a nuclear energy production system, of the major recent advances in material science, of the overall reassessment of the availability of the Uranium resource, of the present state of uncertainty on the moment when breeding will become a necessity and of the strongly modified perception by the public of the danger associated with nuclear waste, the Generation-4 forum (GEN-IV) has decided to retain the MSR as a long term option among the six candidate systems for the new generation of reactors aiming at a sustainable nuclear energy production.

It is in this context, that the CNRS within PACEN has setup the PCR-RSF action which, building on the important legacy of past research at ORNL, aims to revisit the MSR concept, to provide novel perspectives and to produce original results on all the still open major related issues. The CNRS MSR also relies on competences from another PACEN action: GEDEPEON. For a research institution like CNRS, involved in both basic and applied research but not in industrial development, these activities retain the character of an open exploration of the basic questions associated with MSRs. On the other hand, we want to address energy production by MSRs in comprehensive manner, ensuring even paced advances on all issues relating to this type of reactor. The CNRS programme is defined in coherence with the research directions specified by the GEN-IV “System Research Plan” for the MSR option.

The present document has a hybrid character. It both summarizes the status of the CNRS work program and outlines the research planned for the short and medium terms. In Sect. II, we give our motivation for an experimental program on Th-U cycle nuclear data and present the results so far obtained as well as some forthcoming experiments. Sect. III presents an innovative concept proposed by CNRS (a non moderated MSR; the Thorium Molten Salt Reactor or TMSR-NM) and shows that it avoids many of the shortcomings of the MSBR design while opening new perspectives especially with respect to GEN-III-reactor waste destruction. Sect. IV gives the status of the reactor design and outlines the thermal hydraulics research program. Sect. V is devoted to fuel salt reprocessing. In Sect. VI, we indicate which lines of research in material science are pursued by CNRS. Sect. VII analyzes the radiotoxicity of the nuclear waste. Sects. VIII and IX present studies done or planned on safety issues on ^{233}U breeding and on TMSR-NM deployment rate.

II Thorium-Uranium Cycle Nuclear Data

Figure 1 presents the most important reaction and radioactive decay channels of the thorium cycle. In the energy range relevant for nuclear energy the reactions of interest are neutron capture (n, γ), neutron induced fission (n,f), elastic (n,n) and inelastic (n,n') scattering and neutron emission (n, 2n). In order to enable simulations to predict reactor properties or breeding potential with good accuracy, the cross sections associated with the most important reactions must be known with a precision of the order of few % (5-10%). The most important decay channels in this cycle are electron emission (β^-) and gamma decay (γ) in the last stages of the decay chains.

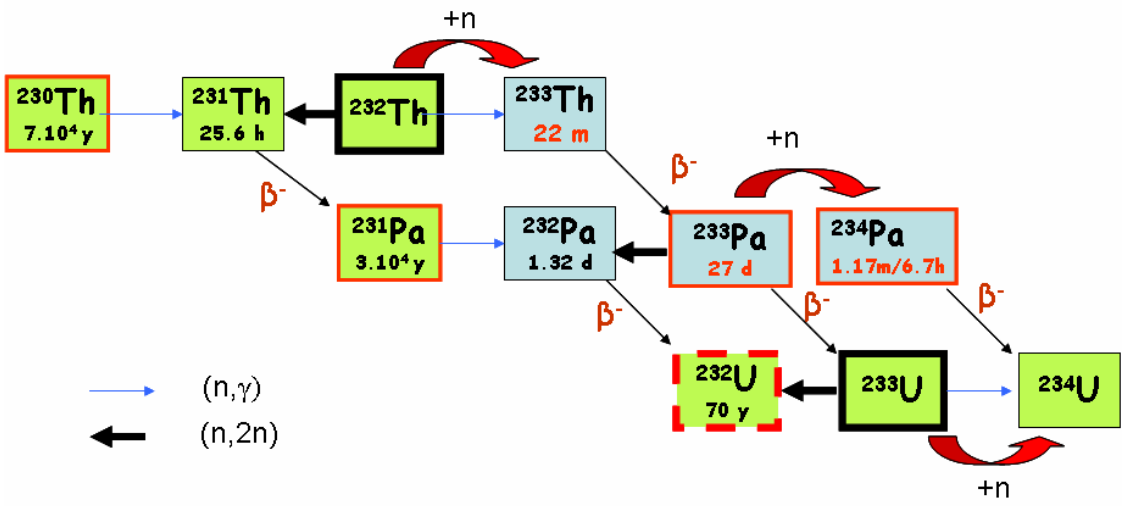

Fig1: Key nuclei of the Th-U cycle. The red arrows correspond to neutron capture processes. In particular, capture on ^{233}U, the fuel of the Th-U cycle, ultimately leads to fission which generates the energy production. On the other hand, capture on the fertile nucleus ^{232}Th is the basis of breeding. Two successive beta decays lead to the formation of ^{233}U.

The past lack of interest for the Th-U cycle explains why present nuclear data files which rely on models and/or extrapolations from odd sets of data points (when they exist for neighbouring nuclei) need to be revisited. Within the European network EFNUDAT, CNRS and CEA teams have started a comprehensive set of measurements on the most important isotopes (^{232}Th, ^{231}Pa, ^{233}Pa, ^{233}U and ^{234}U).

A comparison of the various data tables (ENDF, JENDL, JEFF, …) on the initial capture reaction ^{232}Th(n, γ) as they stood at the end of last century has showed that discrepancies between them were sometimes of the order of 20-30% for neutron energies above 50keV. This has motivated the organization of European collaboration including CNRS/IN2P3 laboratories to revisit this reaction in the energy range [60keV, 2MeV]. With two independent methods for measuring the neutron flux produced by means of the Bordeaux accelerator, a total a 20 new data points has been accurately measured. This work [Kar01] has vindicated the predictions of the JENDL-3 (Japan) data table in contrast with those of the JEFF-2 (Europe) table. The updated version JEFF-3.2 now takes the new data into account.

The nucleus ^{233}Pa plays a central role (analogous to that of ^{239}Np in the U-Pu cycle). It acts as a precursor to the long lived nucleus fissile ^{233}U. The high n-induced fission rate of this latter isotope (92%) is the attractive feature of the Th-U cycle, essentially because it hinders the formation of higher actinides (Np, Pu, Am, Cm…).

The half life of ^{233}Pa is rather long (27d) opening the possibility of a neutron capture in the reactor neutron flux. This is the so-called protactinium effect. It particularly affects reactors operated with solid fertile fuel (ThO_2) (lower reactivity at the start, higher reactivity after stop) but should be avoidable in a MSR whose reactivity can in principle be controlled on line. No reliable data was available for the cross section of this reaction while evaluated data tables could at some energies differ by almost a factor two. Indeed a direct measurement of a neutron induced reaction on this nucleus is extremely difficult because of the high level of radioactivity of a potential ^{233}Pa target (several 10^9Bq/μg). The CNRS-Bordeaux group has thus resorted to the so-called “surrogate” method. In this indirect method the cross-section is extracted by means of a transfer reaction ^{232}Th(^{3}He,p)^{234}Pa leading to the same

compound nucleus ^{234}Pa whose fission and gamma decay can then be investigated. With this method, the Bordeaux group has obtained extensive data for the capture [Boy06] and fission [Pet04] of ^{233}Pa. As shown in Fig.2, the new fission data ("this work") compare well with the few points obtained by a direct experiment at the Geel JRC neutron facility. In contrast, the new measurements differ from both evaluated data files ENDF/B-VI and JENDL-3 which will have thus to be corrected.

Breeding within the U-Pu or Th-U cycles differ in two respects. First, the Th-U cycle allows breeding both in the thermal or non moderated regime while only the fast neutron regime is effective for the U-Pu cycle. Second, the neutron economy of the Th-U cycle is more constrained. As a consequence, an accurate knowledge of the ratio between capture and fission cross sections (the so-called α parameter) of the fissile nucleus ^{233}U averaged over the neutron energy spectrum of the reactor is important to evaluate the potential of the TMSR reactors. The present situation is not satisfactory. This ratio, whose value is about 0.2, is only known from old (1960-70) data which differ by about 25% among themselves.

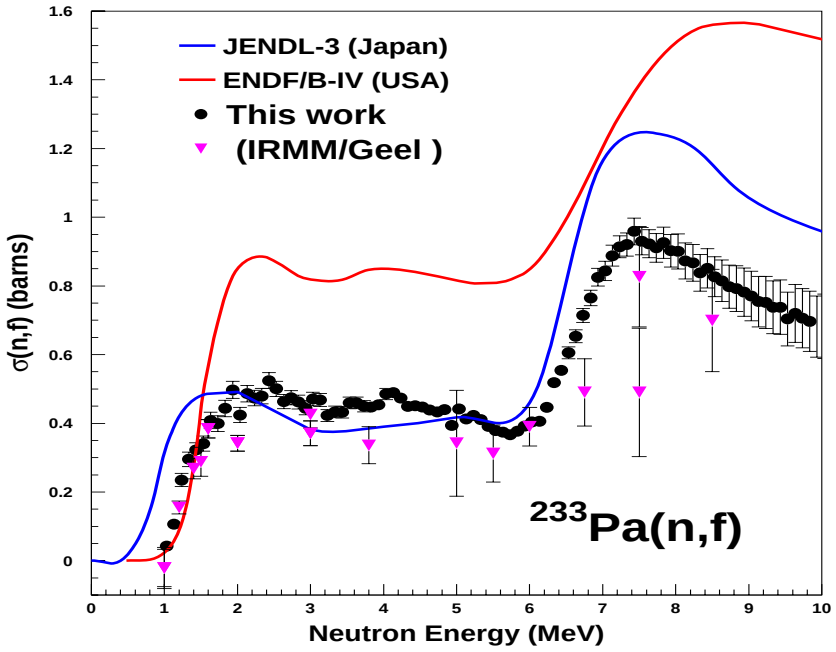

Fig.2. ^{233}Pa fission cross sections for neutron energies ranging between 0 and 10MeV. Comparison of recent CNRS data (black points) with direct measurements (pink triangles) and with two evaluated cross sections in the JENDL (blue curve) and ENDF-B-VI (red curve) data tables.

The CNRS has committed itself to perform a series of experiments dedicated to the simultaneous measurement of fission and capture cross sections (thereby eliminating uncertainties associated with approximate knowledge of the neutron flux) for energies either in the resonance domain (0.1eV to 10keV) or fast neutrons (0.1 to 2MeV). Experiments will be conducted both at Geel, at Grenoble and at the platform AIFIRA in Bordeaux.

III. The non-moderated thorium molten salt reactor concept

As a breeding system, despite its remarkable compactness (as compared to Na cooled fast neutron reactors), mostly because it works in a thermal regime, the Oak Ridge MSBR concept suffers from several weaknesses. For instance, its temperature and void coefficients are positive. This is certainly an undesirable feature from a safety point of view. It also requires an unrealistically large chemical processing unit in order to efficiently eliminate neutron poisons from the salt. Finally, it uses a large mass of graphite as a moderator. It turns out that the life expectancy of the MSBR graphite structure with respect to neutron irradiation is restricted to only few years. French experience with the graphite cores of the decommissioned UNGG reactors shows that managing such a waste is not an easy problem. In that light, it appears interesting to extend the analysis of the MSRs concept beyond that of Oak Ridge in order to devise ways which could circumvent these difficulties.

This task has been performed at CNRS-Grenoble by a set of systematic multi-parametric simulations correlating the geometry and material composition of the core, the reprocessing

performances and the salt composition [Mat05, MHe05, MHe06]. This set covers all configurations from a moderated system similar to that investigated and tested at Oak-Ridge to epithermal and fast systems. The simulations rely on the MCNP neutron transport code [Bri97] coupled with REM: a home-made material evolution code [Nut02]. The former evaluates the neutron flux and the reaction rates within all the cells of the MCNP simulation while the latter solves the Bateman equations for the evolution of the material composition within each cell. The calculations use an accurate description of the geometry and follow the interactions and the radioactive decays of several hundreds nuclei. This work takes into account the input parameters (total power and power density, criticality level, chemistry ...), by a continuous adjustment of the neutron flux and of the material composition of the core structures and of the fuel.

The calculations do indeed reproduce the above mentioned damage level sustained by the carbon core of a moderated thermal MSR such as MSBR. In particular it appears that when graphite is present within the reactor vessel, either irradiation damages are prohibitive or the thermal and void safety coefficients are positive. This led to consider a MSR concept with no moderator: the TMSR-NM. Note however, that, due to some moderation by the salt, the neutron spectrum is not as fast as in a Na or Pb, or He cooled U-Pu reactor. As shown below (Sects. III.C & E), the TMSR-NM seems to be promising both in terms of processing requirements and safety coefficients. The absence of moderation also gives TMSR-NM the capacity to burn most actinides whatever their fission threshold. Section VIII will present its breeding and deployment capabilities. In addition, the simplicity of the core design should translate into reduced irradiation damages on structure materials.

III.A TMSR-NM Core Description

To specify the TMSR-NM concept [HMM06, MHA07, MHA07a, FRL67, MHL07b, MHL06], we consider a 2500 MWth (or 1GWe assuming a 40% efficiency) reactor which when used in energy production mode uses Thorium as fertile fuel and blanket. As initial fissile fuel it can use either ^{233}U or a mix of transuranic elements dominated by Pu. As schematized in Fig. 3, the core is a large empty cylinder (1.25m radius and 2.60m height) which contains 14m^3 of moving fuel salt and a 10m^3 fertile blanket shaped in a 40cm wide annular ring. Most nuclear fissions occur within the cylinder in the up-flowing salt. At any moment, about one third of the 20 m^3 total of fuel carrying salt is outside the core running through pipes, pumps, heat exchangers and in the salt processing system which remove the gaseous and insoluble fission products. Apart from gradients induced by the forced circulation of the salt, the pressure is close to 1bar.

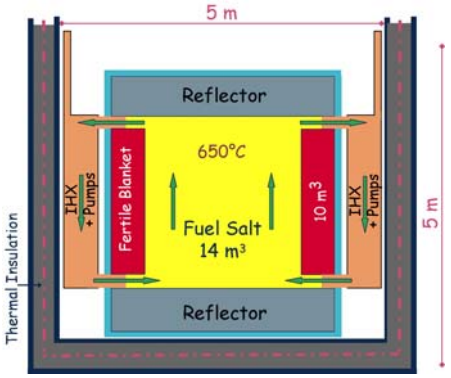

Fig.3. Principle sketch of the vertical section of the TMSR-NM, including pumps and heat exchangers (IHX)

The reactor vessel is protected by reflectors which absorb 80% of the neutron flux. To avoid a thermalization of the reflected neutrons, the axial reflectors are made of NiWCr (Ni-7% W- 6% Cr)[Cur07]. The radial reflector is also the fertile blanket. It consists in a binary fluoride salt LiF-ThF_4 with a 28% concentration[12] of ^{232}Th enclosed in a hollow cylindrical NiWCr structure.

III.B Salt Composition

The proportion of heavy nuclei (HN) within the salt determines the moderation within the reactor core. The small-mass isotopes in the salt affect the neutron energy spectrum and therefore the TMSR-NM actinide burning capacity. Which salt is chosen also has an impact on the fuel inventory because it affects how much heavy nuclei can be dissolved. Finally the chemical nature of the salt constrains

[12] In this document, concentrations for salts and alloys refer to the molar composition.

both operation and safety by imposing temperatures as well as density and dilatation coefficients which in turn determine the thermal and void coefficients.

For values of HN concentration ranging from 20% to 30%, we presently consider as reference salt the binary LiF-ThF_4 whose melting point is close to 570°C. It thus allows a reactor operation at 630°C. This is a significant departure from the salt proposed for MSBR which contains a sizeable fraction of BeF_2. The evolution of safety regulation over the last decades induces to reduce or eliminate Be as much as possible. For smaller proportions of HN, either the operating temperature must be increased or another fluoride must be added in order to lower the eutectic point temperature. Other molecules (CaF_2 for example) are presently under study. The salt density used in our simulations ranges from 3.1 to 4.6 according to the HN proportion. A typical value for the dilatation coefficient is 10^{-3}/°C.

III.C TMSR-NM fuel salt processing requirements

Fuel processing and adjustment of the salt composition (redox potential measurement, reactivity…) are necessary for control of the operation of a MSR reactor. In this section, we only describe processing in so far as it concerns reactor operation deferring a more detailed discussion of chemistry issues to Sect.V.

Fuel processing has two goals. First it eliminates neutron poisons (fission products) which are either harmful to the nuclear chain reaction or to structure materials (corrosion). Second it recovers fissile material mostly in the form of ^{233}U but also higher actinides when operated as a waste burner.

The present view is that processing should be performed simultaneously in two ways. The first consists in a He-bubbling within the reactor-heat-exchanger primary salt loop which extracts all gaseous fission products (FP). Data by ORNL, has shown that bubbling can also eliminate some of the noble metals, non soluble fission products as well as a fraction of tritium. The reactor simulation presented here assumes that this helium bubbling removes half of noble gases and metals present in the salt at a given time over the next 30 seconds. In fact, we have checked that a less efficient He-bubbling extraction efficiency would have little effect on the TMSR-NM properties; indeed, up to extraction times of the order of a few days, the breeding ratio is almost unaffected.

On the other hand, the extraction of the other fission products, mainly lanthanides, is effected in batch mode. A small fraction of the salt is regularly set aside to be processed off-line by a chemical unit. The outcome is a salt cleaned from almost all FPs in which the fissile matter (mostly uranium) is sent back to the reactor core. In Sect. V we present in some details the three stages of this batch process. The amount of salt which is processed every day determines both the size of the chemical unit and the breeding efficiency of the TMSR-NM. The smaller this amount, the less neutron poisons are extracted and the more manageable the processing becomes. On the other hand the presence of a larger concentration in the reactor salt of remaining FP decreases the breeding efficiency. Figure 4 shows the relation between the daily capacity of the batch process and the breeding performance of the TMSR-NM measured in terms of the so-called doubling time (i.e. the time over which one reactor will generate enough ^{233}U to allow the start of another TMSR-NM). The results discussed here only consider reprocessing rates within a technologically accessible range of reprocessed heavy nuclei. Typical values stay within 50kg HN/day to 200 kg HN/day, values which translates into few tens liter per day. As a comparison the Oak Ridge MSBR concept required that over 4000l be reprocessed every day. The processing requirement for a TMSR-NM is thus close to two orders of magnitude smaller.

III.D Initial fissile fuel inventory

III.D.1 ^{233}U

The ^{233}U initial inventory ranges from 2550 kg for a 7.5% HN proportion to 6180 kg for a 27.5% HN proportion (see Table 1). This corresponds to an evolution of the neutron spectrum from epithermal to fast.

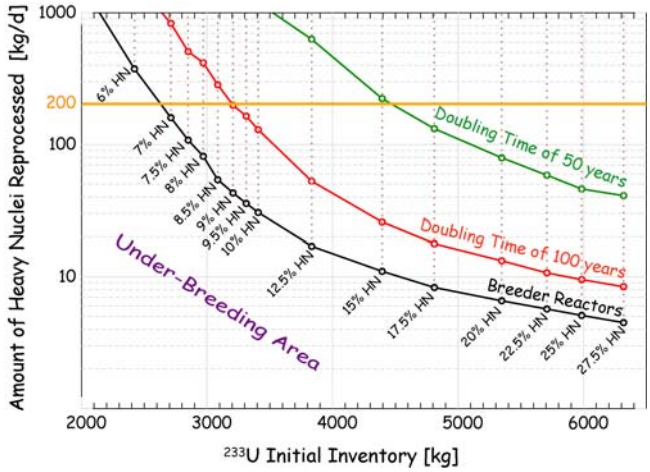

Fig.4. Reprocessing capacity requirement for a TMSR-NM system operated in the Th-U cycle. The ordinate gives the weight of heavy nuclei reprocessed per day while the abscissa corresponds to the initial fissile (^{233}U) inventory. Heavy nuclei proportions in the MSR fuel are indicated (x % HN as in Table 1). The black line separates breeding and non-breeding zones. In the breeding zone, the red and green lines indicate the reprocessing requirements which allow the generation of the ^{233}U needed for a new TMSR-NM reactor with the same inventory over 100y and 50y respectively.

III.D.2 Transuranic Elements

The TMSR-NM presented in section III.a can also be started with an initial actinide load consisting in thorium and in a mix of TRU elements (Pu, Np, Am and Cm) as they exist in the used fuel of the water moderated reactors presently operated with either natural or slightly enriched uranium. For instance, the simulation discussed below assumes a TMSR-NM started with the TRU contents of a PWR UOX fuel for 60GWd/T GEN-III reactor after five years of storage [Pu 87.5% (^{238}Pu 2.7%, ^{239}Pu 45.9%, ^{240}Pu 21.5%, ^{241}Pu 10.7%, ^{242}Pu 6.7%), Np 6.3% , Am 5.3%, Cm 0.9%].

For a typical TMSR-NM configuration with 22.5% of HN in the salt, an amount of 8400 kg of fissile elements is needed initially, corresponding to approximately 4% of Plutonium (See Table 1). This is very similar to the amount of fissile matter needed to start a SFR.

HN proportion	^{233}U-started TMSR-NM		TRU-started TMSR-NM	
	Th	^{233}U	Th	^{239}Pu+^{241}Pu
7.5%	19760	2550	13850	4887
10%	24050	3105	17560	5524
12.5%	27790	3575	20890	6022
15%	33060	4140	25380	6786
17.5%	37230	4650	29150	7297
20%	42380	5170	33640	7968
22.5%	46100	5580	37040	8378
25%	48640	5820	39320	8668
27.5%	52190	6180	42570	9037

Table 1. Initial inventories (kilograms) of Th and fissile matter for ^{233}U-started TMSR-NMs and for TRU-started TMSR-NMs. For the TRU-started TMSR-NM the table only gives the mass of fissile Pu isotopes (^{239}Pu+ ^{241}Pu). The weight of the other Pu isotopes and the minor actinides (Np, Am, Cm) in the initial fuel can be determined from the percentages given in the text. The HN concentrations (left column) are those which label the points on the curves displayed in Figure 4.

III.E Deterministic Safety

The total feedback coefficient at equilibrium is displayed in Fig. 5 as a function of the HN proportion for both a ^{233}U and TRU started TMSR-NM (see Table 1). Fig.5 also shows the contributions of the salt heating and salt density to the feedback coefficient for the ^{233}U case. The safety coefficients remain negative for all HN proportions, including the density coefficient which can be viewed as a void coefficient. The total feedback coefficient, which ranges from -10 pcm/K to -5 pcm/K, ensures a good

level of deterministic safety in all the ^{233}U-started TMSR-NM configurations [MHA07, MHM05]. Thus, safety is not a discriminating factor for the choice of the optimal HN salt composition.

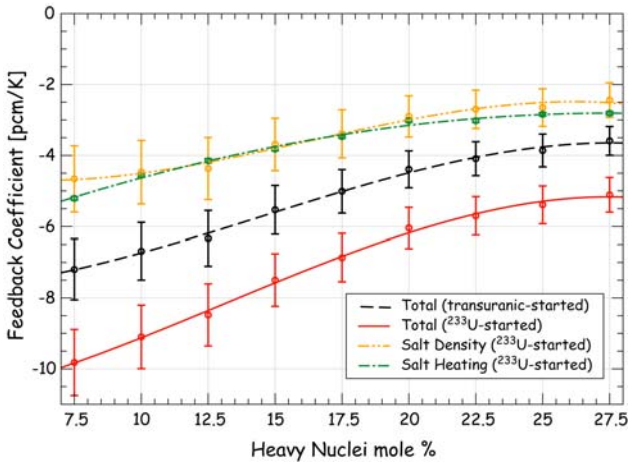

Fig.5. Feedback Coefficients of ^{233}U-started TMSR-NMs (at equilibrium) and of TRU-started TMSR-NMs (after one year of operation) as a function of the HN proportion.

When the TMSR-NM is started with TRU elements, the total feedback coefficients after one year of operation (dark blue curve in Fig.5) turn out to be close to that at startup time. Indeed after just one year of operation, inventories still remain rather close to the initial inventories. The calculation which takes into account the influence of the FPs generated into the salt by the first year of operation indicates that these FPs have only a small impact on the feedback coefficients. Fig. 5 shows that although the safety coefficients are initially slightly smaller in absolute value than those of a ^{233}U-started TMSR-NM, they nevertheless remain negative. When over years of operation of the MSR the equilibrium is reached, the blue curve moves onto the more favorable red curve.

III.F TMSR-NM as an actinide burner

The evolutions of the HN salt composition during the operation of a TMSR-NM started with a ^{233}U fuel (solid curves) and with a TRU initial fuel (dash curves) are shown in Fig.6. Except for Am and Cm, the TRU inventory of the latter is almost equivalent to that of the former after about eighty years of operation when the HN concentration is 22.5%.

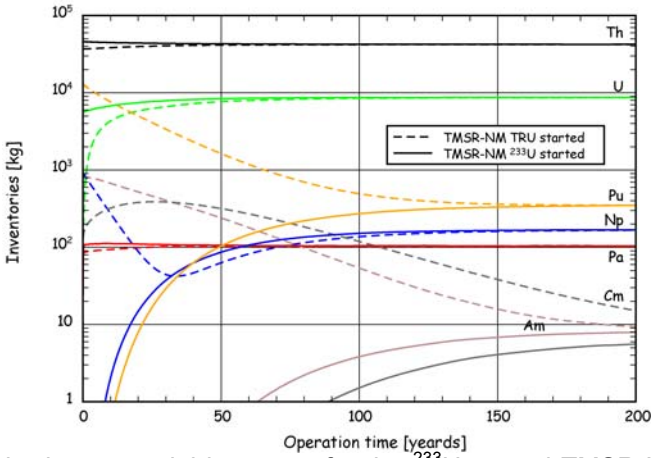

Fig.6. Evolution of the heavy nuclei inventory for the ^{233}U-started TMSR-NM (solid lines) and for the TRU-started TMSR-NM (dashed lines) for a salt containing 22.5 % of HN.

At this moment, more than 85% of the initial TRU inventory is burned. More generally, simulations show that the properties of the Thorium fuel cycle are recovered after 25 to 50 years for a TRU-started TMSR-NM, when the HN proportion varies from 7 to 27.5%.

An isotopic analysis shows that at equilibrium, the value of the ^{232}U/^{233}U ratio in the fuel salt lies between 0.1 to 0.3%. This corresponds to absolute amounts ranging from to 2.5 to 20 kg of ^{232}U, for

HN proportions between 7% and 27.5%. It is our opinion that such a quantity of the ^{232}U isotope, whose presence is sometimes mentioned when discussing the Th-^{233}U cycle characteristics with regard to proliferation, should not generate intractable problems for the reprocessing of the liquid fuel.

IV TMSR-NM Design and Thermal-hydraulics

In the previous section, we have presented the overall specifications of the TMSR-NM concept. Proceeding further, i.e. specifying and addressing the actual scientific and technical challenges associated with a TMSR-NM, requires a preliminary reactor design along with a definition of the main system components (reflectors, blanket, pipes, pumps, reactor vessel, heat exchangers and energy conversion systems). This will help define relevant ranges for the operational parameters (temperature and pressure gradients, fuel velocities as well as the on site chemistry needs) of the primary and secondary circuits. This work should be completed by out-of-reactor analyses (fuel clean-up unit and draining tank) regarding criticality and radiotoxicity issues. A preliminary design, allows one to define a specific work plan detailing the specifications of each component and which investigations are required to fulfil them,, the constraints imposed on these components and whether and how they can be dealt with. In the future, assuming that no show-stopper has been identified, the results can be incorporated into an optimized thorium-fuelled non-moderated MSR pre-conceptual design taking into account all relevant operational parameters.

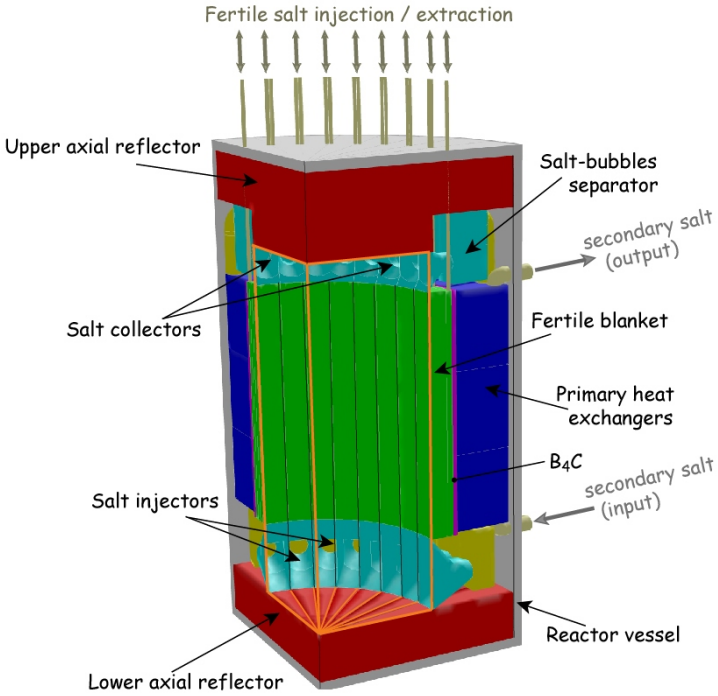

Fig.7 Sketch of a 1/4 sector of a TMSR-NM. Outside the core, the flow circulation is split into 32 angular sector units whose capacity corresponds to about 0.6m^3.

We are now in the process of translating the schematic description of the TMSR-NM as shown in Fig. 3 into a first drawing of a reactor in which all components are identified Fig. 7 displays a quarter section of the reactor with 8 of the 32 angular sectors into which the fluid circulation is divided. An iteration coupling of this design work with analyses of the nuclear energy deposition within the reactor and the thermo-hydraulics simulations, is needed to optimize the global salt circulation and to evaluate the temperature gradients. We expect a pre-design to be available by mid-2008. It will provide order of magnitudes for the operational specifications for each component from which a consistent research program on the components design and structure will be prepared.

As a matter of fact, from the schematic TMSR-NM concept alone, one can deduce some of the basic parameters which characterize the operation of the reactor and consequently already define some research actions. For instance, it would be interesting to investigate whether the temperature range above the salt eutectic can be extended up to 850°C. Similarly, it is established that the maximum salt velocities along pipes and fixed structures will range between 1 and 2 m/s. It has thus

been possible to prepare the design of an experimental circulation loop (see Fig.8) which allows a test of pumps and valves as well as an investigation of corrosion or abrasion effects on vessels, pipes and other components of the reactor. The final design of the loop is expected to be completed by the end of 2008. The construction will begin at CNRS-Grenoble at the beginning of 2009. The associated experimental program is expected to start towards the end of 2009.

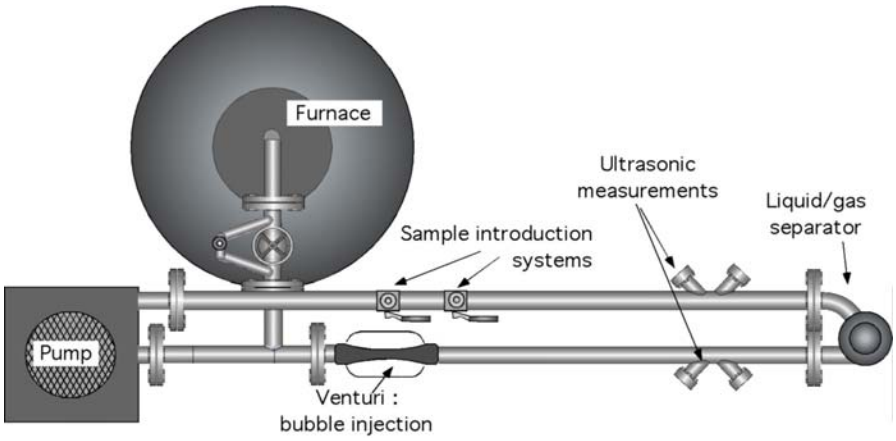

Fig. 8 Schematic design of the salt loop under construction at Grenoble. It allows an investigation of flow control and material resistance in ranges relevant for TMSR-NM operation. On the right side, a test unit for the He-bubbling performance has been included.

V Molten salt chemistry and treatment

V.A Fuel salt properties

The reference salt chosen for the TMSR-NM differs from the thoroughly analyzed MSBR fuel salt. The latter contained a sizeable component of BeF_2. The selection of the ^7LiF-ThF_4 binary system as TMSR-NM salt certainly eliminates the hazard associated with beryllium. On the other hand, the knowledge of its physics-chemistry is still not sufficient. In addition, the optimization of reactor operational parameters such as the melting point, the viscosity, the heat capacity and thermal conductivity or of the trivalent actinide (such as Pu) solubility may require the addition of another fluoride such as NaF. At any rate the solubility of fissile compounds like UF_4 and PuF_3 must be investigated as well as their influence on thermodynamic salt properties. Moreover the smooth operation of TMSR-NM, requires a fine control of the redox potential of the salt with respect to the couple UF_4/UF_3. Finally, the influence of impurities, in particular oxygen has to be carefully assessed.

CNRS teams have joined other EU groups to participate in the experimental determination of the structure of the melt by spectroscopic (NMR, EXAFS) methods and of thermodynamic properties of selected representative systems. This will be used to establish the validity of microscopic numerical simulations (quantum chemistry, molecular dynamics) which by means of extensive numerical exploration should help in the selection of an optimal TMSR-NM salt composition. Collaboration with an institution with adequate facilities (CEA or ITU) is necessary to obtain parameters such as the PuF_3 solubility.

Another important task is the characterization of methods for control of the electrochemical potentials and the activity coefficients of the lanthanides and actinides in the fuel salt which are of crucial importance in the batch processing phase (see Sect. V.b.2) or of the optimal salt redox potential (UF_4/UF_3 couple) in order to minimize corrosion reactions in the primary loop. A (thermo-) chemical model for the fission product chemistry in relation to redox properties and the oxygen impurity in the salt will be established. Appropriately design inactive experiments will be used as test of the calculations.

V.B Fuel salt processing

The salt processing scheme relies on both on-line and batch processes in order to satisfy the constraints for a smooth reactor operation while minimizing losses to waste streams. Support by acquisition of fundamental data for the separation processes is still needed especially for the actinide-lanthanide separation.

Both thermodynamic and kinetic aspects have to be considered to evaluate a reprocessing scheme. Fundamental data (redox potentials, activity coefficients) determined either by experimental measurements [LCB05, WPF05, FPS05, BRL07, DPF08, CMN07, CDL05, LEB93] or by molecular dynamic simulations [SST08], yield efficiencies for each reaction in its equilibrium state. Such types of calculations and the analytical equations they rely on, have already been published [DEL08, DPI05, DSS08, DMH08].

The proposed reference scheme used for the processing of TMSR-NM fuel salt is given in figure 9. The first step involves an on-line gaseous extraction with helium bubbling (green upper left-hand corner box) to remove gaseous fission products, Xe and Kr and noble metals by a flotation process. Presently it is assumed that this bubbling is performed on the entire reactor volume salt over a period of time of the order of the minute. On the other hand, batch process acting at a much smaller speed (few dozens litre per day) separates the actinides which are returned to the reactor salt from the harmful fission products (mostly lanthanides).

V.B.1 On-line extraction of gaseous fission products

Fission leads to the formation of many poorly soluble neutron poisons which have thus to be eliminated. Among the most harmful one finds ^{135}Xe. Each ^{233}U fission atom produces 0.75 noble gas atom (Xe and Kr). Their subsequent radioactive decay generates non volatile residues in the reactor core or along the piping and within the heat exchangers. A fraction (about 15%) of these gases is short-lived and naturally decays within the molten salt. The remaining 85% must be evacuated. Based on the positive experience acquired with the MSRE [CEA76, WMC70, GRI70, SEA70, HEN70], the method envisaged involves an extraction by means of He bubbling. Since gases have a very low solubility in molten salts (approximately 10^{-8} moles/(salt cm^{-3})/atm^{-1}) they tend to spontaneously migrate towards the interface between the gas cover and the salt interface. The injection of He bubbles inside the salt is thus expected to drag to the surface all the insoluble gaseous FP. This first step in the purification process before returning the salt to the reactor involves the further He gas circulation in decay tanks where specific active carbon trap solid FP such as Ba, La, Cs, Rb, Sr, Y and Zr.

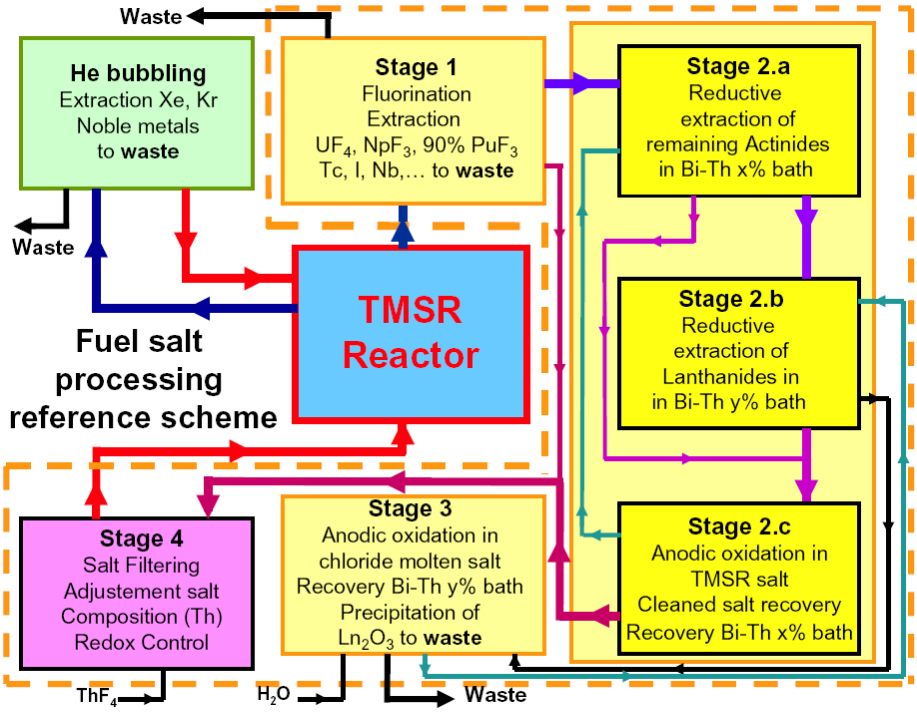

Fig. 9 TMSR-NM reference fuel salt processing. It is effected both on line as “He bubbling” or in batch mode (within the zone circumscribed by the orange thick dash line). The thick solid line follows the fuel salt progression along the four stages of batch processing. The evolution of its colour from blue to red symbolizes the improved cleanliness state. Purple thin lines correspond to actinide circulation within batch processing. Dark exiting lines indicate waste fluxes, while dark input lines for ThF_4 and H_2O corresponds to replenishing of fuel by addition of fertile material and input chemical for batch processing. Finally the blue lines show the internal return circulation of liquid Bi-Th melt.

It is also expected that fission products such as Nb, Ru, Ta, Te, … will be swept away by the He gas. Indeed, because of the low value of the molten salt redox potential these so called noble metals are in their metallic state. Thus they are not solvated in the salt as are most ionic species. Instead, they remain as a suspension with a weak binding to the liquid medium. It is believed that via a flotation mechanism He bubbles will be adsorbed to the noble gas particles allowing them to be carried away by the gas flow. This assumption is supported by the MSRE experiment where a significant part of the noble metals was retrieved inside the decay tanks.

A fraction of tritium (T) produced by fission is also extracted by the He flux. This tritium would then be separated from He by a chemical reaction on copper oxide. It is envisaged to store T, Xe and Kr waste in pressurized canisters for a length of time allowing a later safe release (typically 100 years).

ORNL experiments have already provided relevant data. Still given the present evolutions both in reactor design and in salt specifications, some updating is necessary. When operational at Grenoble, the loop described in Fig.8 should provide a unique testing facility. In the mean time, CNRS has decided to support the construction of a dedicated system (Fig. 10) to begin the exploration of the physical properties of the He bubbling extraction process and to evaluate its performance. The first experiments are planned in 2008.

V.B.2 Batch reprocessing

Batch reprocessing involves the daily (or weekly) removal of a fraction of the salt in order to separate actinides from harmful FP produced during the reactor operation. The volume that can be handled and the reprocessing time are crucial parameters for the overall viability of the MSR concept. Neutron reactor simulations have specified the minimum acceptable level of reprocessing (see Sect. III.B). As compared to the MSBR project [CEA76], TMSR-NM reactor physics requires batch reprocessing rates smaller by about two orders of magnitude.

The reference scheme depicted in Fig.8 involves four stages. The physics and chemistry for the last one “Stage 4: salt composition and redox control” and the corresponding research effort has been touched on in Sect. V.A. We now describe stages 1 to 3 and their impact on the organization of the CNRS effort.

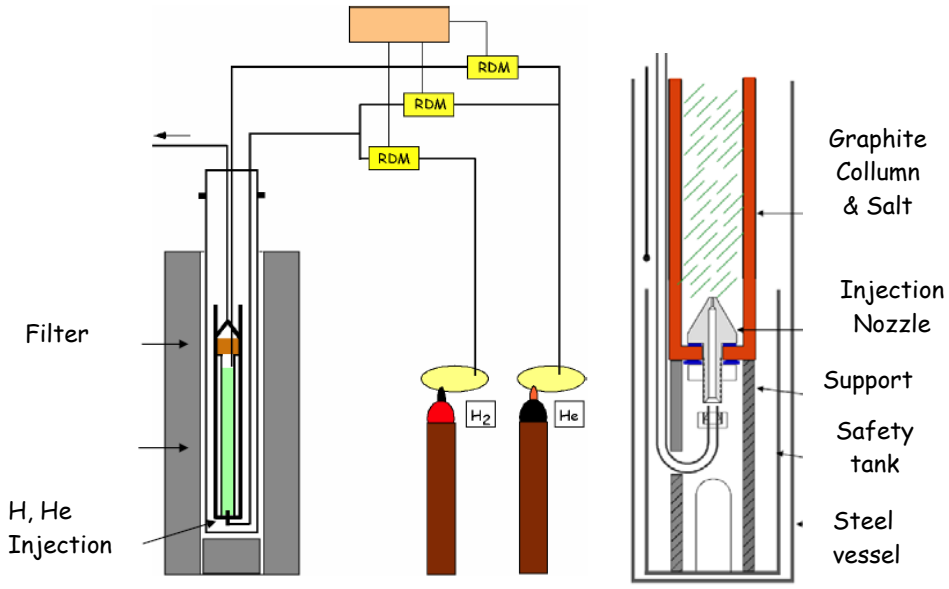

Fig.10 Temperature controlled He and H bubbling experimental column built at CNRS-Grenoble for the evaluation of the extraction efficacy from a molten salt of rare gases and noble metals.

V.B.2.a Stage 1 : Uranium extraction by fluorination

This method has been well tested at ORNL. The extraction yield of Uranium has been shown to exceed 99%. The method proceeds according to the following chemical reaction: $UF_4 + F_2(g) \rightarrow UF_6(g)$. It is performed in a chemical reactor in which the UF_4 laden salt is injected against contrary flowing fluorine (in 50% excess). At the same time, some other FPs are oxidized by F (I, Np, Te, Ru, Nb) as well as some alloying elements of the reactor structure materials (Cr, Mo). All chemical species are then extracted as gases which are circulated on a sequence of selective traps (NaF). The

temperature of the traps decides which molecular species is caught. This technique also allows a selective desorption of UF_6. Simultaneously Np and a significant fraction (~90%) of Pu are extracted as PuF_6 [ORN68] and NpF_6. By means of a H_2 flux, UF_6 NpF_6, PuF_6 are then reduced back to UF_4, NpF_3 and PuF_3 which are further reintroduced into the fuel salt.

A difficulty of this method lies in that fluorine as well as UF_6 (and most other gaseous reaction products) are very oxidizing species with a high corrosion potential. It becomes thus necessary to protect the vessel of the chemical reactor. The commonly used technique is that of the "frozen salt wall" [ORN66] in which the outside of the tank is cooled so as to generate an inside protective layer of crystallized salt with a width of approximately 1.5-2cm. The exothermic fluorination reaction takes place in the middle of the column. This technique is already used by AREVA at an industrial level.

In view of the solid experience already gathered at ORNL giving strong evidence for the practicality and efficacy of the fluorination method, this stage 1 is not presently the subject of extensive studies by CNRS groups.

V.B.2.b Stage 2 : Selective extraction of remaining actinide and lanthanides

As part of the GEN-IV strategy towards sustainable nuclear energy, TMSR-NM is also designed to burn fissile or fertile elements resulting from capture-decay processes inside the reactor. Thus the salt processing should be able to extract all actinides in order to inject them back into the reactor and simultaneously make sure they don't enter the operation waste stream. We have seen that fluorination is effective on U and Np and has a partial (~90%) efficiency regarding Pu extraction. The extraction of the remaining Pu and the heavier elements such as Am remains to be done. In addition within the Th-U cycle itself, a decision has to be taken regarding the relatively long-lived (27d) ^{233}Pa which ultimately will decay into ^{233}U. Whether it should be separated specifically or whether some waiting period will be imposed before effecting a second fluorination (waiting period which will also influence other decays) will have to be decided when scientific information will be of sufficient quality.

The CNRS teams have analyzed two options to separate FPs such as lanthanides and alkaline earths: electrolysis and/or reducing extraction. The experiments explore the electro-chemical behaviour of a LiF-ThF_4 salt at 600°C in presence of lanthanides (we recall that safety constraints have led us to avoid the BeF_2 used in the MSBR concept). The electro-activity domain of this salt is rather extended as a consequence of the strong solvatation of ThF_4 by free fluorides (see left part of Fig. 11) [DPI07, DSS08]. As far as extraction on an inert electrode is concerned, this stability translates in a potential increase by about 260mV which allows the extraction of Nd.

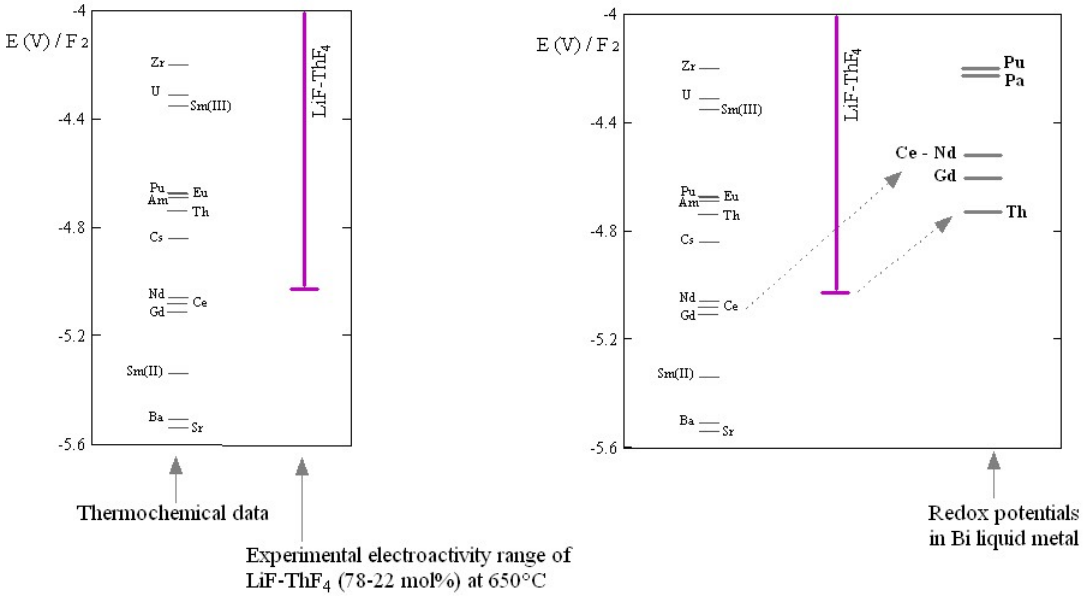

Fig. 11 Comparison between several redox potentials on a inert electrode (left) and on a liquid bismuth electrode (right). The figures also give the electro-activity domain in LiF-ThF_4 at 600°C. (the calculations are performed with the software HSC Chemistry version 4.1).

The scheme adopted as reference for the Stage 2 is shown in Fig.12.

An analysis of the performance of reducing extraction in presence of a metal (Bi-Th) has led us to select this method for the reference scheme. It is indicated by the boxes labelled Stage 2.a and

Stage 2.b in Fig.9. The principle of this extraction is given in the right part of Fig.11 which gives the redox potential of many elements by liquid bismuth (other metals than Bi can be and will be tested). It is seen that a selective extraction of actinides is possible when an external potential of -4.4V is imposed. All elements whose potential lies above -4.4V are extracted while the lanthanides remain in the salt phase. If Zr turns out to be undesirable for the operation of the reactor, it will have to be extracted first. The following sequence of chemical extractions can thus be envisaged: Zr first then Pa-Am-Pu-Pa (as well as remaining U and Np) and finally Ln.

Changing the composition of the metal (Bi) bath (noted in Fig.9 as x% and y% (x<y)) provides a way to control the potential. Indeed, Fig. 11 shows that the potential difference between lanthanides (Ce, Nd, Gd) and Th is about 100mV in liquid Bi. This suggests limited extraction efficiency with Bi only. It certainly can be improved by increasing the volume of Bi relative to that of the molten salt [DSS08]. On the other hand the extraction potential of Th can also be lowered in order to increase selectivity. This is achieved by controlled saturation of liquid Bi with Th. The extraction yield is enhanced by a repetition of the basic operation: contact between Bi-Li-Th_{sat} and the molten salt. Studies are presently underway to estimate extraction yields of Nd, Gd and Sm.

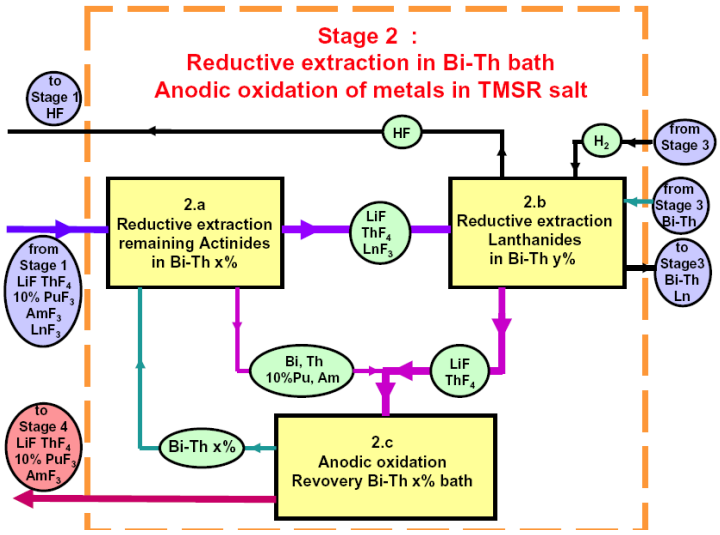

Fig. 12 Stage 2 of the TMSR-NM Fuel salt (Li-ThF_4) processing reference scheme (see Fig.9). The thick solid line follows the fuel salt progression along the four stages of batch processing. The evolution of its colour from blue to purple-red symbolizes improving cleanliness. Purple thin lines correspond to actinides circulation within batch processing. The letters x and y refer to different composition of the liquid phase (different concentrations of Th in the liquid metal bath [Bi here] or different volumes of liquid metal) chosen to favour selective and effective transfer into the liquid metal of either the actinides or the lanthanides. Dark lines show the side flows of either H_2 or HF gases while the blue lines indicate the internal return circulation of liquid Bi-Th melt at x% concentration within Stage 2 or from Stage 3.

Thermodynamic studies performed at ORNL have shown that Ba and Cs cannot be reduced and thus extracted from the salt. In the MSBR concept, all the salt had to be replaced after about 8 years of operation. A similar analysis of the impact of increasing Ba and Cs concentration in the salt is underway for the TMSR-NM. The amounts of Ba, Sr and Cs (radioactive daughters of Xe and Kr) depend on the efficiency of the on-line extraction (Sect. V.B.1)

The final step of Stage 2 (Stage 2.c) is the reverse of Stage 2.a performed by anodic electrolysis on a salt cleaned from its lanthanide content. After 2.c, one recovers a fuel salt which contains all the actinides extracted in Stage 2.a to which UF_4, NpF_3 and the PuF_3 extracted in the fluorination stage (Stage 1) can then be reincorporated.

V.B.2.c Stage 3 : Extraction of waste Lanthanide as oxides

In order to close the process, the metal bath (Bi-Th y% in Fig.9 and Fig.12) must be regenerated and the lanthanides extracted in their waste form. In the MSBR mode of operation, it was initially planned to realize this by an interaction with molten LiCl. This would transfer the lanthanides into the salt while thorium remains within bismuth. As shown in Fig.13, we have rather opted for an anodic oxidation of lanthanides in the molten mixture LiCl-KCl (58.8 - 41.2 mol%). The reaction whose

efficiency is going to be tested involves a lanthanide oxide precipitation in the presence of water vapour (lower left corner in Fig.13) followed by a regeneration of LiCl, KCl.

V.B.3 Flow sheet

Although the feasibility and performance of a TMSR-NM ultimately depends on the safe, reliable and efficient operation of both its on-line and batch fuel processes, the present status of our knowledge on the viability of the reference scheme schematized in Fig.9 present has not reached a stage adequate for the construction of an integrated processing design. The research will therefore be essentially conducted on He bubbling and the poorly known individual reactions shown for instance in Fig.12 and 13. From these studies which will extend over the next few years, one expects to reach a coherent flow sheet including performance assessment with respect to fuel salt cleaning and conditioning, speed and processing potential as well as the expected waste streams. Then it will become possible to build a first consistency evaluation of the overall scheme beyond what is initially warranted by the basic network of chemical reactions and the known laboratory performances. If no showstopper is uncovered, one will be in position to work on optimization, durability and economic issues. Corrosion and technical feasibility studies such as those described in Sect. VI should be undertaken as early as possible once chemical analyses have determined the most promising paths. Iterative feedback on the impact of the salt processing performance on reactor operation will have to be effected as part of the optimization process.

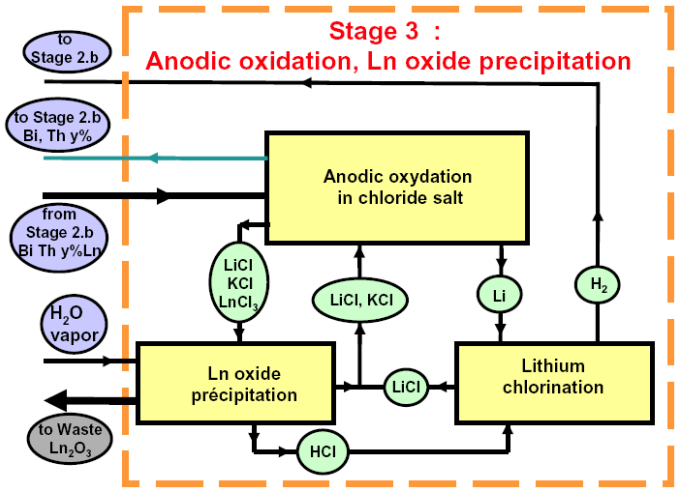

Fig.13 Extraction scheme of lanthanides from the Bi-Th y% bath and transformation into an oxide as ultimate waste.

VI Structural materials

Considerable documented past experience on material choices exists thanks to the two experimental molten salt reactors built in the US and elsewhere [AST07, Chap. V and references quoted therein]. The essential problem identified for the alloys used for the structural components was that of corrosion by molten fluorides. Accordingly, the first reactor built in the 1950's (the "Aircraft Reactor Experiment" or "ARE") used one of the best corrosion resistant alloys then available, i.e. a Ni-Cr alloy (an Inconel alloy of composition Ni-15%Cr-7%Fe, in wt %). It was found that although this alloy has excellent oxidation resistance in air, Cr was oxidised by the fluoride producing CrF_2 leading to severe corrosion. Hence the next experimental reactor, in the 1960's, the "Molten Salt Reactor Experiment" (or "MSRE") used a Cr free Ni alloy (Hastelloy B with composition Ni-29%Mo-5%Fe in wt%). This showed excellent corrosion resistance to molten fluoride salts, but unfortunately suffered from oxidation spalling on the air side. Further alloy development led to Hastelloy N (Ni-16%Mo-7%Cr-5%Fe-0,05%C, in wt%). Here just sufficient Cr was added to achieve a compromise performance between molten fluoride salt corrosion and oxidation in air. The added C led to improved creep resistance through grain growth control. Subsequently minor additions of Nb and Al to improve strength and oxidation resistance led to a new alloy which was tested in molten fluorides with a measured corrosion rate of 50 μm after about 11 years at 700°C. At that time, these alloys could be

produced as tubes, plates, bars, forgings and castings and they are readily welded and brazed. It should also be noted that over 40 years substantial improvements in industrial production have been made and alloys with substantially reduced levels of unwanted (impurities) or unnecessary (introduced via ferroalloys) elements can be achieved. It can be concluded that very reasonable solutions exist for structural materials for a molten fluoride salt reactor operating below 750°C.

Nevertheless, even if past experience has provided a wealth of information and well developed alloy solutions, a number of problems still require more work. As mentioned above, the alloy composition has to be a compromise between low Cr content for tolerable fluoride corrosion and yet enough to obtain acceptably low oxidation rates. To obtain acceptable fluoride corrosion it is in general sufficient to control the redox potential of the salt. This is achieved by means of the UF_4/UF_3 system used as a redox buffer. The natural trend of the molten salt potential is to evolve towards high and thus oxidizing values. It then becomes necessary to reduce a fraction of UF_4 into UF_3. This is done by an injection of small amounts of metallic Be which induces the reaction: $Be+2UF_4 \rightarrow 2UF_3 + BeF_2$. To stay within acceptable redox conditions, the typical value for the ratio UF_4/UF_3 should be 100. Under these conditions, overall corrosion, as measured in forced convection loops (~2m/s) is approximately 3μm/year for the core MSBR salt (with UF_4), but can reach 50μm/year for the molten salt considered for the secondary loop ($NaBF_4$-NaF). Finally, some fission products such as tellurium may lead to the grain boundary embrittlement observed in Hastelloy-N. This would occur via grain boundary diffusion, even though the diffusion coefficients are small. The precise mechanisms for the embrittlement (brittle intermetallic grain boundary phase, grain boundary segregation,) remain unknown. Recently russian teams have suggested that an addition of 0.1% Mn in modified Hastelloy-N would reduce this effect.

Irradiation resistance of Ni based alloys is also a problem which needs to be addressed: the helium production could be a limiting factor. In a fast neutron flux, the transmutation of Ni leads to the production of He atoms, which when they diffuse and are allowed to combine, can produce severe intergranular embrittlement. For austenitic steels (i.e. Ni containing stainless steels) this effect can be mitigated by a fine dispersion of precipitated carbide particles: the He atoms are then trapped at the matrix/carbide interfaces. Similar effects are expected in Ni based alloys provided an adequate microstructure is obtained. Little is known for irradiation induced swelling, creep and solute segregation. These effects should be investigated in candidate materials and eventually reduced to acceptable values given the irradiation conditions by a fine tuning of the alloy composition and microstructure.

The CNRS teams have so far focused their research on less well known Ni-W-Cr alloys in which Mo is substituted by W. They have a number of potential advantages. Indeed, the substitution should lead to an increase in creep resistance by at least an order in magnitude. Moreover long term activation problems should be reduced. These alloys are strengthened by solid solution effects and by the presence of strong short range atomic order. It is expected that the corrosion resistance in molten salts will be similar, while the presence of W improves the oxidation resistance in air. It might thus be possible to further reduce the Cr content. Another interesting feature is that large W content leads to grain boundary precipitation of W (see Fig. 14) which, unlike with Hastelloy-N, can also improve the creep resistance allowing a higher operating temperature of the reactor.

Following initial laboratory studies, several compositions will soon be produced on a semi-industrial scale by the French firm “Aubert et Duval”. Work in progress concerns both the molten fluoride corrosion and oxidation resistance in air. Since corrosion is strongly chemistry specific, further work is necessary in order to study corrosion of these alloys in the secondary loop ($NaBF_4$-NaF).

Finally, at the present early stage of selection of reactor specifications, other materials can be envisaged. For instance, instead of Ni-W-Cr alloys, ZrC could be used for the axial reflectors. The fabrication of large reactor ZrC components is challenging, in terms of synthesis, manufacturing, mechanical properties and behaviour under irradiation. A substantial research effort is required here to address the specific needs of molten salt reactors. Graphite might also be envisaged for some components. Specific high density and low porosity graphites had been developed for the MSRE. Unfortunately much of the know-how has been lost. Nevertheless, development has continued for the high temperature reactor leading to substantial improvement of life expectancy in irradiation conditions. The key factor has been the improvement of the isotropy and the homogeneity. Still, the difficulties involved in producing the appropriate grades should not be underestimated.

As of today, it does not seem that there exist materials problems so intractable as to forbid the conception of a TMSR-NM,.For the metallic structural components, the proven Hastelloy N is already a good candidate for temperature up to 750°C. Presently ongoing research holds the promise for substantially improved alloys (in terms of corrosion and oxidation resistance, high temperature creep, reduced long term activation, ..). Given recent advances, graphite does not appear to raise such major

concerns as was the case at the time of MSBR. Nevertheless, more work is required on the effects of irradiation on the Ni based alloys. In addition, an investigation of material compatibility with the several batch processing fluids (liquid Bi-Th alloys, chloride melts) is also needed.

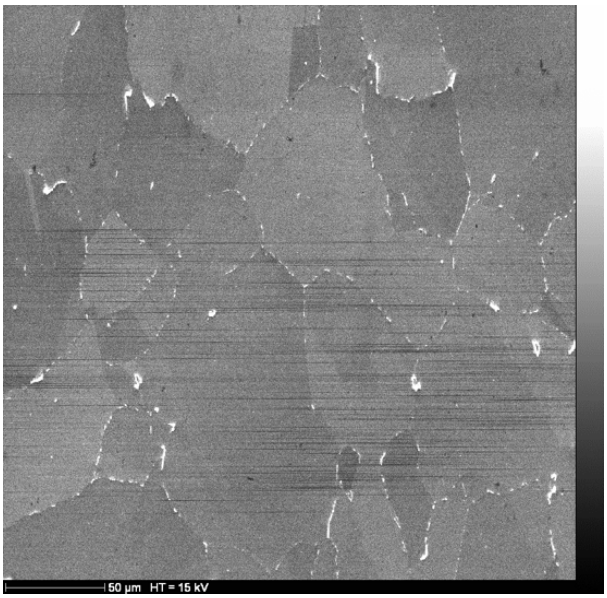

Fig. 14 Re-crystallized microstructure of a Ni-W-Cr alloy tailored for a usage in contact with fluoride salts. The white spots at the grain boundaries are W precipitates.

VII Nuclear waste

The time scales of the nuclear physics underpinning the technology of fast reactors (determined by rather small fission and capture cross sections) as well the neutron fluxes compatible with the resistance of present and foreseeable structure materials, imply that a significant effect in terms of transmutation can only be achieved if such reactors are operated over a long time. The abscissa time scale used in Fig.6 is representative of this rather slow evolution of fuel compositions. It also shows that the equilibrium state is only reached after many decades. For that reason, any family of reactors presently envisaged in the context of GEN-IV is meant to be deployed over a long period. The typical order of magnitude considered in most scenarios is one to two centuries. Given human past history, the sometimes chaotic evolution of our societies and the replacement rate of technologies, this is already a rather long period. Although there is no strong indication that availability of natural resources is going to fix a limit of its own, it would be somewhat presumptuous to imagine that we are working on the definition of a smoothly evolving line of energy production systems for a much longer span of time, say for instance, a millennium.

These general considerations set the frame for a discussion of the nuclear waste generated by a system and lead us to consider two different contributions to the waste. First there is that fraction which is produced during the operation of the reactor as defined by its original mission of energy production. Second one must include that which results from the decision taken in an uncertain future to abandon this specific line of reactors and to move over to another technology. In this section, for the purpose of allowing a numerical evaluation, we arbitrarily fix to 200 years after the start of TMSR-NMs operation as the time when the decision to stop them is taken. It will be seen that changing this date by a factor 2 one way or the other (100y to 400y) does not crucially affect our conclusions[13]. If future technologies replacing fission do not have a usage for the inventory remaining in the core of the stopped reactors (and also for the matters in the cycle facilities), these matters must also be considered as a waste. It is usual to label “End of Game” this situation.

It does not seem appropriate to advertise the virtue of a specific GEN-IV system on its radio-toxicity legacy to future generations by solely considering its merits with respect to the minimization of its accumulated waste stream. This is especially the case if the radiotoxic impact of the inventory

[13] It is anyhow likely that any decision to stop will be implemented in a progressive manner at various times in different geographical zones.

waste (which is also handed over to the same generations after several centuries) turns out to be significantly larger. For instance, this could lead to inadequate conclusions as to the geological storage capacity needs.

On the example of TMSR-NM, which as a fast neutron system shares many of the characteristics of the other GEN-IV open cycle reactors, we discuss the two contributions to the nuclear waste[14] Traditionally, the first waste component is measured relative to the service given by the reactor, i.e. the TW.h of nuclear energy or better of electricity [TW.h(e)] produced. The second component must be considered in absolute terms irrespective of the electricity produced before the decision to stop the reactor fleet is taken. On the other hand, the geological storage capacity which has to be prepared is only concerned with absolute amounts.

VII.A Waste stream

From a physicist point of view, the true nuclear waste consists in the fission products whose nuclear energy content is either zero or non exploitable. Although the unstable FPs are a major (initially dominant) contributor to the radio-toxicity and the heat production of the waste for the first few centuries, it is considered that, because of their generally shorter half live, they do not a priori create insurmountable challenges for present civil storage engineering technology. With the possible exception of a few mono-isotopic elements their transmutation is also not considered. The quantity of fission products is directly proportional to the number of fissions and thus for a given typical reactor efficiency to the amount of produced electricity. Within minor qualitative and quantitative differences, it is the same for both Th-U and U-Pu cycle. It is well documented both in Sievert magnitude and long term evolution (see Fig. 15 and 16). Apart from the influence of the reactor electricity production efficiency, a point which presently cannot be precisely assessed for any GEN-IV system, the FP waste per TWh(e) generated by a TMSR-NM won't differ much from that other GEN-II, GEN-III or GEN-IV reactors.

At the industrial level, of course, the actinides also contribute to the waste stream. Political decisions (for instance on the status of the uranium and plutonium in the spent fuels) may also determine which of these elements is a waste or a valuable future resource. Since the TMSR-NM is proposed here as a candidate for the sixth class of GEN-IV, it should be compared with other GEN-IV systems which also rely on the closed cycle and consider Pu and possibly other trans-uranium elements as a fuel rather than a waste. Indeed, as it is envisaged for most GEN-IV systems, the first TMSR-NMs will be started with the Pu and heavy elements from the used fuels of GEN-III fleet (see Sect.III.D.2). Recycling implies that during the operation of the reactor as much as possible of the actinide content of the spent fuel of this new class of reactors is sent back into the next cycle of operation. Once they have been started, the only external feed-in of breeders is fertile matter, either ^{238}U (depleted U in fact) or ^{232}Th. As a consequence, in a first approximation, it is solely the efficiency of the chain of chemical separations of the actinides which determines the actinide waste stream. Today, the chemical treatment of molten salt as described in Sect.V does not have the same status as that of the hydro metallurgic reprocessing of solid fuel (UO_2 or even metallic fuel). Indeed while the latter can already rely on a large body of experience at both technical and industrial levels for the extraction of uranium, neptunium, plutonium and americium, the performances of the sub-processes outlined in Sect.V are often only evaluated, if not just estimated, at the laboratory scale.

Nevertheless, taking the scheme proposed in Sect.V as a starting point and using all available data, we can already obtain a reasonably good estimate of the performances of the various stages involved in Figs. 9, 12 and 13. Since the on-line He-bubbling only involves extraction of fission products (rare gases, some noble metals) its efficiency will not impact actinide waste but only reactor operation in determining how much neutron poison remains within the salt. The efficiency of the first stage of batch processing (fluorination) is rather well known from ORNL work. Extraction of uranium and neptunium has been demonstrated at a better than 99% level. On the other hand only 90% of plutonium is extracted at this stage. Stage 2.a effects the remaining extraction by means of an interaction of the salt with a liquid metal bath (Bi for instance). Its efficiency will depend on several factors such as the value chosen for the liquid metal Th saturation factor. This factor will control how much of the lanthanides are extracted together with the actinides. The kinematics of the reaction and the acceptable level of poisoning of the fuel (which in turns affects the reactor operation and the

[14] It is usual to consider that each reactor considered individually will be operated over several decades (60 years is often quoted as a typical value). At the end of its life, the dismantling of the reactor will also produce additional waste which has to be taken into account. The recycling facilities (an integral part of the reactor for a MSR) as well as many other components of the reactor also have a finite life expectancy (often much shorter than 60y). Only when TMSR-NM will have reached the integrated preliminary design phase will it be possible to perform an evaluation of the nature and quantity of the associated waste.

breeding capacity) are still open parameters. How many duplications of each stage should be considered to reach the a priori desired extraction efficiency has also to be analyzed. Given the present knowledge and unknowns, in order to perform evaluations we have considered that, as in present industrial installations treating spent UO_2 fuel, it should be possible to extract 99.9% of the actinides remaining after stage 2.a. After this second extraction (stage 2.b) the actinides left in the salt will be taken along with lanthanides into stage 2.c and transferred to stage 3 and from there to the waste stream.

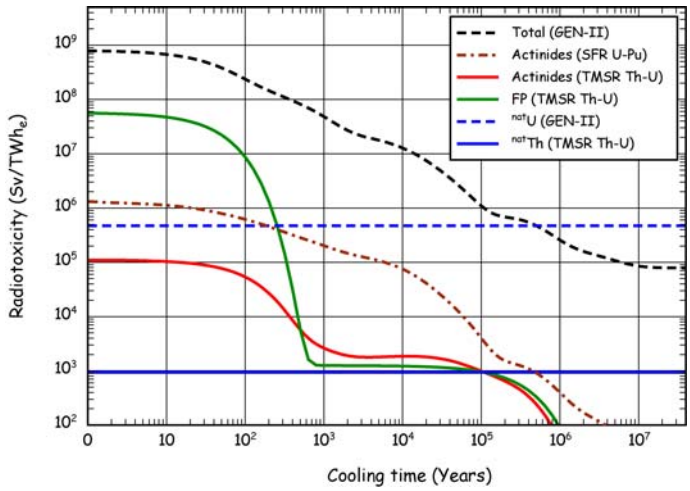

Fig.15 Time evolution of the radio-toxicity of the waste stream of nuclear reactors per TWh of produced electricity. The upper curve "Total (GEN-II)" (Black-dash) corresponds to reactors presently in operation. The curves "FP (TMSR Th-U)" (Green-solid) and "Actinides (TMSR Th-U)" (Red-solid) respectively give the radio-toxicities associated with the FP and the actinide waste stream of a TMSR-NM assuming a 40% energy efficiency (see text for assumed recycling performances). The lines "^{nat}U (GEN-II)" (Blue-dash) and "^{nat}Th (TMSR Th-U)" (Blue-solid) give the radio-toxicities of the needed natural resources. For the GEN-II reactor the curve assumes a 4% enrichment level. The brown dash-dot curve "Actinides (SFR U-Pu)" provides a comparison with the actinide waste stream of a GEN-IV Sodium Fast Reactor within the U-Pu cycle.

Fig. 15 compares the radio-toxicity resulting from the operation of a typical UOx GEN-II reactor (N4, 4% enriched U, 47.5 GWj/t, no recycling) with that associated with TMSR-NM per TWh of electricity produced [SDT00, Sal95]. For the waste of the TMSR-NM the contribution of FPs has been drawn separately from that of the actinides. The reprocessing rate is that considered in Sect.III (~40l of salt per day). As mentioned above, the PF curve dominates for the first centuries. For the first millennium, the main contributions are from ^{238}Pu ($T_{1/2}$=88y) and ^{241}Am ($T_{1/2}$=432y). Later on, the dominant contribution originates from that small fraction of ^{231}Pa ($T_{1/2}$=33000y) and ^{233}Pa (via its daughter ^{233}U, $T_{1/2}$=160000y) which has not been recycled. Between 10^4 and 10^6 years one also observes a revival of the ^{238}Pu contribution via its grand daughter ^{230}Th (T1/2=75000y). It thus appears that the extraction efficiencies of U, and especially Pa and Pu are crucial. Note also that similar assumptions on efficiencies have been made for the Am and Cm elements for which presently no data is available[15]. The collaboration on such questions of CNRS teams with an institution with adequate processing facilities such as for instance ITU at Darmstadt (SUMO FP7 proposal) will be attempted.

Given that the radiotoxicity of the TMSR actinide waste depends mostly on the efficiency of Pu and Pa extraction, one can say that if the efficiency of stage 2.a (Figs. 9 & 12) were only 99% (instead of 99.9%) the curve " Actinides 'TMSR Th-U)" would move up by a factor ten. In view, on the one hand, of the remaining uncertainties on all systems, and on the other hand, of the much better established fuel recycling performances (at least for U, Np, Pu and Am) for solid fuel, a comparison with other GEN-IV systems (Sodium, Lead or He cooled) using solid fuel and global recycling is difficult to perform. One can nevertheless say that with the above considered extraction performances for stage 2.a (99% or 99.9%) the TMSR-NM within the Th-U cycle performs at least as well or better with respect to the radiotoxic content of the waste stream (see Fig.15).

We have mostly discussed the waste associated with the actinides which dominates the long term radiotoxicity. The total radiotoxicity of the waste stream also includes that of the fission products. In Fig. 16 we show the radiotoxicity evolution of the total (actinide + FP) GEN-III waste stream (dash)

[15] Yet, the efficiencies we have chosen for Am and Cm are compatible with the redox potentials of these elements or with those of neighbour or analogous elements.

accumulated over 200 years[16]. We compare it to the total TMSR waste stream over the same period (dash-dot). The main modification as compared to Fig.15 is that Fig.16 takes into account of the radioactive decay of the (mostly short lived) fission products. The solid curve which almost covers the dash-dot curve gives the total waste stream radioactivity after the inventory incineration phase described in next subsection. This figure illustrates that it is only after several hundred years that the benefit of moving over to the closed cycle brings about a significant reduction, by more tan two orders of magnitude, of the global waste stream radiotoxicity.

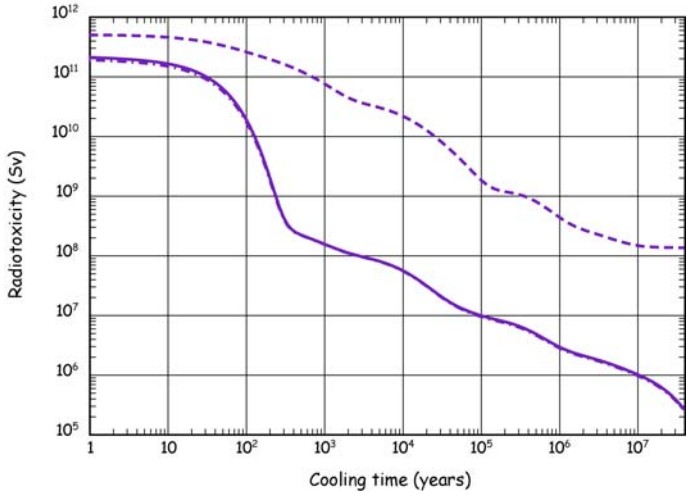

Fig. 16 Time evolution of three total waste streams radio-toxicities (FP+Actinide) The dash curve corresponds to a GEN-III reactor operated over 200 years (open cycle). The dash-dot curve (almost undistinguishable from the solid curve) corresponds to a TMSR-NM of the same power operated over the same period of time (closed cycle). The solid curve shows the total waste stream after the inventory incineration process.

VII.B End-of-Game waste

TMSR-NM as all other closed cycle GEN-IV systems is a fast neutron reactor. Consequently, its actinide fuel inventory is large. For instance, the 2.5GWth (1GWe) TMSR-NM considered in this document has an inventory of 8.74t U, plus (0.35t Pu, 0.17t Np, 0.10t Pa, 0.008t Am, 0.006t Cm)[17]. Because the radio-nuclides present in the Th-U cycle differ from those in the U-Pu cycle, for the first millennia, the intrinsic radiotoxicity of the core of a TMSR-NM is smaller than that of other GEN-IV fast reactors of the same power, by approximately a factor 5. On the other hand, the present uncertainties on all GEN-IV systems as well as a resurgence around 10^5 years of the radiotoxicity of the TMSR-NM inventory due to the decay of ^{233}U does not necessarily make this an overly significant argument in favour of TMSR-NM.

In Fig.17, we compare the time evolutions of several radio-toxicities. As an illustration of the interest[18] of a transition to the closed cycle, the purple dash curve gives, as a reference, the contribution to the radiotoxicity of the actinide waste which would result from 200 years of operation of a GEN-III reactor. As already shown in Fig. 15, it is about two orders of magnitude larger than the actinide waste stream from a TMSR-NM producing the same amount of energy (purple dash-dot curve).

The purple curves correspond to actinide waste streams and the three green curves to reactor inventories, respectively. The dash green curve shows the GEN-III reactor actinide waste which (apart from the depleted U which is effectively treated as a low-level waste) has been used as the initial inventory to start the TMSR-NM. The dash-dot curve corresponds to the TMSR-NM inventory after 200 years. One sees that this curve is not significantly different from that of the initial inventory of the TMSR-NM. In addition, because it lies above that of the TMSR waste stream (purple dash-dot curve)

[16] As for all similar curves, a convolution to take into account the decay of the short lived isotopes has been effected.

[17] We do not consider the 46t of Th in the core as waste destined to a low-level final storage. Indeed even in the steady state of the reactor they mostly consist in ^{232}Th with few additional kg of ^{230}Th ($T_{1/2}$=75000y) and a quarter of a kg of ^{229}Th ($T_{1/2}$=7340y). We assume that all thorium of the final inventory is chemically extracted and handled separately from the high activity elements. Its status is thus similar to that of the depleted uranium of used fuels which is dominated by ^{238}U.

[18] In addition to the sustainability of energy production since no input of ^{235}U resource is anymore needed.

accumulated over two hundred years it will dominate the final waste. With the log scale used in Fig.17, an operation of the fast TMSR-NM over 400 years instead of 200 which would induce an upward shift of the three purple curves by less than a third of a division (factor 2), does not change this conclusion.

Thus, for societies which consider that the choice of a technology covering their present energy needs must take into account their legacy to future generations, there is a definite incentive to optimize global radiotoxicity including both the waste stream and the inventory. In that respect, finding ways to reduce the radiotoxicity of the TMSR-NM inventory as one enters the End-of-Game phase to the level of that of the accumulated waste stream is of major interest. For the TMSR-NM, this requires a destruction of ^{233}U whose decay dominates the radiotoxicity of the inventory between 10^4 and 10^5 years.

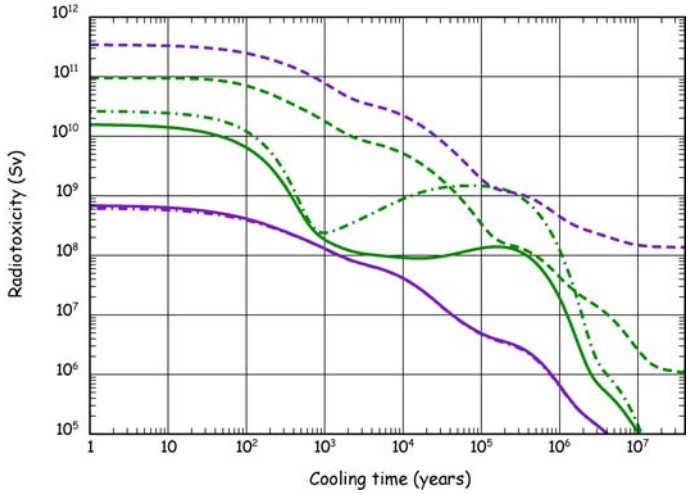

Fig.17 Time evolution of several waste radio-toxicities. The purple dash curve at the top is the actinide waste radiotoxicity resulting from 200 years of operation of a GEN-III reactor; i.e. a two century open cycle. The green dash curve corresponds to the matters extracted from the spent fuel of a GEN-III reactor which are used as the initial inventory of the TMSR-NM. The green dash-dot curve corresponds to the contribution of the radioactive inventory of the fuel in a TMSR-NM core and its recycling facility when the steady state has been reached. The green solid curve corresponds to the transformed inventory after sixty additional years of incineration in a dedicated TMSR. The purple dash-dot curve gives the accumulated actinide waste stream produced over 200 years of TMSR-NM operation while the purple solid curve shows how this waste stream radiotoxicity is modified by the contribution of the additional actinide waste stream during the 60 years devoted to reduction of the inventory.

If a decision to abandon fission and to move over to a new technology is taken at some date in the future[19], apart from the decision to immediately consider the inventory as an ultimate waste to be disposed of in a geological storage, it seems that the only acceptable alternative will be to work on the destruction of this inventory. Should this choice be made, it is unlikely that it will be decided to design and built an entirely novel system relying on fission for that sole purpose. On the other hand, as Physics tells that low energy neutrons are by far the cheapest way to achieve the fission of actinides, it seems reasonable to assume that one will try to hand this task over to an already available and well-tested fission-based technology albeit with a changed mission. Now, instead of the generation of energy and brreding, the priority will be given to the incineration of as much as possible of the inventory in order to reduce the burden on the storage[20]. This is a scenario that we have started to investigate as another illustration of the versatility of the TMSR-NM concept.

To that aim, we consider a situation where, in fleet of TMSR-NMs which it has been decided to stop, for every seven reactors[21] an incinerator TMSR is started which, over a sixty year period, will burn the inventory of the 7 reactors. Keeping in mind that one of the criterions is the minimization of technological untested innovations as one enters the End-of-Game phase, we choose a TMSR with

[19] We assume that this novel technology does not rely on nuclear fission and has no use for the energy content of actinides.

[20] Of course, this incineration will also contribute to an additional production of energy

[21] As a matter of fact the exact value of the ratio is 6.9 to 1 which for this preliminary study, we approximate as 7 to 1 for the sake of redaction simplicity.

the same basic design and characteristics described in Sect III and IV. Since ^{233}U generation is now to be avoided, the main modification is that the fuel of this TMSR will not anymore contain Th. The reactor will also not have breeding blankets. This TMSR has the same nominal power 2.5GWth (1 GWe) as those studied for energy production and will thus also produce energy (at the level 1 / 7). For the incinerating function which is now assigned to the TMSR, the salt must be changed to the so-called FLiNaK (46.5% LiF, 11.5% NaF, 42% KF) which has a low fusion temperature compatible with a small load of heavy elements (680kg) while ensuring a a not too much thermalized neutron spectrum.

Element	7 TMSR-NM Inventory	TMSR-NM Incinerator Inventory	Reduction Factor
U	60.240	5.520	10.9
Np	1.160	0.422	2.7
Pu	2.400	1.210	2.0
Am	0.055	0.039	1.4
Cm	0.039	0.078	0.5
HN	63.894	7.269	8.8

Table 2 Comparison of the inventories of 7 TMSR-NMs (see note 21) with that of the incinerator TMSR after sixty years of operation. HN stands for heavy nuclei. Masses are given in ton. The last column gives the destruction efficiency.

In Fig. 17, the two solid curves correspond to the situation after sixty years of operating the incinerator TMSR[22]. The purple solid curve shows the effect of the additional contribution to the actinide waste stream. Expectedly, it barely increases the waste stream resulting from a 200 year operation of the TMSR-NMs. The green solid curve gives the evolution of the inventory of the original TMSR after 60 years of incinerating mostly ^{233}U but also some neptunium and plutonium (see Table 2). Although the radiotoxicity of this inventory still exceeds the total actinide waste stream over 260 years, the comparison with the green dash-dot curve shows an improvement by at least a factor 10 for the period considered critical for the dependability of geological storage civil engineering (beyond several 10^3 years).

In principle, this process which already decreases the reactor fleet by a factor 7 can be repeated to complete the End-of-Game phase. In this way, after sixty years a reactor fleet of the size of that presently operating in France would be reduced to just one or two reactors terminating the process of destruction of the U and Pu elements. Once their task is completed, these few remaining reactors would be stopped. At that moment, the radio-toxicities of the accumulated waste stream and of the inventory for the entire initial TMSR-fleet would have been reduced to about the same magnitude[23].

At any rate even considering the sum of this remaining inventory and the waste stream after 260 years, the transition from the GEN-III open cycle (red dash-curve) to GEN-IV using TMSR-NM (sum of green solid curve plus purple solid curve) already leads to a decrease of radiotoxicity by more than a factor 10. Except for the fact that the waste streams of fast reactors working with the U-Pu cycle is likely to be larger, these conclusions should qualitatively hold for any other GEN-IV type of reactor.

This discussion aimed to show that any optimization of the interim storage and the final disposal capacities for the global nuclear waste issue cannot restrict itself to a consideration of only the waste stream accumulated during the energy production phase. The End-of-Game inventory which may well turn out to be the dominant contribution to the waste has to be considered simultaneously even if this requires making additional assumptions as to when in the future it is decided that the game should end. The TMSR-NM offers interesting perspectives in that respect.

In the framework of the CNRS program, further studies are being planned which will investigate not only radio-toxicities but also heat emission from the waste. We plan also to investigate schemes where the small amount of americium and curium produced during the exploitation of a TMSR-NM instead of being kept in the salt is regularly discarded to the waste stream. We also have to consider the modifications to the chemistry of the recycling batch process (Sect. V.B.2.b & c) which result from the elimination of Th in the incinerating phase.

[22] The contribution of the incinerator to both waste stream and inventory have been rescaled (factor 1/7) to that of the original TMSR-NM.

[23] Once the FP contribution to the waste stream radiotoxicity has disappeared in the background of that of the actinides

VIII Safety

The work of the CNRS community on the question of the safety of TMSR-NM has just started. Of course, the analyses performed at Oak Ridge for the MSRE and the MSBR as well as some technical options then adopted are a great help in so far they can be transposed to a non moderated system such as the TMSR-NM. Nevertheless, this section of the document corresponds more to a to-do list than to a progress report. Of course, in Sect III we have mentioned that the safety potential of the TMSR-NM core can be considered to be high due to its favourable feedback coefficients, in particular, its negative thermal expansion coefficient. Neither the temperature nor the pressure conditions appear as a major safety constraining factors. On the other hand, the very original design of the core and some specific features of MSR (moving delayed neutron precursors, coupling to a processing facility) require a much more detailed definition and analysis of incidental and accidental events.

As a matter of fact, even safety guidelines for a MSR (taking into account the increased level of the safety standards since the MSBR project was abandoned) have to be established both for the core and salt recycling facility. These imply an identification of the potential risks (source term, residual heat) specifically attached to the use of molten salt loaded with radioactive elements (delayed neutron precursors mobility, criticality) and to the specific design and operating conditions (efficiency of helium bubbling, rate of fuel-salt extraction for reprocessing). The integration of the processing unit into the reactor operation scheme generates additional original problems for the overall safety.

Other questions not yet investigated involve reactor operations (start-up and shut-down regimes), accident initiators and scenarios. These have to be identified, characterized and categorized. This task will become easier once the preliminary reactor design (Sect. IV) is completed. Of great use will be the transposition of transient analyses computer codes built for the safety analysis of solid fuel reactors to that of non-moderated MSRs.

The CNRS teams have contributed to the definition of a FP7 EURATOM proposal which among other themes addresses some these safety questions.

X Deployment Capacities

Despite the vast thorium natural resources, all of present nuclear energy production relies on the U-Pu cycle. The front-end and back-end of the operation of GEN-II (GEN-III) reactors generates large quantities of depleted uranium as well as a used fuel heavily loaded with TRU which carry most of the radio-toxicity. These matters are presently kept in dedicated interim storage facilities. For various reasons, the creation of geological repositories for the permanent disposal of waste has met with many difficulties and has turned out to be a very difficult task (even for sites designed to handle only a small fraction of the TRU contents of the used fuel). Thus, there is a definite incentive to find a usage for the GEN-II TRU and to limit to a minimum the ultimate waste of future nuclear energy production.

We have shown that as other reactors, the TMSR-NM can be considered among the systems with a capacity to incinerate a significant fraction of the GEN-III TRU. In turn, this answers the question of the deployment of a fleet of TMSR-NM in spite of the absence of naturally available ^{233}U. Intrinsically, the situation is the same for other GEN-IV reactors since ^{239}Pu, which all of them depend on to start operation, is no more available in nature than is ^{233}U. The only difference is that ^{239}Pu (along with other TRU) is already now present and will be present in the waste of LWRs. The results presented in Section III establish that a TMSR-NM can burn this Pu just as well as any other GEN-IV system and will use it to effect a transition towards the Th-U cycle. As a matter of fact, when it comes to starting any GEN-IV reactors, it appears that the real worth of GEN-III used fuel lies more in the stock of fissile ^{239}Pu needed to start any future fast neutron reactor fleet than in the much larger associated amount of depleted uranium[24]. One should also keep in mind that, in many developed countries, just as depleted uranium, thorium is already available, mostly as the unwanted by-product of industrial rare-earth extraction. For instance, given the present annual French nuclear electricity production (400TWh), the ^{232}Th needs of an entire TMSR-NM fleet would be covered for over a century by the quantity of thorium presently in interim storage on French territory[25].

Another question can be raised as to whether the smaller breeding potential of the Th-U cycle (both in the thermal and fast regime) as compared to that of the U-Pu cycle (in fast reactors) would

[24] A significant fraction of the presently available and future depleted uranium from GEN-II and GEN-III reactors is bound to become a waste unless breeders are used for millennia.

[25] One can estimate that the amount of ThO_2 within French interim storage facilities will soon be of the order of 8000t while a 60GWe fleet of TMSR-NM would use about 60t per year.

allow a smooth transition from GEN-III reactors to a TMSR-NM fleet. Before getting into an evaluation of the breeding capacity of the TMSR-NM, one may recall that to start a SFR using solid fuel one must have available the TRU amount for about two cores (to take into account the necessary cooling period before reprocessing). In contrast, only one core load is needed for a TMSR-NM since the reprocessing is performed as a (semi)continuous procedure. This compensates for the difference in the breeding potential of the two cycles. As a result, the transition from a GEN-III to a GEN-IV fleet can be initiated just as fast with TMSR-NMs as with SFRs.

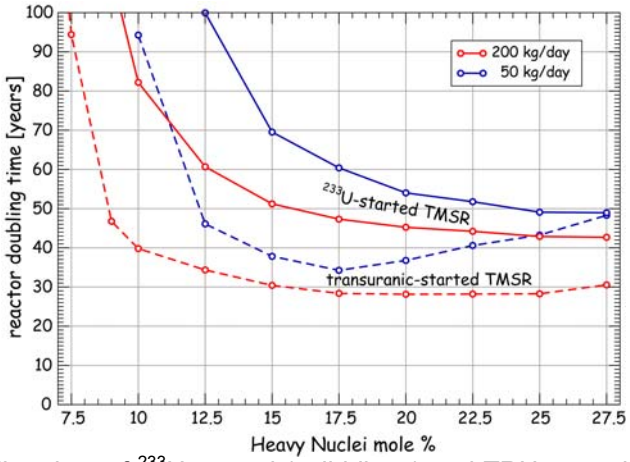

Fig.18. Reactor doubling time of ^{233}U-started (solid lines) and TRU-started TMSR-NMs (dashed lines) for the different HN proportions and for two off-line reprocessing capacities: 200 kg/day or ~60l/day (red lines) and 50 kg/day or ~15l/day (blue lines).

The “doubling time” defined as the period of time over which the TMSR-NM must be operated to produce the ^{233}U inventory for another reactor is a quantity relevant for estimating the deployment potential of the TMSR-NM. This quantity is plotted in Fig.18 for a wide range of values of heavy nuclei fractions in the salt. Since the breeding capacity depends on how well the salt is cleaned from the neutron-poisoning FPs, the doubling time is also evaluated for two different reprocessing performances (50kg/day and 200kg/day). Not surprisingly, the cleaner the salt, the shorter the doubling time. It is also seen that in TRU-started TMSR-NMs significantly larger amounts of ^{233}U can be extracted during their first 20 years of operation when the incineration of the initial TRUs allows for the saving of most of the ^{233}U produced in the core. For the selected heavy nuclei concentration of 22.5%, the doubling times vary between 30-40 years depending on the reprocessing rate. For ^{233}U-started TMSR-NM operated with a salt with the same concentration, the doubling time is in the range 45 to 50 years. Smaller values are obtained with higher concentrations. It thus appears that a TMSR-NM is able to cope with energy strategies in which the nuclear energy production grows at an annual rate between 1.5 and 2.2%. (a factor 2 to 3 over fifty years).

X CONCLUSION

Within GEN-IV, it appears that the main nuclear energy countries have given the highest priority to the Sodium Fast Reactor concept. This reflects their confidence in a technology which, although it has not yet reached the level of economic and safety performance required for a GEN-IV reactor, has already been tested with several reactors. From that point of view, because of the long time scales typical of nuclear industry, the TMSR-NM may today appear more as a long term or as a fallback option. On the other hand, it may already be appealing to countries which hold important natural thorium resources[26] and choose not to take too early or too restricted commitments in the definition of their nuclear energy policy strategy. As a matter of fact, when it comes to the deployment of GEN-IV reactors, the question of the resource lies much less in the availability of fertile material (whether it is ^{238}U or ^{232}Th) than in that of ^{239}Pu which is the truly useful legacy of GEN-II and GEN-III reactors. In that respect, on the basis of simulations, TMSR-NM is at least in as good position as any other GEN-IV fast reactor.

[26] We have already mentioned that France, although it does not have natural thorium resources, turns out to possess a stock of refined Th on its soil which would cover its energy needs for over a century within the Th-U cycle.

Non-moderated TMSR-NM configurations with high HN proportions (typically 22.5%) present particularly interesting characteristics concerning safety performances. They too can be started with a fuel made from the TRU of the used fuel of today PWRs. The TMSR-NM concept has some attractive features compared to earlier versions of MSRs. The reactor core is simple and simulations do not point to major reprocessing constraints. In particular, the requested reprocessing fluxes should allow a batch mode treatment in the immediate vicinity of the reactor. First studies of the scientific feasibility of the on-line control of the salt composition and of its chemical and physical properties have not uncovered insuperable obstacles.

Despite the rather large initial fissile inventory of a TMSR-NM with 22.5% of heavy nuclei, (in fact not larger than that of a SFR of the same power), its effective ^{233}U breeding allows a deployment rate compatible with the needs of a developed country. The technology which in principle does not require transportation of fissile material outside the reactor site as well as the presence of small quantities of ^{232}U within the fuel are sometimes considered as assets with respect to proliferation risks.

In spite of the vast amount of knowledge accumulated by the ORNL teams over several decades, much work remains to be done on questions relating to reactor design, thermal-hydraulics, processing, material resistance to corrosion, stress, on reprocessing aspects and on safety methodology. In particular, the question of the impact on the long-term behaviour of structures in the presence of impurities in the fuel should be addressed in more detail. It is also necessary to move to a less schematic design of a TMSR-NM reactor in order to firmly establish relevant domains for the research regarding the influence of fluid velocities, temperature gradients within the primary circuit, as well as of thermal and mechanical stresses on the longevity of the structures. The results of thermal-mechanical simulations on a realistic design will help define the experimental and simulation programme within an adequately circumscribed subspace for the reactor operation parameters. The availability of a versatile forced convection molten salt loop also appears to be desirable for an orderly progression of the experimental programme.

To solve the ensemble of questions raised by the physics and chemistry of the TMSR-NM, PACEN is working within as comprehensive and as coherent a planning as is compatible with the CNRS and French universities human and material resources.

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