Molten salts for 4th generation nuclear reactors?

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If we are to combat climate change significantly, massive use of nuclear energy is essential. This requires upstream management of the risks associated with nuclear power and its acceptability to society. Nuclear generation will also have to be able to keep up with the load of a fleet that includes a large proportion of renewable energies.

This article presents and discusses a new approach to meeting these challenges: liquid fuel reactors, or molten salt reactors, which are attracting growing interest. It describes the developments made in France with the CNRS MSFR (molten salt reactor with fast neutron spectrum) concept, which offers prospects for the incineration of transuranics and the management of intermittent energy sources.

Assessment criteria for a nuclear reactor of the future

Three assessment criteria, directly related to the question of the societal acceptability of such an industry, can be defined: safety, waste management and its role in supporting the deployment of intermittent renewable energies.

For the public to accept new nuclear developments, not only must no accident lead to the evacuation of the population, but it must also be made credible to the public by eliminating all risks of over-accidents. It is impossible to prevent an accident from occurring as a result of human error or a natural disaster. It is therefore important that the reactor configuration ensures that such an accident cannot escalate through a domino effect. For example, in a pressurised water reactor (PWR), it would be preferable to make it impossible to produce hydrogen rather than to install systems to absorb the hydrogen produced. More generally, new reactors will have to be intrinsically safe, which will also reduce costs.

It will also be necessary to reduce the amount of radioactive waste produced and the length of time it needs to be stored. To achieve this, it is essential that the only highly radioactive waste should be the least radiotoxic fission products (which are unavoidable), whose activity falls below the fuel's natural activity after only a few hundred years. This requires the fuel to be recycled, which will also make the process more economical. It will therefore be necessary to use one of the two breeder cycles[a], which have the advantage of consuming only one tonne per GW_{el} per year of natural uranium or thorium instead of 150 to 200 tonnes for the current cycle.

Lastly, the equally large-scale deployment of intermittent modes of energy production envisaged by most of the scenarios also means that these new reactors will have a role to play in offsetting intermittency. They will therefore need to be highly flexible in their operation and rapidly dispatchable over a wide power range.

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Scale model (1/6) of the Molten Salt Reactor Experiment (MSRE) at Oakridge (USA). The MSRE diverged on[1] June 1965, and reached full power (7500 kW) in May 1966.

The benefits of liquid fuels

Back in the 1960s, engineers at ORNL (Oak Ridge National Laboratory in the USA) discovered that, irrespective of the type of reactor, the use of liquid fuel as both moderator and coolant had great potential in terms of intrinsic safety and controllability. These fuels have a number of very interesting characteristics.

1- The fission heat is deposited directly in the coolant and transported out of the core in a matter of seconds (for solid fuel, it must first be diffused inside the fuel rods). The disappearance of this delay means that thermal counter-reactions can take effect very early on, which greatly stabilises the reactor. This characteristic also enables rapid power variations.
2- In the event of an accidental increase in reactivity, the power level rises very quickly, causing the fuel to heat up significantly. If the fuel is liquid, it expands and leaves part of the highly neutron-active zone: it then becomes slightly more transparent to neutrons, which reduces reactivity and returns the reactor to its nominal state. Numerical simulations and experiments on a critical liquid fuel reactor such as SILENE [1] have shown that a Chernobyl-type accident is impossible.
3- In an out-of-control situation, the liquid fuel can be drained from the reactor core in a few minutes, without pumps and by simply opening a drain plug. The fuel, which continues to heat up because of its residual power, will then be passively cooled in complete safety in an emergency drain tank. A Fukushima-type situation can then be managed even without operator intervention, with the reactor automatically waiting to be restarted in the weeks or months following the accident.
4- The liquid state of the fuel means that it can be drawn off and added without stopping the reactor. It is therefore possible to ensure that there is no excess reactivity and therefore eliminate any risk associated with this.
5- An accidental increase in reactivity through fuel densification is impossible because liquid fuel is incompressible.
6- The liquid state of the fuel eliminates the need to build and deconstruct assemblies, which can be fragile and complex to cool if the fuel is recycled several times.
7- Minor actinides, currently considered waste, could be used in these fuels, which do not require the manufacture of pellets and assemblies.
8- The fuel is homogenous because it is constantly being stirred as it passes through heat exchangers, eliminating the need for the complex loading plans required for solid fuel.

At present, however, these advantages are only potential: only a judicious choice of liquid type and reactor configuration will enable them to be realised.

Choice of liquid fuel

Regardless of the configuration of the reactor used, a good liquid must meet a number of criteria, ranging from transparency to neutrons to the non-production of troublesome radioisotopes. Chemical properties for heat dissipation, stability in operation, solubility of fuel elements and reprocessing. Ionic salts (fluorides or chlorides) enable these criteria to be met: these are known as Molten Salt Reactors (MSRs).

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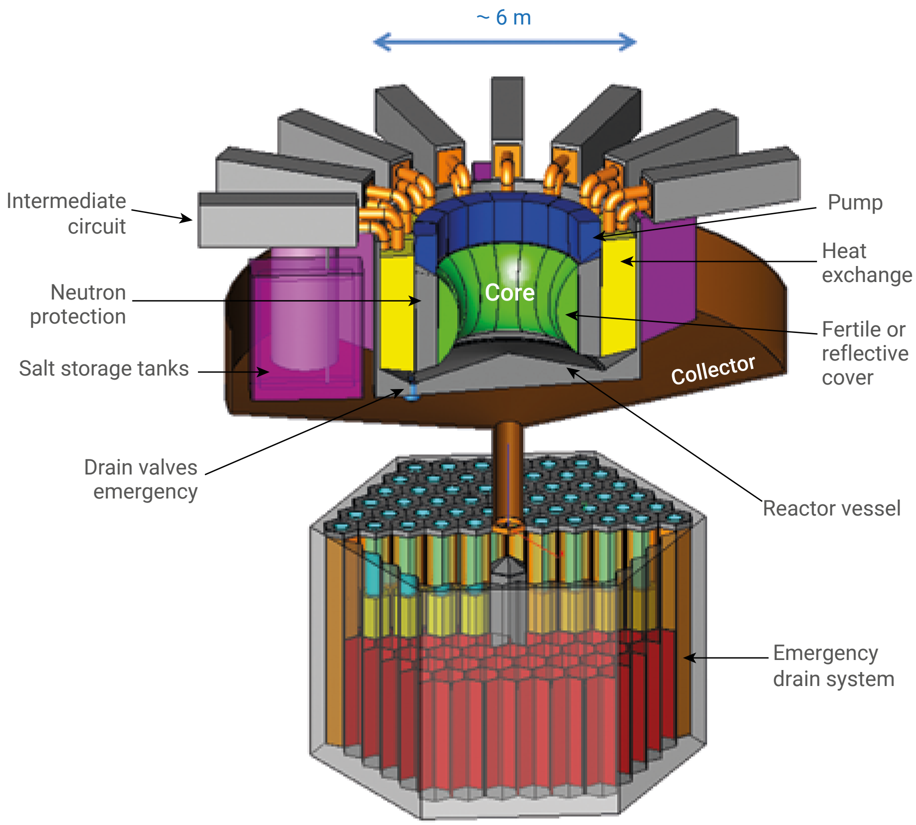

1. MSFR integrated geometry

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Fluorine has several inelastic neutron scattering resonances of a few hundred keV, which greatly reduces the number of neutrons at these energies. As a result, the neutron flux is much less damaging to structural materials, but is highly detrimental to the uranium-plutonium cycle, which needs these energetic neutrons to ensure proper regeneration of fissile nuclei. Fluorides are therefore better suited to the thorium-uranium cycle. In this case, the best solvent is lithium fluoride LiF, with a fusion eutectic at 585°C for around 22% actinides. It is necessary to use lithium enriched to more than 99.9% in ^{7}Li, as the ^{6}Li isotope has a very large neutron capture cross-section and produces too much tritium.

To use chlorides, the ^{37}Cl isotope must be enriched to over 99.9%. Natural chlorine contains 75.77% ^{35}Cl, which produces radioactive ^{36}Cl by neutron capture, with a half-life of 301 000 years, making it impossible to store. These chlorides are well suited to the U-Pu cycle, despite a neutron spectrum that is more damaging to structural materials. The best solvent is sodium chloride NaCl, with 38% actinides and a eutectic melting temperature of around 450°C.

History of molten salt reactors

Two molten salt reactors have already been in operation at ORNL [2]. In 1954, the US Air Force, wishing to have a nuclear-powered aircraft, built a small molten-salt reactor with a thermal power of 2.5 MWth, the Aircraft Reactor Experiment (ARE) was tested on the ground for around a hundred hours. This type of reactor was in fact compact and allowed the wide variations in power over short periods of time required for an aircraft. The test showed that liquid nuclear fuel is relatively simple to use, but that the fluoride salts corroded the structural materials excessively.

ORNL physicists then built the Molten Salt Reactor Experiment (MSRE, see photo p. 99), where the use of a nickel-based alloy, Hastelloy-N, solved the corrosion problems. This reactor operated for five years with a rated power of 8 MWth. It had a thermal neutron energy spectrum, thanks to a graphite matrix in which molten salt circulated. The MSRE used three different types of fissile material: enriched uranium, uranium 233 and plutonium; however, it never contained enough fertile material to regenerate its fuel and was never loaded with thorium. Power variation tests on the MSRE have successfully demonstrated the excellent load-following capabilities of this type of reactor.

The next project, the Molten Salt Breeder Reactor (MSBR), which could produce electricity, was halted at ORNL, but studies were carried out by a CNRS-EDF collaboration in the early 2000s. They demonstrated the unavoidable problems of this concept, with the reprocessing of the fuel and the management of large masses of irradiated graphite. However, the advantages of liquid fuels were still sufficient to justify systematic research into new configurations, using neutron simulations.

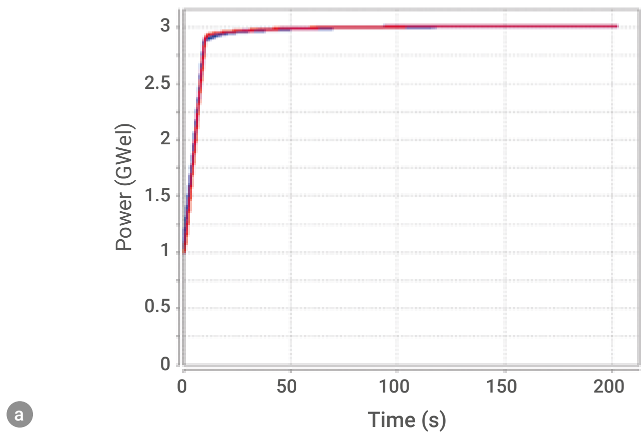
The French approach since 2000

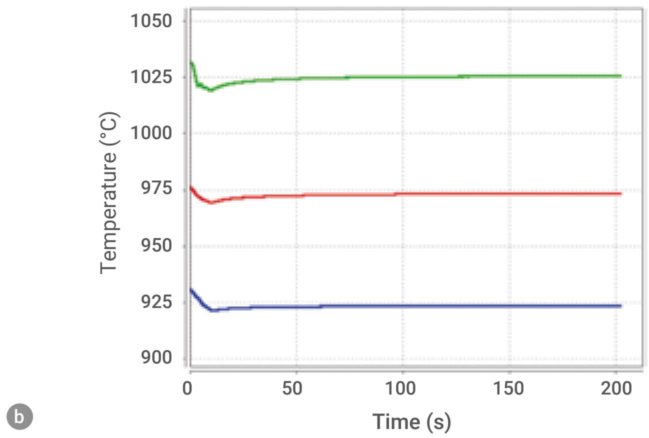
An innovative concept, meeting the criteria of the Generation IV International Forum (GIF) and eliminating graphite from the reactor core, was adopted in 2008 under the name "Molten Salt Fast Reactor" (MSFR) [3, 4]: this is a molten-salt nuclear reactor with a "fast" neutron energy spectrum, because the neutrons are no longer moderated. This configuration, which is largely breeder in terms of fissile material, would have very high operating stability; however, the start-up of each reactor would require a large quantity of fissile nuclei (3600 kg/GW_{el}). Until sufficient fuel can be produced by fast-breeder reactors, the target of 3,000 GW_{el} in thirty years set out in the various scenarios will require the use of uranium enriched to 13% ^{235}U and plutonium from current PWR reactors. It will even be possible, and even preferable, to include in the fuel the minor actinides from the spent fuel of current PWRs *(i.e.* highly radioactive elements in the actinide series other than uranium, plutonium and thorium, produced artificially in small quantities by current reactors). Research work on the MSFR has been supported by Europe as part of the EVOL *(Evaluation and viability of liquid fuel reactors*) project, and since 2015 the safety and behaviour of these reactors in the event of an accident have been the subject of the SAMOFAR *(Safety assessment of molten salt fast reactor*) and SAMOSAFER *(Severe accident modelling and safety assessment for fluid-fuel energy reactors*) projects under the Euratom H2020 programme.

The configuration we are now going to describe (fig. 1) corresponds to an MSFR reactor with a power of 3 GWth in a Th/U cycle using fluorides; lower power versions, as well as the use of the U/Pu cycle with chlorides, are also being studied.

Fuel circuit

Traditionally, in a molten-salt reactor, the liquid fuel leaves the core to pass through heat exchangers and pumps before returning to the core: there is therefore an unacceptable risk of a fuel leak one day. In the MSFR concept, the fuel circuit is entirely confined in a vessel and is not subjected to mechanical stress or excessive temperatures. Easily transportable and manipulable cooling sectors (on the right in Figure 1) are attached to the inside edge of the tank, which contains 18 m^3 of molten salt, including 9 m^3 in the central zone. These sectors include a pump, a heat exchanger, protection limiting the neutron flux on the pump and exchangers, and a neutron reflector or fertile blanket; the atoms making up the latter are transformed into fissile elements under the effect of neutrons. The pump shafts are magnetically driven to avoid penetrating the cover. Another salt, known as "intermediate" salt, enters the vessel to cool the fuel circuit via the heat exchangers, but also the walls in contact with the fuel and the fertile blanket.





2. Load following from 1 GWth **to 3** GWth **in ten seconds.**
(a) Changes in power demand (red curve) and power supply (blue curve below the red curve).
(b) Evolution of fuel salt temperatures at exchanger inlet (green curve), exchanger outlet (blue curve) and average (red curve).

The vessel is closed by a watertight cover containing a neutron reflector to protect the outside. The bottom of the vessel, which also contains a neutron reflector, supports active and passive valves that allow the fuel to be drained in an emergency into a tank designed to contain it in a highly sub-critical geometry, and to passively evacuate the residual power. This draining would not normally have to take place even in the event of an accident: in all the situations currently envisaged, the fuel would remain in the core thanks to passive cooling using the natural convection of the intermediate salt. The heat extracted in this way would be transferred to the reactor building, which is itself cooled by the outside air.

Reactor control

Since the energy produced by fission is deposited directly in the coolant, a liquid fuel reactor can be driven directly by heat extraction, eliminating the need for a neutron-absorbing 'control rod'.

In principle, if the power demand increases, the fuel salt is cooled, which causes an increase in reactivity and therefore an increase in power and finally a return to the initial temperature with greater fission power. This adjustment to external demand takes place in a few seconds, but can lead to undesirable temperature variations in the heat exchangers. The solution is to obtain the power variation by simultaneously modifying the flow rates in the fuel and intermediate circuits. To study the normal and accidental

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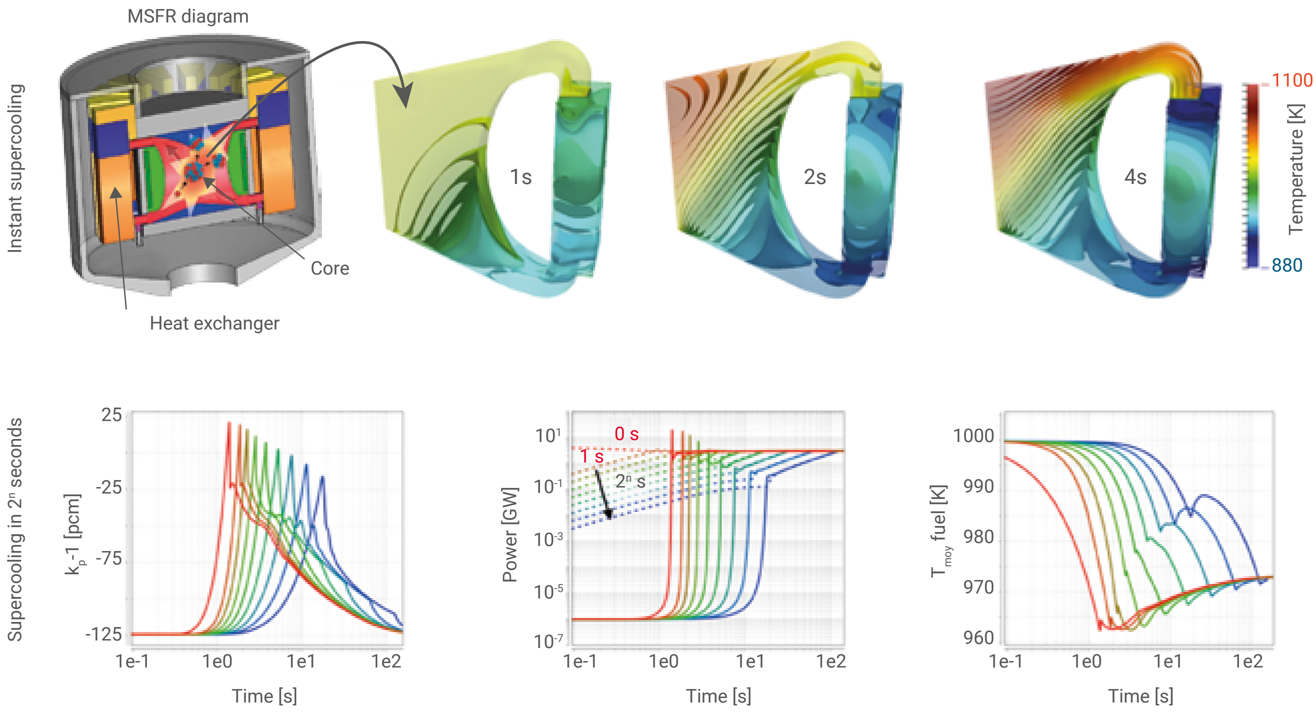

3. Numerical simulation of a supercooling accident with power ranging from 1 kW_{th} to 3 GW_{th}.
Top: evolution of the temperature field in the core and a heat exchanger for an instantaneous transient (the white part is the fertile blanket). Bottom: time variation of the prompt criticality margin k_p-1(which is related to the control of excess neutrons to sustain the chain reaction), power and average fuel temperature for transients lasting from 0 to 128 s (colour-coded) [5].

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operation of the MSFR, the CNRS has developed simulation tools that are unique in the world, taking into account the physical phenomena specific to this type of circulating fuel reactor [MSFR2]. Figure 2 shows the simulation of such load following for a power output increasing from 1 GWth to 3 GWth in ten seconds! This ability to rapidly modify the power produced without affecting the integrity of the reactor would greatly facilitate integration into an energy mix with a high proportion of intermittent renewable energies.

Supercooling accident

The last few years have been devoted mainly to studying the safety of the MSFR and the possibility of serious accidents leading to a massive release of radioactivity into the environment. Such accidents have not yet been identified: the most worrying situation that has come to light is a massive and sudden over-cooling of the fuel in the case of a new fuel salt producing only 1 kW_{th} of fission through spontaneous fissions. This salt is assumed to be in nominal circulation, while the intermediate salt is shut down. Following an operator error, the intermediate salt is put into nominal circulation in a very short time. Figure 3 above shows the evolution of the temperature field under the unrealistic assumption of an instantaneous transient. After four seconds, the temperatures have stabilised and the power supplied is 3 GW_{th}. The bottom left of Figure 3 shows the variation over time of the margin to prompt criticality(b) for different durations of this supercooling. If the transient is short enough, the reactor exceeds the prompt criticality threshold. If the fuel were solid, this situation would lead to its destruction, which is what happened at Chernobyl. The centre shows the instantaneous fission power in the core. There is a power peak of up to 20 GW_{th} when the transient lasts less than ten seconds. However, on the right, we can see that the effect on temperatures is small enough for there to be no mechanical effect: there is no violent change in the behaviour of the core that could lead to a nuclear risk.

Prospects

Work on molten salt reactors is booming, not only in research centres (sixteen countries have been taking part in meetings organised by the IAEA on this technology since 2016), but also through *start-ups* all over the world. In France, industrial companies such as Framatome, ORANO and CORYS have started to take an interest in this research since 2018, after some twenty years in the academic world. Recently, an annual national workshop on the subject of molten salt reactors has been organised by the CEA and the

CNRS [6]; in 2022, this workshop brought together 150 participants, demonstrating the growing interest in France in this type of reactor.

As is rightly mentioned time and time again, current nuclear technology is leading to the accumulation of "long-lived radioelements", which are considered to be waste, and which will remain dangerous for millennia. In reality, what is waste today could become fuel by changing the type of nuclear reactor. Studies are now underway at European level on intrinsically safe molten salt reactor concepts, such as the MIMOSA project for "Multi-recycling strategies of spent nuclear fuel focusing on molten salt technology" under the Horizon Europe programme, led by the industrial company ORANO. In this context, the national ISAC project ("Innovative solution for actinide recycling") financed by the public investment bank BPI as part of the recovery plan, was launched in 2022 with the CEA, CNRS, EDF, Framatome and ORANO as partners. The aim is to carry out pre-design studies for a molten-salt reactor that would incinerate plutonium and americium.

Molten salt reactors can also be designed to be dispatchable and to support intermittent sources. This type of nuclear reactor would therefore have an additional advantage and could support the development of renewable energies.

Finally, some of the new reactors under study are *Small Modular Reactors* (SMRs), which are seen as nuclear cells, with reduced size and power so that they can be manufactured in a factory and transported to their place of use [7]. Such small molten-salt reactors would be very compact, with cores of a few hundred litres that could be transported on a lorry and installed in *plug-and-play* mode on the user's premises to produce electricity or process heat, while burning nuclear waste. This would make it possible to replace thousands of diesel engines with equipment that emits no greenhouse gases. French *start-up* NAAREA has been positioning itself in this type of niche for the past three years, with the development of the XSMR, a molten salt microreactor capable of generating several dozen megawatts, and the aim of building a demonstrator in the next few years. The CEA is also in the process of setting up a spin-off company: in 2023, the start-up Stellaria will have developed a molten salt reactor concept with a capacity of several hundred megawatts. The France 2030 national investment plan launched in 2022 by the French President includes a funding component for start-ups developing such small modular reactors, in order to give a French boost to this area of R&D. ▮

(a) There are two fast breeder reactor cycles: the uranium-plutonium cycle, in which the fissile isotope is plutonium 239 and the fertile isotope is uranium 238 (transmuted in the reactor into fissile plutonium 239); and the thorium-uranium cycle, in which the fissile isotope is uranium 233 and the fertile isotope is thorium 232.

(b) A critical nuclear reactor can be driven by its delayed neutrons. When it becomes critical without requiring the contribution of delayed neutrons, it is no longer controllable. The margin to prompt criticality is the reactivity margin before it is no longer controllable.

“What is waste today could become fuel by changing the type of nuclear reactor.”

1• F.Y. Barbry, "A review of the Silene criticality excursions experiments", Rapport technique, CEA Centre d'Études de Fontenay-aux-Roses, 1993.

2• P.N. Haubenreich et J.R. Engel, "Experience with the molten salt reactor experiment", *Nucl. Appl. Technol.* **8** (1970) 107–117.

3• “Status of Molten Salt Reactor Technology”, Technical Report Series no. **489** (2021), International Atomic Energy Agency, Vienne. https://cutt.ly/preprint-iaea

4• M. Allibert *et al*., “Chapter 7 - Homogeneous Molten Salt Reactors (MSRs): The Molten Salt Fast Reactor (MSFR) concept”, *Handbook of Generation IV Nuclear Reactors*, Second Edition, Elsevier Science Publishing (2023).

5• A. Laureau *et al*., “Transient coupled calculations of the Molten Salt Fast Reactor using the Transient Fission Matrix approach”, *Nuclear Engineering and Design*, **316** (2017) 112-124.

6• https://cutt.ly/youtube_1

7• « Les réacteurs nucléaires modulaires de faible puissance (SMR) : état des lieux et perspectives », Avis et Rapport du Comité de Prospective en Énergie de l'Académie des sciences (octobre 2022).