# **High-temperature** reactors: a recent past, a near future

While high-temperature reactors did experience major developments in the past, in Europe in particular, significant R&D efforts are required, if – a major innovation – deployment is to be made possible, of modular reactors having the capability of being coupled, in reliable, economic fashion, to an industrial process. The aim? The construction, before the next decade is out – more swiftly than is feasible for other fourth-generation systems – of an industrial prototype, coupled to such a process. The Areva Group takes up this approach, with its ANTARES project.

The basic technologies for high-temperature reactors (HTRs) were developed over the period extending from the 1960s to the 1980s. A number of experimental reactors were built: Dragon in the United Kingdom, AVR in Germany, Peach Bottom in the United States. Industrial power reactors were built, and operated in the 1970s and 1980s: Fort Saint Vrain in the United States, and THTR 300 (Thorium High-Temperature Reactor) in Germany.

These reactors shared a number of common characteristics. This reactor line employs **helium** as **coolant**, and **graphite** as **moderator**. The reactors use a specific **fuel** technology, in the form of particles, encapsulated in several coating layers. They take advantage of the high heat capacity of the graphite used for the **core**, and **reflectors**; of the high-temperature characteristics of core components; the stability, and chemical inertness of the coolant fluid, fuel, and moderator; the high **fission-product** retention capacity exhibited by the fuel coatings; the properties of the coolant helium, exhibiting no phase change; and, finally, a negative core **temperature coefficient**.

## Modular high-temperature reactors

A new generation of high-temperature reactors was developed in the 1980s, both by the German firm Siemens-Interatom (now trading as Areva NP), and US corporation General Atomics. These are so-called modular reactors - which do, obviously, exhibit the general characteristics of HTRs. Their specific feature is their use, for decay heat removal, of conduction, and thermal radiation, from the metallic vessel to an externally mounted cooling circuit - which may be of the passive kind - while not exceeding the temperature beyond which the particle fuel loses its integrity. This is achieved, on the one hand, by restricting core overall power, and volumetric power, and, on the other hand, by going for a reactor configuration such as to ensure that natural decay heat removal processes retain the ability, in an accident situation, to restrict fuel temperature to levels for which there is no significant release of fission pro-



ducts from the reactor to the environment. The low **power density** exhibited by such reactors, together with the desire to effect decay heat removal through a passive vessel thermal radiation process entail a necessarily considerable vessel size.

These design choices, with respect to fuel, core, coolant fluid, and vessel give modular HTRs outstanding safety characteristics. Their natural behavior, in the event of an accident, does not result in any signifiCVD (chemical vapor deposition) station in the GAIA facility, at Cadarache, which allows  $UO_2$  balls to be obtained, with their various ceramic coating layers.



Figure 1. Sectioned view of a TRISO particle.

cant consequences, allowing requirements for safeguard systems to be limited. Further, in order to enhance reliability, the functioning of such systems is, as far as feasible, passive. Calls for corrective action are thus minimized, and the reactor's thermal inertia gives time to implement them.

Two projects exemplifying this philosophy were developed during the 1980s: the German HTR Modul reactor, and the US MHTGR (Modular High-Temperature Gas-Cooled Reactor). The designers submitted these projects for scrutiny by their respective nuclear safety authorities.

## **Design principles**

### Fuel

The design selected for the **fissile** particles is that of a spherical particle (a few hundred **microns** in diameter), of the **TRISO** type (see Figure 1), comprising a kernel of **uranium** fuel (uranium oxide [**UO**<sub>2</sub>], or in some cases uranium **carbide** [UC<sub>2</sub>], or a mixture of oxide and carbide: oxycarbide [UCO]), enriched to less than 20%, possibly also containing **plutonium** (mixed oxide, or 100% PuO<sub>2</sub>). The particle delivers power, however it further ensures the function of retaining its own fission products. The kernel is contained in a sandwich, comprising three

TRISO HTR fuel balls, fabricated in the UO<sub>2</sub> Laboratory's GAIA facility, at CEA's Cadarache Center.



layers of dense ceramic materials: one layer of silicon carbide (SiC), held between two layers of dense pyrocarbon (PyC). The kernel and containment layers are separated by a buffer, made of highly porous pyrocarbon, the role of which is to shield the dense layers from fission product recoil, provide a plenum to accommodate gases released by the kernel (fission gases, and carbon monoxide), and ensure mechanical separation between the kernel, and dense layers, thus limiting the effects, for the latter, of kernel irradiation swelling. The intermediate SiC layer fulfills an essential function, of fission product containment, further ensured by the contribution from the two dense pyrocarbon layers, which also play a crucial part, in delaying the onset of tensile stress in the SiC layer, due to the effects of rising internal pressure, from fission gases and CO accumulating inside the buffer – and thus allow high irradiation rates to be achieved. In the case of fertile particles, the amount of fission products to be contained being smaller, a single dense pyrocarbon layer is as a rule adequate, rather than the "sandwich" configuration (so-called BISO particles).

A reactor core holds several billion such particles. Mixed with graphite powder, particles are assembled, by compaction, into compacts, as in the ANTARES project (see below), or pebbles, as in German reactors, or in the PBMR (Pebble-Bed Modular Reactor) project.

Provided they are subjected to adequate dimensioning, fabrication, control, and employment, TRISO particles exhibit the remarkable property of not releasing their fission products, up to very high **burnup** values, and temperatures of about 1,800 °C (beyond which point, the SiC layer loses its stability). The reactor is so dimensioned (power density, core size, operating temperature, etc.) as to ensure, in all and any situation – whether normal or accidental – conditions allowing the fuel particles' containment function to be maintained.

### The various reactor configurations

Conditioning the particles into pebbles, or compacts entails different reactor technologies. In pebble-bed reactors, the fuel is charged loose into the core cavity, the coolant helium being circulated across the pebble bed. Pebbles are taken out at the base of the reactor on a continuous basis, being reintroduced at the top, or replaced by new pebbles, if they have achieved *discharge burnup*. Compacts, on the other hand, are inserted into channels in hexagonal, prismatic graphite blocks, while the coolant helium circulates in separate, specific channels. The reactor core then consists of a stack of such blocks, being reloaded periodically.

The energy conversion part equally results in two distinct technologies, the so-called **direct cycle**, or the **indirect cycle** (see Focus C, *Thermodynamic cycles and energy conversion*, p. 23). In direct-cycle reactors, the coolant helium passes directly through a turbine engine, driving the alternator, this involving no exchange with a **secondary circuit**. In indirect-cycle reactors, the coolant helium transfers its heat to an intermediate circuit, by way of an intermediate heat exchanger, which may be either a **steam generator**, or a gas-to-gas heat exchanger (see *Gas*-

technology energy conversion: common ground for the new fast reactors and [V]HTRs, p. 91).

### **Current programs**

In the 1980s, development of high-temperature reactors was driven by process heat supply applications, and **cogeneration** capabilities, for which modular HTRs proved suitable. This was one of the considerations that led **JAERI**, in Japan, and **INET**, in China, to build small experimental reactors: HTTR (High-Temperature Engineering Test Reactor), and HTR-10, respectively.

In the early 1990s, the preferred option, for hightemperature gas-cooled reactor (HTGR) electricity production, switched away from the steam cycle, to a gas turbine system (**Brayton cycle**). This change resulted from the advances achieved in the field of gas turbine technology (see Focus C, *Thermodynamic cycles and energy conversion*, p. 23).

The following paragraphs provide an overview of the main concepts, or projects currently undergoing development, or trials, whether they concern experimental reactors (in China, and Japan), power reactor projects, such as the direct-cycle reactors developed in South Africa and Japan (PBMR, and GTHTR-300, respectively), or indirect-cycle projects, as developed in China, and France (HTR–PM, and ANTARES, respectively), along with the NGNP program, developed in the United States.

### **Experimental reactors**

The two experimental reactors, HTTR and HTR-10, allowing as they do temperatures to be achieved, at the core outlet, of 950 °C and 900 °C, respectively, are to be used to support research and development activities, to validate safety principles for this type of reactor, together with gas-turbine electricity generation performance, and uses of heat for high-temperature industrial applications.

#### Modular power reactors

The PBMR has been seen, from 1993 on, by South African power utility **Eskom**, as a way of boosting their electricity generation capacity. A technical and economic viability study to that effect was completed in early 1997. Development for this pebble-bed reactor takes as reference the HTR Modul, developed in Germany during the 1980s. PBMR (Pty) Ltd corporation is currently carrying out design studies for such a modular nuclear power reactor, coupled to a direct-cycle gas turbine, i.e. one directly fed with primary helium (see Figure 2).

A number of concepts involving high-temperature modular reactors, coupled to a gas turbine, are being developed in Japan, work on such projects being coordinated with the HTTR program. One of these projects, GTHTR-300, is a 600-MWth reactor, using a direct-cycle gas turbine (see Figure 3). One of its specific features is the presence of three main vessels: one for the core, which is made up from hexagonal blocks; one for the turbine engine; and one for the heat exchangers involved in the direct cycle.

The Chinese HTR–PM project, for its part, draws on the experience gained with the HTR-10 program. The aim is to build a pebble-bed reactor, delivering



power of several hundred MWth, coupled to a steam cycle.

In the United States, **DOE** has initiated the NGNP (Next-Generation Nuclear Plant) program, aiming to build a reactor demonstrating HTR capability, as regards providing electricity at high efficiency, and ability to be coupled to **hydrogen** production processes involving no  $CO_2$  emissions, whether by high-temperature water **electrolysis**, or **thermochemical** water splitting.

Figure 2. The PBMR (Pebble-Bed Modular Reactor), a modular, direct-cycle HTR developed by Eskom, in South Africa. PBMR (Pty) Ltd.

## What is the market for the HTR/VHTR? What is the product for this market?

Among fourth-generation systems, the HTR/VHTR exhibits one highly specific characteristic: beyond mere electricity production, it has the ability to provide heat for industrial processes, at a wide range of temperatures, up to 800 °C or so with current technologies and materials, probably beyond this in the longer term (see *Towards nuclear energy applications other than electricity production*, p. 123). High tem-



### Figure 3.

Mockup of the Japanese GTHTR-300C project for a cogeneration plant, showing, on the left, the hydrogen production plant, and (right) the nuclear island, with its 600-MWth reactor (shown in brown), the intermediate heat exchanger (gray), the direct-cycle gas turbine (green), and the isolation valves (blue). The core of this reactor is made up from hexagonal graphite blocks.





The HTTR (High-Temperature Engineering Test Reactor), a 30-MWth modular high-temperature reactor, developed by JAEA (formerly JAERI), in Japan, went critical at the end of 1998. This reactor should ultimately be coupled to a hydrogen production plant.

> perature provides it with a first advantage, by enabling it to make inroads into the process heat market, though the major simplification it allows, as regards safety design, at power levels limited to a few hundred megawatts. The modular concept, adopted by all recent HTR/VHTR projects, further opens the way to development of competitive, medium-power reactors, better suited than large electricity production reactors to the energy requirements of an industrial platform, or small-scale electric power grids. The inherent safety characteristics of modular HTRs are of further advantage, with respect to the acceptability of this type of reactor.

Figure 4. General view of an ANTARES system, a multi-application, high-temperature heat source, being investigated by Areva NP, showing the nuclear reactor (in purple), the intermediate heat exchanger (blue), and the energy conversion unit, to the right. At the same time, at a time when oil and natural gas prices have become highly volatile, the cost stability exhibited by heat generated by a nuclear reactor turns into a further advantage yet. Contrary to a widely held belief, the breakthrough of HTRs/VHTRs into the industrial market is thus in no way bound up with the hypothetical emergence, in the longer term, of a "hydrogen civilization." HTRs could find, as of now, their place in a fast-expanding process heat market, provided a credible offer can be put on the table.



pled to an industrial process. What product should be developed? The temperature range involved in uses of process heat is quite large (see Figure 1, in *Towards nuclear energy applications other than electricity production*, p. 123). However, since the issue is that of meeting market requirements at the earliest possible time, there is no point in targeting, initially, very-high-temperature - beyond 800 °C - applications (VHTR), which may only be contemplated at the outcome of protracted development work, on innovative materials, and fuels. As regards catering for an application at the earliest date, an altogether adequate response will be to meet the challenge, which has never been taken up, of coupling the reactor with a heat generation application involving a more reasonable temperature level. At the same time, whereas electricity production is effected through coupling of a standard turbine engine to the reactor, industrial processes that may be considered, to be coupled to the reactor, are many, and exhibit diverse characteristics (power and temperature levels). The reactor must thus be sufficiently flexible to adjust to a variety of situations. Moreover, as is the case with natural gas-fired plants, the expectation will be that, even as it provides heat, the reactor should go on supplying electricity, in cogeneration mode, to provide an all-in-one energy supply solution for an industrial platform, or, as the case may be, be wholly dedicated to electricity production, depending on local power grid requirements.

## The ANTARES program

In 2004, Areva NP launched the ANTARES (Areva New Technology based on Advanced gas-cooled Reactor for Energy Supply) program, with a fourfold goal: competitiveness, with regard to capital cost and energy output; flexibility to adjust to a variety of industrial applications; making optimum use of the inherent safety characteristics of modular HTRs, to simplify system design; and the ability to be put on the market at the earliest possible date. All of which entails the construction, in the shortest possible time, of a prototype industrial-scale reactor, coupled to an industrial heat generation application. The design choices for ANTARES (see Figure 4) stem from these goals:

To meet the requirement for flexibility to adjust to a variety of industrial requirements, an indirect cycle was selected, allowing a relative uncoupling of the boiler (dedicated to generation of nuclear energy; removal of that energy from the reactor core; and containment of **radioactivity**), and the secondary



circuit, which, as shown in Figure 5, may be adjusted, to suit various electricity and/or heat production applications, involving diverse temperature levels, with no major impact on boiler design. Such uncoupling, together with the use, in the secondary circuit, of a coolant exhibiting properties close to those of air, make it possible to rely, for the secondary circuit architecture, and components, on proven, nonnuclear industrial technologies.

# The intermediate heat exchanger: a key component

Out of a number of major components (see below), the key component, with respect to the indirect cycle, is the intermediate heat exchanger (IHX). Going for a compact design, with a single primary **loop**, and a single intermediate heat exchanger, was the preferred option (see Figure 6), on economic grounds. As the aim is to ensure the exchange of several hundred megawatts by way of this device, which is to be set up in a vessel of the same type as that enclosing the reactor, the technology being considered is that of *plate heat exchangers*, allowing as this does greater compactness than tube heat exchanger technology (see Gas-technology energy conversion: common ground for the new fast reactors and [V]HTRs, p. 91). A number of variations on this technology are being investigated (see Figure 7), however, inasmuch as use of a plate heat exchanger for a nuclear application, on the scale and at the temperatures being contemplated, presents a major technological challenge, a fallback, multiple-loop solution, involving several tube heat exchangers, is being kept in hand.

## Safety, and economics

The reactor design systematically puts to advantage the inherent safety characteristics exhibited by this type of reactor, and its fuel, to simplify system design, with respect to safety features. In particular, the core is designed in such a manner as to allow, in an accident situation, the **chain reaction** to be brought to a stop through the effects of thermal **feedback**, and enable decay heat removal on the basis of simple,



Figure 6.

ANTARES – Conception compacte à un seul échangeur intermédiaire.



#### Figure 5.

Principle schematic of the ANTARES project, designed by Areva NP. Potential applications range from hydrogen production to desalination, and heating, through coal liquefaction, or gasification, treatment of tar sands, and biomass processing.

well-controlled physical processes (conduction, radiation...).

TRISO SiC coated-particle technology, shared as it is by all modern HTR projects, was naturally adopted for ANTARES, along with uranium (in the form of  $UO_2$ ) as the fissile material. To maximize reactor power (~ 600 MWth), the option of a core made up of hexagonal blocks was selected, rather than the pebble-bed option. Particles are held together in small graphite cylinders, about 50 mm high, for a diameter of some 12 mm – the *compacts* – each holding approximately 2,000–6,000 particles. The compacts in turn are inserted into the channels in the hexagonal blocks, which are positioned in an annular configuration, thus forming the reactor core.

The economic optimum, for the fuel cycle, will certainly correspond to a high discharge burnup value, probably lying around 150 GW · d/tHM, provided the demonstration can be made, that the fuel retains, right up to that point, its outstanding ability to contain its own fission products - something that still lies outside the past experience achieved with HTR fuel. To minimize development leadtimes, only materials that are already being manufactured, and have seen use on an industrial basis are eligible for consideration. In particular, for the reactor vessel, the reference solution is a modified 9Cr1Mo steel grade (with, as fallback solution, the steel used for pressurized-water reactor vessels); and, for the intermediate heat exchanger, a nickel-base alloy. The choice of these materials will determine the maximum operating temperatures for the reactor: 400-450 °C (350 °C for the fallback solution) at the core inlet, about 850 °C at the outlet.



Figure 7. Intermediate heat exchanger concept, of Alfa-Laval technology, investigated for the ANTARES project.





Fort Saint Vrain, the second US HTR after Peach Bottom, built from 1968, went critical at the end of 1974, operating till 1989. In this reactor, fuel particles are held together in a carbon matrix, to form "compacts," which are inserted into hexagonal graphite blocks, making up the core.

### The main goals for the associated R&D

Even though HTR reactor technology has experienced major developments in the past, in Europe in particular, with the construction, and operation of industrial prototypes, significant R&D efforts are required, if modern modular reactors are to be deployed.

Since no reactor, in the past, has ever been coupled to an industrial process, a large-scale demonstration is required, to demonstrate the viability of such a coupling, and convince industry that it may be implemented effectively. Ensuring that such a major innovation is feasible, and reliable thus needs must stand as the major goal for R&D, as regards HTRs.

#### The other components

Other components, aside from the intermediate heat exchanger, lie outside the realm of extant industrial experience, such as the helium circulators for the pri-



The Chinese HTR-10 experimental reactor, built at Tsinghua University, near Beijing. mary circuit, the large helium valves, and the ducts to channel hot gases.

All these components have to undergo trials, first in order to select the options (e.g. to select the plate concepts under consideration for the IHX), and then to qualify the solution thus selected. Tests to point the design to the most appropriate solution may be carried out on small mockups, in an air environment. However, final qualification must be carried out in large test loops, with representative, large-scale mockups, under representative conditions, with respect to temperatures, flow rates, pressure, and chemical environment (see Gas-technology energy conversion: common ground for the new fast reactors and [V]HTRs, p. 91). For the ANTARES program, IHX concept selection will be achieved through trials in CEA's CLAIRE air loop, at Grenoble, under low-flowrate, low-temperature conditions; in EDF's PAT air loop, at Chatou, for high-flow-rate, low-temperature conditions; and **ENEA**'s HEFUS 3 helium loop, at Brasimone (Italy), under low-flow-rate and medium-temperature conditions, or in the future HELOKA loop, to be set up at Forschungszentrum Karlsruhe. Following this, choice of concept will be validated in the 1-MW HELITE helium loop, currently at the definition stage, at CEA/Cadarache (see Figure 8). To achieve final qualification, a larger facility, of at least 10 MW power, will be required.

### Materials: a critical point

Materials selection, and qualification stand as a critical point, with respect to component development, not only for the IHX, but equally for the vessels, and internal structures: even though existing industrial materials are to be preferred, they will be employed outside their usual domain, in terms of temperatures, and chemical environment. Past experience with HTRs is inadequate, even though the AVR experimental reactor did see operation, over an extended period, at high temperature (950 °C). The reactor's metallic components were held at much lower temperatures, conditions that cannot apply, as regards an IHX kept at core outlet temperature (for past HTRs, maximum temperature stood at around

700 °C). Much further data, with respect to materials mechanical properties, will have to be collected, for the purposes of component design. Moreover, the unavoidable presence of impurities in the helium (CO, CO<sub>2</sub>, H<sub>2</sub>O, H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>...), in however small amounts, may have a very strong, negative impact on the mechanical properties of metallic materials, e.g. through carburization/decarburization reactions, in the absence of sufficient amounts of oxygen, able to form protective oxide coatings (see Metallic materials, one of the keys for the fourth generation, p. 71). Such processes prove particularly critical, as regards the very thin walls ensuring heat transfer in the IHX. The IHX walls' behavior, with respect to the secondary fluid environment (a mixture of helium and nitrogen), also has to be investigated.

Thus, an extensive program of empirical investigations on the interactions of high-temperature materials with their specific environment is to be initiated, to select the materials most suited to operations in an HTR environment. The composition of these materials must be adjusted, to enhance corrosion resistance; and acceptable proportions have to be specified, with respect to helium impurities (to be controlled, inside the reactor, by the helium purification system), to minimize the impact of such impurities on materials behavior. These tests are to be carried out in specific helium loops, in a precisely controlled atmosphere. A number of such loops are currently being used, in concerted fashion (e.g. the CORINTH loop, at CEA/Saclay), or are being developed, at CEA, EDF, or Areva.

It should also be said that past experience with various types of gas-cooled, graphite-moderated reactors is not directly applicable, for the design of future HTRs, since the graphite grades used in the past are no longer available on an industrial basis. All of the characterizations, all the tests carried out to determine the graphite's behavior under irradiation, in an oxidizing atmosphere (under normal, or accident conditions), have to be carried out anew, to select the most suitable grades, out of those that are currently available, and to collect, for design purposes, a full set of data, for the grades selected.

## Fuel: fabrication, and qualification

A further major challenge, set by modern modular HTR development, is that of fuel development. Fuel for the German HTR-Modul reactor was qualified for operation at 700 °C, and 80 GW  $\cdot$  d/tHM. In such operating conditions, it was shown that, in the event of accidental heating up, right up to 1,600 °C, no unacceptable release of **radionuclides** occurred. A large number of experiments provide evidence there are margins, extending beyond the HTR-Modul fue-l's qualification domain, such as to allow contemplating harsher operating conditions.

For ANTARES, and the current HTR programs, required performance levels include higher temperatures (850 °C), and, if possible, higher burnups (150 GW  $\cdot$  d/tHM) than those achieved in past programs.

In the early 2000s, CEA worked on recovering the groundwork, underpinning that experience, and on small-scale fabrication process development: production of UO2 kernels, on the laboratory-bench



experiment scale, and of coating layers, in a CVD (chemical vapor deposition) furnace, on simulation kernels. The preliminary stage allowed kernels, and coating layers to be obtained, that met initial specifications, as set out on the basis of German experience. On this basis, CEA and Areva NP put into service, in 2005, a laboratory fabrication line, CAPRI, also catering for compact fabrication.

Presently, the CAPRI line has produced its first TRISO particles, featuring a **depleted uranium** kernel, in its facilities sited at CEA/Cadarache, together with its first compacts, holding simulation particles, at the compaction development station run by CERCA, a Romans (France)-based subsidiary of Areva NP. The aim is to achieve fabrication of fuel at least equal, in terms of quality, to that of the German fuel; and to verify it can achieve, while maintaining its integrity, target performance levels, set higher than what has been demonstrated in the past, and, if required, secure improvements.

These technologies will have to be mastered on an industrial scale, and possibly further elaborated, if the high-quality fuel is to be produced, that is required for such operating conditions. A significant program, covering irradiations, post-irradiation investigations, and temperature rise tests will have to be carried out, using dedicated experimental facilities, allowing online measurement of fission product release, to ascertain fuel performance, and qualify it for industrial operation, prior to licensing it. The European Union's JRC (Joint Research Center) irradiation facility, in the HFR (High-Flux Reactor), at Petten (Netherlands), used for irradiations carried out under the aegis of **FP5** and FP6, together with Figure 8. The HELITE loop, due to be built at CEA/Cadarache (power 1 MW; flow rate 0.4 kg/s; temperature 850 °C).

Haynes 230 alloy test sample, before (mirror finish, left), and after exposure to a 900 °C helium stream, in the presence of impurities, in the CORINTH facility for the investigation of impurities in helium, at CEA/Saclay. The test evidences uniform sample corrosion.







AVR, the first German HTR, built from 1961 at the Jülich research center, went critical in 1966, operating till 1988. In this reactor, fuel particles are conditioned in spherical graphite matrixes some 6 cm in diameter, known as pebbles.

the irradiation device being developed for the OSI-RIS reactor, at CEA/Saclay, are indispensable tools, as regards completing this program (see Figure 9).

### **Computation codes**

Finally, its has been shown, over the past ten years, that there was a major requirement as regards modeling, and the qualification of existing computing resources for the purposes of HTR design, and certification (see Box). The neutronics benchmark drawn up by IAEA has highlighted the fact that most of the tools used by participants in the scheme did not yield correct predictions as to initial criticality, for the HTTR and HT-10 reactors. New codes will have to be qualified, and this will probably require a specific critical experiment. The IAEA benchmark for fuel codes further showed that the phenomenological laws, used in the various codes, should also be reconsidered. These codes will have to be qualified on the basis of fuel irradiation trials, and tests in accident conditions.

Likewise, CFD (computational fluid dynamics) codes will have to be qualified by means of tests on representative mockups, in particular for the computation of conditions critical for HTR design (mixing

# Characterization, and modeling of fuel particle thermal properties

For the purposes of characterizing the thermal properties of heterogeneous, multiscale materials, as a function of temperature, experimental and numerical instruments have been developed at the Microstructure and Behavior Laboratory (Laboratoire microstructure et comportement), at CEA's Le Ripault Center. They have been applied to the thermal characterization of the various layers in a hightemperature reactor (HTR) fuel ball.

To ascertain thermal diffusivity<sup>(1)</sup> at various scales, so-called photothermal techniques are employed. These allow the measurement, and analysis of the periodic rise in a material's surface temperature, induced by absorption of a focused laser beam, intensity-modulated at a selected frequency. Two devices have been developed. around two of the processes generated by local heating in the material. The first device is a photoreflection microscope (PM), measuring, on the scale of a few micrometers, variations in the reflection coefficient due to changes in the refraction index, which is temperature dependent. For that purpose, the light flux from a second laser is collected, by means of a photodiode, after reflection on the surface of the sample that is heated by a first laser. The resulting signal is proportional to the local temperature variation. The

(1) Thermal diffusivity: the velocity at which a thermal wave penetrates into a medium: it is equal to the ratio (expressed in m<sup>2</sup>/s) of conductivity, over the product of density by specific heat.

second device is an *infrared microscope* (IRM), measuring, on a scale of a few hundred micrometers, the modulated component of the *infrared radiation* emitted by the surface. The signal obtained is again proportional to the local temperature variation.

An independently controlled heating stage allows the sample to be heated to 1,500 °C, making it possible to plot thermal diffusivity as a function of temperature.

The complex materials subjected to laboratory investigations entail the development, on the one hand, of analytical models, to take thermal and/or structural **anisotropy** effects into account, in order to allow thermal diffusivity values to be extracted; and, on the other hand, of **computation codes**, for the homogenization of thermal properties in heterogeneous materials, on the basis of photographs, or tomographs of their microstructure, taking as input data the values for the thermal properties of the basic constituents of these media, as measured by PM, or IRM.

The particles investigated (see Figure 1, in *High-temperature reactors: a recent past, a near future*, p. 51) comprise a  $ZrO_2$  kernel (acting as a phantom for  $UO_2$ ); a layer of porous pyrocarbon (PyC: ~ 90  $\mu$ m thick; also known as the buffer); a layer of dense PyC (IPyC: ~ 35  $\mu$ m thick); a layer of sili-



Figure 1 a) 10 × 10  $\mu$ m<sup>2</sup> mapping of thermal signal phase, measured at 300 kHz on the buffer skeleton (the scale is graduated in degrees of angular variation). b) Fit of isophases selected for the determination. evidencing, by their circular shape, the skeleton's thermal isotropy, and allowing thermal diffusivity to be estimated, in this case at  $5 \cdot 10^{-6} \text{ m}^2/\text{s}$ .

occurring in the core lower **plenum**, computation of core bypasses, flow-rate distribution in the IHX collectors...). Finally, an effort will have to be directed at computation resources to address radiocontaminant transport, and the computation of complex situations, involving e.g. a coupling of graphite oxidation models, and natural circulation models.

# Public support, and international cooperation

Owing to the level of capital outlay required, the R&D support it calls for, and the risks of the project, the development, construction, and commissioning, before the end of the next decade, of an HTR demonstrator, coupled to an industrial heat-using process will entail major public funding support, and international cooperation. That initial demonstration is the key to making the emergence of HTRs on an industrial scale feasible, within timescales that may be shorter than is the case for most other Generation IV systems.

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Figure 9. The HTR irradiation device, being developed for the OSIRIS reactor, at CEA/Saclay.



The photoreflection microscope (above) and infrared microscope (at right) used by the Microstructure and Behavior Laboratory (Laboratoire microstructure et comportement), at CEA/Le Ripault, to characterize the thermal properties of HTR fuel particles

con carbide (SiC: ~  $30 \ \mu m$  thick); and a layer of dense PyC (OPyc: ~  $35 \ \mu m$  thick). For the dense layers (<  $40 \ \mu m$  thickness), only PM measurements may be contemplated. Owing to the structure of these materials, the measurements made yield representative values for the entire layer.

On the other hand, the two photo-

thermal microscopy benches do not allow thermal diffusivity measurements to be made of the buffer as a whole. Indeed, the extent of the region subjected to PM probing is restricted, by the requirement to ensure an adequate signal-to-noise ratio. Conversely, in the IRM case, optical diffraction processes do not allow ade-





# Characterization, and modeling of fuel particle thermal properties (next)

quate spatial resolution to be achieved, to ensure the probed region is contained within the buffer.

The approach selected consists, first of all, in using PM to evaluate the thermal diffusivity of the buffer's solid skeleton (see Figure 1), then estimating the layer's effective thermal conductivity tensor,<sup>[2]</sup> through a homogenization computation, taking into account conduction heat transfer through the pores (see Figure 2).

To achieve experimental validation of this approach, IRM measurements on much thicker buffers (~ 500  $\mu$ m; see Figure 3a) were compared with effective diffusivity values, as computed by a "numerical flash" (NF) method. Its principle consists in illuminating one of the sides of the heterogeneous sample by a uniformly distributed pulsed excitation flux. Heating over the opposite side of the sample (known as a thermogram) is computed over time. Effective thermal diffusivity is determined by minimizing, through a least-squares technique,<sup>[3]</sup> the standard deviation between the thermogram yielded by the "numerical experiment," and the theoretical thermogram. The latter is obtained by resolving the 1D heat equation, along the direction perpendicular to the two opposite sides, and substituting, for the heterogeneous material, an equivalent homogeneous, isotropic material.

Effective circumferential thermal diffusivity, as determined in this manner (see Figure 3b) is equal to  $4.45 \cdot 10^{-6}$  m<sup>2</sup>/s. The same numerical experiment, this time carried out along the direction perpendicular to the flash (i.e. along the radial direction), yields a diffusivity value of  $4.1 \cdot 10^{-6} \text{ m}^2/\text{s}$ . The anisotropy factor thus stands at 1.08. IRM measurements, carried out across the section (see Figure 3a), allowed circumferential thermal diffusivity to be estimated at  $4.41 \cdot 10^{-6}$  m<sup>2</sup>/s, along the direction shown in red (see Figure 4a), with an anisotropy factor standing at 1.06 (see Figure 4b), thus allowing radial diffusivity to be derived, at  $4.16 \cdot 10^{-6} \text{ m}^2/\text{s}$ .

The values ascertained thus lie quite close to those computed by NF. These findings thus corroborate the approach selected, to characterize the nominal buffer.

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### Figure 2.

Homogenization computation, using a "numerical hot stage" technique:

a) Micrograph of the computation domain, and boundary conditions; local thermal properties are associated either to the white pixels (buffer solid skeleton), or to the black pixels (air held in the pores); b) Computed temperature field, allowing effective thermal conductivity to be evaluated, along the direction perpendicular to the temperature gradient (radial direction), as being equal, in this case, to 0.4 W/m/K.



#### Figure 3.

At a: equatorial section of a thick buffer; shown above the red outline: principle of NF method: deposition of pulsed energy  $\delta$  Q (t) is effected on one side of the sample, and heating on the opposite side is computed over time. Local thermal properties are associated either to the white pixels (buffer solid skeleton), or to the black pixels (air held in the pores); at b: thermogram, yielded by an NF experiment carried out on the region outlined in red in photograph a, and its theoretical fit. A region of low thickness was selected, to avoid, as far as possible. effects related to the curvature of the microstructure, apparent in a.



Figure 4. a) 300 × 400 µm<sup>2</sup> mapping of thermal signal phase, measured at 160 Hz on the buffer equatorial section shown in 3a (the scale is graduated in degrees of angular variation). b) Fit of isophases selected for the determination.

(2) Thermal conductivity tensor: in the case of an isotropic material, thermal conductivity is a scalar quantity. For a thermally anisotropic material, if that property is to be described, it must be related to the material's structure: the tensor concept is thus used, this allowing conductivity values to be given along the principal axes of the material.

(3) Least-squares technique: an adjustment method, allowing the impact to be minimized, of measurement errors on experimental data, as compared to a mathematical model.

# FOCUS A

# The components of a nuclear system

nuclear system comprises a Anuclear reactor and the fuel cycle associated to it. It is the object of overall optimization, when industrially deployed - from raw materials to waste. In such a system, for which it forms the lynchpin, the reactor is given the ability to **recycle** fuel - so as to recover for value-added purposes fissile materials (uranium, plutonium), or even fertile materials (uranium, thorium) - and to minimize, through transmutation, production of long-lived waste, by burning, to a large extent, its own waste - namely, the minor actinides (MAs). Some systems may also feature online reprocessing plants.

The reactor itself, whichever **technology** line it may come under (see Focus B,



Virtual 3D imagery of the components and circuits in a reactor of the PWR type.

*Reactor lines, generations, and neutron spectra*, p. 14), invariably comprises the same main components (as regards fission technology at any rate, since fusion reactors make use of altogether different nuclear processes).

The core, i.e. the area where chain reactions are sustained, holds the fuel, bearing fissile, energy-yielding materials (heavy nuclei), as well as fertile materials which, subjected to the action of neutrons, turn in part into fissile materials. The fuel may come in a number of forms (pellets, pebbles, particles), and fuel elements may be brought together in rods, pins, or plates, these in turn being grouped together in assemblies, as is the case, in particular, in water-cooled reactors.

The moderator, when required, plays an

essential part. This is a material consisting in light **nuclei**, which slow down neutrons by way of *elastic* scattering. It must exhibit low neutron**capture** capability, if neutron "wastage" is to be avoided, and sufficient density to ensure effective slowing down. Thermal-spectrum reactors (see Focus B) require a moderator – as opposed to fast-spectrum reactors (which, on the other hand, must compensate for the low probability of **fast-neutron**-induced fission through a steep rise in neutron numbers) - to slow down the neutrons, subsequent to the fission that yielded them, to bring them down to the optimum velocity, thus ensuring in turn further fissions. One example of a moderator is graphite, which was used as early as the first atomic "pile," in 1942, associated to a gas as coolant fluid.

The coolant fluid removes from the core the thermal energy released by fission processes, and transports the calories to systems that will turn this energy into useable form, electricity as a rule. The coolant is either water.<sup>[1]</sup> in "water reactors" (where it also acts as moderator), or a liquid metal (sodium, or lead), or a gas (historically, carbon dioxide, and later helium, in gas-cooled reactors [GCRs]), or yet molten salts. In the last-mentioned case, fuel and coolant are one and the same fluid, affording the ability to reprocess nuclear materials on a continuous basis, since the actinides are dissolved in it.

The choice of technology line has major repercussions on the choice of materials (see Focus E, *The main families of nuclear materials*, p. 76). Thus, the core of fast-neutron reactors may not contain neutron-moderating substances (water, graphite), and their coolant must be transparent to such neutrons.

Control devices (on the one hand, control rods, or pilot and shutdown rods, made of neutron-absorbent materials [boron, cadmium...], and, on the other hand, neutron "poisons") allow the neutron population to be regulated and, in the process, by acting on its **reactivity**, to hold reactor power at the desired level, or even to quench the chain reaction. The rods, held integral and moving as one unit (known as a **cluster**) are inserted more or less deeply into the core. Poisons, on the other hand, may be adjusted in concentration within the cooling circuit.

A closed, leakproof, primary circuit contains the core, and channels and propels (by means of circulators – pumps or compressors) the coolant, which transfers its heat to a secondary circuit, by way of a heat exchanger, which may be a steam generator (this being the case equally in a pressurizedwater reactor, or in the secondary circuit of a fast reactor such as Phénix). The reactor vessel, i.e. the vessel holding the core immersed in its cooling fluid, forms, in those cases when one is used, the main component of this primary circuit.

The secondary circuit extends out of the "nuclear island," to actuate, by way of a turbine, a turbo-alternator, or to feed a heat-distribution network. In heavy-water reactors,<sup>(1)</sup> and in some gas-cooled reactors, heat is transferred from gas to water in conventional heat exchangers.

A **tertiary circuit** takes off the unused heat, by way of a **condenser**, to a cold source (water in a river, or the sea), or the air in a cooling tower, or yet some other thermal device (e.g. for hydrogen production).

Other components are only found in certain reactor lines, such as the pressurizer in pressurized-water reactors (PWRs), where pressurization keeps the water in the liquid state by preventing it from boiling. On the other hand, boiling is put to work in boilingwater reactors (BWRs), the other line of light-water reactors (LWRs), where the primary circuit water comes to the boil, and directly actuates the turbine.

(1) *Heavy water*, in which **deuterium** is substituted for the **hydrogen** in ordinary water, was the first kind of moderator, used for reactor concepts requiring very low neutron absorption. *Light water* became the norm for operational, second-generation reactors. For the future, *supercritical water*, for which thermodynamic and transport properties are altered as it goes through the critical point (temperature of 374 °C, for a pressure higher than 22 MPa [221 bars, i.e. some 200 times atmospheric pressure]), may be used, to enhance the reactor's **Carnot efficiency** (see Focus C, *Thermodynamic cycles and energy conversion*, p. 23).

# FOCUS B

# Reactor lines, generations, and neutron spectra

Nuclear reactor lines correspond to the many combinations of three basic components: coolant, moderator (when required), and fuel – almost invariably uranium, possibly mixed with plutonium (see Focus A, *The components of a nuclear system*, p. 10).

Numerous setups have been experimented with since the onset of the industrial nuclear energy age, in the 1950s, though only a few of these were selected, for the various generations of operational power generating reactors.

The term technology line, or reactor line, is thus used to refer to one possible path for the actual construction of nuclear reactors having the ability to function under satisfactory safety and profitability conditions, and defined, essentially, by the nature of the fuel, the energy carried by the neutrons involved in the chain reaction, the nature of the moderator, and that of the coolant.

The term is used advisedly, implying as it does that this combination stands as the origin of a succession of reactors, exhibiting characteristics of a technological continuum. More or less directly related to this or that line are research and trials reactors, which are seldom built as a series. Such reactor lines are classified into two



The four PWR units of EDF's Avoine power station, near Chinon (central France), belong to the second generation of nuclear reactors.

main families, depending on the **neutron** spectrum chosen: thermal, or fast (an operating range partly straddling both domains is feasible, for research reactors), according to whether neutrons directly released by fission are allowed to retain their velocity of some 20,000 km/s, or whether they are slowed down to bring them into thermal equilibrium (thermalizing them) with the material through which they scatter. The neutron spectrum, i.e. the energy distribution for the neutron population present within the **core**, is thus a thermal spectrum in virtually all reactors in service around the world, in particular, in France, for the 58 PWRs (pressurizedwater reactors) in the EDF fleet. In these reactors, operating with enriched uranium (and, in some cases, **plutonium**), heat is transferred from the core to **heat exchangers** by means of water, kept at high pressure in the **primary circuit**.

Together with **BWRs** (boiling-water reactors), in which water is brought to the boil directly within the core, PWRs form the major family of light-water reactors (LWRs), in which ordinary water plays the role both of coolant, and moderator.

Use of the fast spectrum is, currently, restricted to a small number of reactors, operated essentially for experimental purposes, such as Phénix, in France, Monju and Joyo, in Japan, or BOR-60, in Russia. In such fast reactors (FRs), operating as they do without a moderator, the greater part of fission processes are caused by neutrons exhibiting energies of the same order as that they were endowed with, when



yielded by fission. A few reactors of this type have been built for industrial production purposes (Superphénix in France, BN600 in Russia), or investigated with such a purpose in mind (mainly **EFR**, a European endeavor, in the 1980s and 1990s, BN800 in Russia, CEFR in China, PFBR in India).

Electrical power generation reactors fall into four generations. The *first generation* covers reactors developed from the 1950s to the 1970s, which made possible the takeoff of nuclear electricity production in the various developed countries, comprising in particular the UNGG (or NUGG: natural uraniumgraphite-gas) line, using graphite as moderator, and carbon dioxide as coolant, in France; the Magnox line, in the United Kingdom; and, in the United States, the first land-based<sup>(1)</sup> pressurized-water reactor (PWR), built at Shippingport.

While comparable in some respects to firstgeneration reactors, the Soviet Union's **RBMK** line (the technology used for the reactors at Chernobyl) is classed under the second generation, owing, in particular, to the time when it came on stream. RBMK reactors, using graphite as moderator, and cooled with ordinary water, brought to boil in pressure tubes, or channels, were finally disqualified by the accident at Chernobyl, in 1986.

The *second generation* covers those reactors, currently in service, that came on stream in the period from the 1970s to the 1990s. Solely

(1) In the United States, as in France, the first pressurized-water reactors were designed for naval (submarine) propulsion. built for electricity generation purposes, most of these (87% of the world fleet) are watercooled reactors, with the one outstanding exception of the British-built AGRs (advanced gas-cooled reactors). The standard fuel they use consists of sintered enriched uraniumoxide pellets, to about 4% uranium-235 enrichment, stacked in impervious tubes (rods), which, held together in bundles, form assemblies. PWRs hold the lion's share of the market, accounting for 3 nuclear reactors out of 5 worldwide. This line includes the successive "levels" of PWR reactor models built, in France, by Framatome (now trading as Areva NP) for national power utility EDF. Russian reactors from the VVER 1000 line are comparable to the PWRs in the West. While operated in smaller numbers than PWRs, BWRs (boiling-water reactors) are to be found, in particular, in the United States, Japan, or Germany. Finally, natural-uranium powered reactors of the CANDU type, a Canadian design, and their Indian counterparts, form a line that is actively pursued. These are also pressurized-water reactors, however they use heavy water  $(D_2 O)$ for their moderator, and coolant, hence the term **PHWR** (pressurized-heavy-water reactor) used to refer to this line.

The *third generation* corresponds to installations that are beginning to enter construction, scheduled to go on stream from around 2010. This covers, in particular, the French–German EPR, designed by Areva NP (initially: Framatome and Siemens), which company is also putting forward a boiling-water reactor, the SWR-1000, at the same

time as it has been coming together with Japanese firm Mitsubishi Heavy Industries. This generation further includes the AP1000 and AP600 types from Westinghouse, a firm now controlled by Toshiba; the ESBWR and ABWR II from General Electric, now in association with Hitachi; the Canadian ACRs, and the AES92 from Russia; along with projects for smaller integral reactors.

Programs for modular high-temperature reactors, of the GT–MHR (an international program) or PBMR (from South African firm Eskom) type, belong to the third generation, however they may be seen as heralding fourth-generation reactors.

The fourth generation, currently being investigated, and scheduled for industrial deployment around 2040, could in theory involve any one of the six concepts selected by the Generation IV International Forum (see Box, in The challenges of sustainable energy production, p. 6). Aside from their use for electricity generation, reactors of this generation may have a cogeneration capability, i.e. for combined heat and power production, or even, for some of models, be designed solely for heat supply purposes, to provide either "low-temperature" (around 200 °C) heat, supplying urban heating networks, or "intermediate-temperature" (500–800 °C) heat, for industrial applications, of which seawater desalination is but one possibility, or yet "high- (or even veryhigh-) temperature" (1,000-1,200 °C) heat, for specific applications, such as hydrogen production. **biomass** gasification. or hydrocarbon cracking.

# FOCUS C

# Thermodynamic cycles and energy conversion

In the large-scale conversion of heat into electricity, a thermodynamic cycle must be involved. Conversion efficiency  $\eta$  is always lower than the Carnot efficiency:

$$\eta = 1 - \frac{T_c}{T_t}$$

where  $T_h$  is the temperature of the hot source, and  $T_c$  is the temperature of the cold source.

Generally speaking, a distinction is made, for energy conversion, between the **direct cycle**, whereby the fluid originating in the hot source directly actuates the device using it (a turbo-alternator, for instance), and, conversely, the **indirect cycle**, whereby the cooling circuit is distinct from the circuit ensuring the energy conversion itself. The **combined indirect cycle** may complement this setup by adding to it a gas turbine, or, by way of a steam generator, a steam turbine.

Any system built around a nuclear generator is a heat engine, making use of the principles of thermodynamics. Just as fossil-fuel- (coal-, fuel oil-) burning thermal power plants, nuclear power plants use the heat from a "boiler," in this case delivered by **fuel elements**, inside which the **fission** processes occur. This heat is converted into electric energy, by making a fluid (water, in most reactors currently in service) go through an *indirect* thermodynamic cycle, the so-called Rankine (or Hirn-Rankine) cycle, consisting of: water vaporization at constant pressure, around the hot source; expansion of the steam inside a turbine; condensation of the steam exiting the turbine at low pressure; and compression of the condensed water to bring that water back to the initial pressure. In this arrangement, the circuit used for the water circulating inside the core (the primary circuit; see Focus A, The components of a nuclear system, p. 10) is distinct from the circuit ensuring the actual energy conversion. With a maximum steam temperature of some 280 °C, and a pressure of 7 MPa, the net energy efficiency (the ratio of the electric energy generated, over the thermal energy released by the reactor core) stands at about one third for a second-generation pressurized-water reactor. This can be made to rise to 36-38% for a third-generation PWR, such as EPR, by raising the temperature, since the Carnot equation clearly shows the advantage of generating high-temperature heat, to achieve high efficiency. Indeed, raising the core outlet temperature by about 100 degrees allows an efficiency improvement of several points to be achieved.



Figure. Brayton cycle, as implemented in an open-cycle gas turbine.

The thermodynamic properties of a coolant gas such as helium make it possible to go further, by allowing a target core outlet temperature of at least 850 °C. To take full advantage of this, it is preferable, in theory, to use a direct energy conversion cycle, the Joule–Brayton cycle, whereby the fluid exiting the reactor (or any other "boiler") is channeled directly to the turbine driving the alternator, as is the case in naturalgas, combined-cycle electricity generation plants, or indeed in a jet aero-engine. Using this cycle, electricity generation efficiency may be raised from 51.6% to 56%, by increasing Tc from 850 °C to 1,000 °C.

Indeed, over the past half-century, use of natural gas as a fuel has resulted in a spectacular development of gas turbines (GTs) that can operate at very high temperatures, higher than around 1,000 °C. This type of energy conversion arrangement stands, for the nuclear reactors of the future, as an attractive alternative to steam turbines. GT thermodynamic cycles are in very widespread use, whether for propulsion systems, or large fossil-fuel electricity generation plants. Such cycles, known as Brayton cycles (see Figure) simply consist of: drawing in air, and compressing it to inject it into the combustion chamber  $(1 \rightarrow 2)$ ; burning the air-fuel mix inside the combustion chamber  $(2 \rightarrow 3)$ ; and allowing the hot gases to expand inside a turbine  $(3 \rightarrow 4)$ . On exiting the turbine, the exhaust gases are discharged into the atmosphere (this forming the cold source): the cycle is thus termed an open cycle. If the hot source is a nuclear reactor, open-cycle operation, using air, becomes highly problematical (if only because of the requisite compliance with the principle of three confinement barriers between nuclear fuel and the ambient environment). In order to *close* the cycle, all that is required is to insert a heat exchanger at the turbine outlet, to cool the gas (by way of a heat exchanger connected to the cold source), before it is reinjected into the compressor. The nature of the gas then ceases to be dictated by a combustion process.

# FOCUS D

# What is multiphysics, multiscale modeling?

Multiphysics, multiscale modeling approach, arising out of the requirement to take into account, when modeling a system for which behavior is to be predicted, all processes – these in practice being coupled one with another – acting on (or prevailing in) that system. This is the most complete form of modeling, for a concatenation of various processes, of highly diverse scales, bringing together as it does all of the relevant knowledge, whether theoretical or empirical, at a variety of scales, into elementary building blocks, which then have to be assembled.

In physical terms, this takes into account the couplings arising between basic processes of diverse nature. In the area of reactor physics, for instance, coupling occurs between structural mechanics, neutronics, and thermal-hydraulics.

This kind of modeling further aims to provide a description of processes at different scales. In the area of materials physics, the aim will be, e.g., to derive the macroscopic properties of a polycrystalline material, from its description at the most microscopic scale (the atom), by way of nested levels of description (molecular dynamics, dislocation dynamics).

The issue is that of connecting these various levels of description, by using the correct information to pass from one scale to the next with no break in continuity, and of handling in modular fashion such behavior laws, valid as these are at diverse scales (see Figure).

Thus it is numerical computation of a composite character, depending on the spatial scale being considered, that "drives" the overall model. All the more composite, since researchers are led to "chain" *deterministic*, and *probabilistic* models, whether it be for lack of an exhaustive knowledge of the basic processes involved, or because the numerical resolution of the deterministic equations would prove too difficult, or too heavy a task. Hence the adoption of such methods as the Monte-Carlo method, in particular.

Finally, multiscale modeling joins up, through superposition techniques, numerical models at different scales. This makes it possible – to stay with the example of materials – to "zoom in" on



#### Figure.

Improving nuclear fuel reliability, and cost-effectiveness calls for finescale modeling of that fuel, through a multiscale approach, from reactor to fuel microstructure (in this instance, MOX fuel). Microstructural characteristics (porosity, cluster size and distribution, grain size...) have a direct impact on fuel rod behavior under irradiation, and thus on reactor ease of operation, and on that rod's lifespan. regions that are particularly sensitive to **stresses**, such as fissures, welds, or supporting structures.

Multiphysics, multiscale modeling thus raises, in acute fashion, the issue of the compatibility, and consistency of the computation **codes** making up the elementary building blocks in the description. However, the outcomes are on a par with the difficulty: in the area of metallic materials, in particular, it is now possible to implement an approach predicting macroscopic properties from "first principles," of atomic physics and molecular dynamics (ab-initio method, see note (1) p. 79), by way of the physical description of microstructures. In the nuclear energy context, the investigation of materials subjected to irradiation provides a good illustration of this approach, since it has now become feasible to bridge the gap between knowledge of defects at the macroscopic scale, and modeling of point defect formation processes, at the atomic scale.

While physics naturally provides the first level, in this type of modeling, the two other levels are mathematical, and numerical, insofar as the point is to connect findings from measurements, or computations, valid at different scales, going on to implement the **algorithms** developed. Multiphysics, multiscale modeling has thus only been made possible by the coming together of two concurrent lines of advances: advances in the knowledge of basic processes, and in the power of computing resources.

CEA is one of the few organizations around the world with the capability to develop such multiphysics, multiscale modeling, in its various areas of research and development activity, by bringing together a vast ensemble of modeling, experimental, and computation tools, enabling it to demonstrate, at the same time, the validity of theories, the relevance of technologies, and bring about advances in component design, whether in the area of nuclear energy (in which context coupling is effected between partial codes from CEA and EDF), or, for example, in that of the new energy technologies.

# FOCUS E

# The main families of nuclear materials

The specific conditions attributable to radiation conditions prevailing inside nuclear reactors mean it is imperative to look to materials exhibiting special characteristics, which may be grouped under two main categories: cladding and structural materials, on the one hand, and fuel materials, on the other. For either group, the six concepts for fourth-generation systems selected by the Generation IV International Forum mostly require going for innovative solutions, as the favored option (see Table, p. 71).

The characteristics, in terms of resistance to temperature, pressure, fatigue, heat, corrosion, often under stress, that should be exhibited, as a general rule, by materials involved in any industrial process must, in the nuclear energy context, be virtually fully sustained, notwithstanding the effects of irradiation, due in particular to the **neutron** flux. Indeed, irradiation speeds up, or amplifies processes such as **creep** (irradiation creep), or causes other ones, such as **swelling**, or growth, i.e. an **anisotropic** deformation occurring under the action of a neutron flux, in the absence of any other stress.

Structural materials in the reactor itself are subject, in particular, to the process of **activation** by neutron bombardment, or bombardment by other particles (**photons**, **electrons**).

Materials employed for fuel structures (assemblies, claddings, plates, and so on) are further subjected to yet other stresses. Finally, the fuel itself is a material, taking the form, in current light-water reactors, for instance, of sintered uranium and/or plutonium ceramics, in the form of pellets.

Neutron **irradiation** can cause a major alteration in the properties exhibited by the materials employed in the various components of a reactor. In metals, and metal alloys, but equally in other solid materials, such as ceramics,<sup>(1)</sup> such alterations are related to the evolution of the **point defects** generated by this irradiation, and to the

or incorporated into composites, which may be of the cercer (a ceramic held in a matrix that is also a ceramic) or cermet (a ceramic material embedded in a metallic matrix) types. With regard to nuclear fuel, this takes the form of a closely mixed composite of metallic products, and refractory compounds, the fissile elements being held in one phase only, or in both. extraneous **atoms** generated by nuclear reactions, substituting for one of the atoms in the **crystal** lattice. The nature, and number of such defects depends both on the neutron flux, and neutron energies, however the neutrons that cause appreciable structural evolutions are, in **thermal-neutron reactors** as in **fast-neutron reactors** (**fast reactors**), the **fast neutrons**.

A crystal invariably exhibits some defects, and irradiation may generate further defects. Point defects fall under two types: vacancies (one atom being expelled from its location in the crystal), and interstitials (one extra atom positioning itself at a supernumerary site, between the planes of the crystal lattice).

Dislocations, marking out a region where the crystal stack is disturbed by local slipping, affecting a single atomic plane, in turn act as sources, or sinks of point defects. Vacancies may come together to form vacancy clusters, loops, or cavities, while interstitials may form interstitial clusters, or dislocation loops. At the same time, copper, manganese, and nickel atoms, e.g. in a vessel steel alloy, tend to draw together, to form clusters, resulting in hardening of the steel. Finally, grain boundary are defects bounding two crystals exhibiting different orientations, and thus act as potential factors of embrittlement. Many of the metal's properties are subject to alteration at these boundaries.

The damage occasioned to such materials is expressed in terms of displacements per atom (dpa), with n dpa implying that every atom in the material has been displaced n times, on average, during irradiation.

## **Crystal structures**

Metallic materials exhibit a crystal structure: they are formed by an elementary unit, periodically repeating across space, known as a unit cell, consisting of atoms, in precise, definite numbers and positions. Repetition of such structures endows them with specific properties. Three of these structures, defining the position of the atoms, are of importance:

the body-centered cubic structure (that found in iron at ambient room temperature, chromium, vanadium); such materials as a rule exhibit a ductile-brittle behavior transition, depending on temperature;
the face-centered cubic structure (nickel, aluminum, copper, iron at high temperature); • the **hexagonal structure** (that of zirconium, or titanium).

Depending on temperature and composition, the metal will structure itself into elementary crystals, the grains, exhibiting a variety of microstructures, or phases. The way these arrange themselves has a major influence of the properties exhibited by metals, steels in particular. The ferrite of pure iron, with a body-centered cubic structure, turns into austenite, a face-centered cubic structure, above 910 °C. Martensite is a particular structure, obtained through *tempering*, which hardens it, followed by annealing, making it less brittle. Bainite is a structure intermediate between ferrite and martensite. likewise obtained through tempering followed by annealing.

Among metals, high-chromium-content (more than 13%) stainless steels, exhibiting as they do a corrosion and oxidation resistance that is due to the formation of a film of chromium oxide on their surface, take the lion's share. If the criterion for stainless ability (rustproofness) is taken to be chromium content, which should be higher than 13%, such steels fall into three main categories: ferritic steels, austenitic steels, and austenitic-ferritic steels.

## Steel families

Ferritic steels, exhibiting a *body-centered* cubic structure (e.g. F17), are characterized by a low carbon concentration (0.08–0.20%), and high chromium content. As a rule containing no nickel, these are iron–chromium, or iron–chromium–molybdenum alloys, with a chromium content ranging from 10.5% to 28%: they exhibit no appreciable hardening when tempered, only hardening as a result of work hardening.

They exhibit a small expansion coefficient, are highly oxidation resistant, and prove suitable for high temperatures. In the nuclear industry, 16MND5 bainitic steel, a low-carbon, low-alloy (1.5% manganese, 1% nickel, 0.5% molybdenum) steel, takes pride of place, providing as it does the vessel material for French-built **PWRs**, having been selected for the qualities it exhibits at 290 °C, when subjected to a fluence of  $3 \cdot 10^{19}$  n  $\cdot$  cm<sup>-2</sup>, for neutrons of energies higher than 1 MeV.

Martensitic steels, exhibiting a *body-centered cubic* structure, are ferritic steels containing less than 13% chromium (9–12% as a rule), and a maximum 0.15% carbon,

<sup>(1)</sup> Ceramics are used on their own,



Pressure-vessel nozzle shell for EDF's Flamanville 3 reactor, the first EPR to be built on French soil.

which have been subjected to annealing: they become martensitic when quenched, in air or a liquid, after being heated to reach the austenitic domain. They subsequently undergo softening, by means of a heat treatment. They may contain nickel, molybdenum, along with further addition elements. These steels are magnetic, and exhibit high stiffness and strength, however they may prove brittle under impact, particularly at low temperatures. They have gained widespread use in the nuclear industry (fastenings, valves and fittings...), owing to their good corrosion resistance, combined with impressive mechanical characteristics.

Austenitic steels, characterized by a facecentered cubic structure, contain some 17-18% chromium, 8-12% nickel (this enhancing corrosion resistance: the greater part, by far, of stainless steels are austenitic steels), little carbon, possibly some molybdenum, titanium, or niobium, and, mainly, iron (the remainder). They exhibit remarkable ductility, and toughness, a high expansion coefficient, and a lower heat conductivity coefficient than found in ferritic-martensitic steels. Of the main grades (coming under US references AISI<sup>[2]</sup> 301 to 303, 304, 308, 316, 316L, 316LN, 316Ti, 316Cb, 318, 321, 330, 347), 304 and 316 steels proved particularly important for the nuclear industry, before being abandoned owing to their excessive swelling under irradiation. Some derivatives (e.g. 304L, used for internal structures and fuel assembly end-caps, in PWRs; or 316Ti<sub>ɛ</sub>, employed for claddings) stand as reference materials. In fast reactors, they are employed, in particular, for the fabrication of hexagonal tubes (characteristic of reactors of the Phénix type) (316L[N] steel), while 15/15Ti austenitic steel has been optimized for fuel pins for this reactor line, providing the new cladding reference for fast reactors.

# FOCUS (Cond't) E

Austenitic-ferritic steels, containing 0%, 8%, 20%, 32%, or even 50% ferrite, exhibit good corrosion resistance, and satisfactory weldability, resulting in their employment. in molded form. for the ducts connecting vessels and steam generators.

One class of alloys that is of particular importance for the nuclear industry is that of nickel alloys, these exhibiting an austenitic structure. Alloy 600 (Inconel 600, made by INCO), a nickel (72%), chromium (16%), and iron (8%) alloy, further containing cobalt and carbon, which was employed for PWR steam generators (along with alloy 620) and vessel head penetrations, was substituted, owing to its poor corrosion resistance under stress, by alloy 690, with a higher chromium content (30%). For certain components, Inconel 706, Inconel 718 (for PWR fuel assembly grids), and Inconel X750 with titanium and aluminum additions have been selected. in view of their swelling resistance, and very high mechanical strength. For steam generators in fast reactors such as Phénix, alloy 800 (35% nickel, 20% chromium, slightly less than 50% iron) was favored. Alloy 617 (Ni-Cr-Co-Mo), and alloy 230 (Ni-Cr-W), widely employed as they are in the chemical industry, are being evaluated for gas-cooled VHTRs.

Ferritic-martensitic steels (F-M steels) exhibit a body-centered cubic structure. In effect, this category subsumes the martensitic steel and ferritic steel families. These steels combine a low thermal expansion coefficient with high heat conductivity. Martensitic or ferritic steels with chromium contents in the 9-18% range see restricted employment, owing to their lower creep resistance than that of austenitic steels. Fe-9/12Cr martensitic steels (i.e. steels containing 9-12% chromium by mass) may however withstand high temperatures, and are being optimized with respect to creep. For instance, Fe-9Cr 1Mo molybdenum steel might prove suitable for the hexagonal tube in SFR fuel assemblies. Under the general designation of AFMSs (advanced ferritic-martensitic steels), they are being more particularly investigated for use in gas-cooled fast reactors.

Oxide-dispersion-strengthened (ODS) ferritic and martensitic steels were developed to combine the swelling resistance exhibited by ferritic steels, with a creep resistance in hot conditions at least equal to that of austenitic steels. They currently provide the reference solution for fuel cladding, for future sodium-cooled reactors. The **cladding material** in light-water reactors, for which stainless steel had been used initially, nowadays consists of a zirconium alloy, selected for its "transparency" to neutrons, which exhibits a compact hexagonal crystal structure at low temperature, a face-centered cubic structure at high temperature. The most widely used zirconium-iron-chromium alloys are tin-containing **Zircaloys** (Zircaloy-4 in PWRs, Zircaloy-2 in BWRs, ZrNb - containing niobium - in the Russian VVER line), owing to their outstanding behavior under radiation, and capacity with respect to creep in hot conditions.

After bringing down tin content, in order to improve corrosion resistance, a zirconium-niobium alloy (M5<sup>®</sup>) is presently being deployed for such cladding.

Among nuclear energy materials, graphite calls for particular mention: along with heavy water, it is associated with reactors that must operate on natural uranium; it proves advantageous as a moderator, as being a low neutron absorber.

For **GFRs**, novel ceramics, and new alloys must be developed, to the margins of high fluences. Researchers are storing high hopes on refractory materials containing no metals.

In particle fuels, uranium and plutonium oxides are coated with several layers of insulating pyrocarbons, and/or silicon carbide (SiC), possibly in fibrous form (SiCf). These are known as coated particles (CPs). While SiC-coated UO<sub>2</sub>, or **MOX** balls stand as the reference, ZrC coatings might afford an alternative.

At the same time, conventional sintered uranium oxide (and plutonium oxide, in MOX) pellets might be supplanted by advanced fuels, whether featuring chromium additions or otherwise, with the aim of seeking to overcome the issues raised by pel**let-cladding interaction**, linked as this is to the ceramic fuel pellet's tendency to swell under irradiation.

Oxides might be supplanted by **nitrides** (compatible with the **Purex** reprocessing process), or carbides, in the form e.g. of uranium-plutonium alloys containing 10% zirconium.

(2) This being the acronym for the American Iron and Steel Institute.

# The six concepts selected by the Gen IV Forum

Of the six concepts selected by the **Generation IV International Forum** for their ability to meet the criteria outlined, three – and ultimately four – make use of **fast neutrons**, while three (ultimately two) use **thermal neutrons**. At the same time, two of the six concepts use gas as a coolant (they are thus gas-cooled reactors [GCRs]). The six concepts are the following:

# GFR

The gas-cooled fast reactor system (GFR) is a high-temperature, gas-cooled (helium-cooled as a rule), fast-neutron reactor allowing actinide recycle (homogeneous, or heterogeneous), while sustaining a **breeding** capability greater than unity. The reference concept is a helium-cooled, direct- or indirect-cycle reactor, exhibiting high efficiency (48%). Decay heat removal, in the event of depressurization, is feasible through natural convection a few hours after the accident. Maintaining forced circulation is a requisite, during the initial accident stage. Core power density is set at a level such as to restrict fuel temperature to 1,600 °C during transients. The innovative fuel is designed to retain fission products (at temperatures below the 1,600 °C limit), and preclude their release in accident conditions. Reprocessing of spent fuel for recycling purposes may be considered (possibly on the reactor site), whether by means of a pyrochemical or a hydrometallurgical process. The GFR is a high-performance system, in terms of natural resource utilization, and long-lived waste minimization. It comes under the gas-cooled technology line, complementing such thermal-spectrum concepts as the GT-MHR,<sup>(1)</sup> PBMR,<sup>(2)</sup> and VHTR.

# Le SFR

The sodium-cooled fast reactor system (SFR) is a liquid-sodiumcooled, fast-neutron reactor, associated to a closed cycle, allowing full actinide recycle, and plutonium breeding. Owing to its breeding of fissile material, this type of reactor may operate for highly extended periods without requiring any intervention on the core. Two main options may be considered: one that, associated to the **reprocessing** of metallic fuel, results in a reactor of intermediate unit power, in the 150–500 MWe range; the other, characterized by the Purex reprocessing of mixedoxide fuel (MOX), corresponds to a high-unit-power reactor, in the 500-1,500 MWe range. The SFR presents highly advantageous natural resource utilization and actinide management features. It has been assessed as exhibiting good safety characteristics. A number of SFR prototypes are to be found around the world, including Joyo and Monju in Japan, BN600 in Russia, and Phénix in France. The main issues for research concern the full recycling of actinides (actinide-bearing fuels are radioactive, and thus pose fabrication difficulties), in-service inspection (sodium not being transparent), safety (passive safety approaches are under investigation), and capital cost reduction. Substitution of water with supercritical CO<sub>2</sub> as the working fluid for the power conversion system is also being investigated

# LFR

The lead-cooled fast reactor system (LFR) is a lead- (or lead-bismuth alloy-) cooled, fast-neutron reactor, associated to a closed fuel cycle, allowing optimum **uranium** utilization. A number of reference systems have been selected. Unit power ranges from the 50–100 MWe bracket, for so-called battery concepts, up to 1,200 MWe, including modular concepts in the 300–400 MWe bracket. The concepts feature long-duration (10–30 years) fuel management. Fuels may be either metallic, or of the **nitride** type, and allow full actinide recycle.







GT–MHR: Gas-Turbine Modular Helium Reactor.
 PBMR: Pebble-Bed Modular Reactor.



# control rods uppercritical water turbine generator electrical power heat sink pump

#### coolant control sali rods electrical purified reactor generator nower turbine recuperator pump fuel reprocessing plant salt freeze plug heat amua sink emergency dump tanks heat exchanger intercooler compressor

# VHTR

The very-high-temperature reactor system (VHTR) is a very-high-temperature, helium-gas-cooled, thermalneutron reactor, initially intended to operate with an open fuel cycle. Its strong points are low costs, and most particularly safety. Its capability, with regard to sustainability, is on a par with that of a third-generation reactor, owing to the use of an open cycle. It may be dedicated to hydrogen production, even while also allowing production of electricity (as sole output, or through cogeneration). The specific feature of the VHTR is that it operates at very high temperature (> 1,000 °C), to provide the heat required for water splitting processes, by way of thermochemical cycles (iodine-sulfur process), or high-temperature **electrolysis**. The reference system exhibits a unit power of 600 MWth, and uses helium as coolant. The core is made up of prismatic blocks, or pebbles.

# **SCWR**

The supercritical-water-cooled reactor system (SCWR) is a supercritical-water-cooled, thermal-neutron reactor, in an initial stage (open fuel cycle); a fast-neutron reactor in its ultimate configuration (featuring a closed cycle, for full actinide recycle). Two fuel cycles correspond to these two versions. Both options involve an identical operating point, with regard to supercritical water: pressure of 25 MPa, and core outlet temperature of 550 °C, enabling a thermodynamic efficiency of 44%. Unit power for the reference system stands at 1,700 MWe. The SCWR has been assessed as affording a high economic competitiveness potential.

# **MSR**

The molten salt reactor system (MSR) is a molten salt (liquid core, with a closed cycle, through continuous online pyrochemical reprocessing), thermal-neutron - more accurately epithermal-neutron - reactor. Its originality lies is its use of a molten salt solution, serving both as fuel, and coolant. Fissile material breeding is feasible, using an optional uranium-thorium cycle. The MSR includes as a design feature online fuel recycling, thus affording the opportunity to bring together on one and the same site an electricity-generating reactor, and its reprocessing plant. The salt selected for the reference concept (unit power of 1,000 MWe) is a sodium-zirconium-actinide fluoride. Spectrum moderation inside the core is effected by placing graphite blocks, through which the fuel salt flows. The MSR features an intermediate fluoride-salt circuit, and a tertiary, water or helium circuit for electricity production.