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The long-term future for civilian nuclear power generation in France: The case for breeder reactors
Breeder reactors: The physical and physical chemistry parameters, associate material thermodynamics and mechanical engineering: Novelties and issues

La production d'électricité d'origine nucléaire en France, dans le futur à long terme : Le cas des surgénérateurs
Les réacteurs nucléaires surgénérateurs : Les paramètres physique et physico-chimiques, la thermodynamique associée des matériaux et de l'ingénierie mécanique : Nouveautés et options

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ABSTRACT

The author firstly gives a summary overview of the knowledge base acquired since the first breeder reactors became operational in the 1950s. "Neutronics", thermal phenomena, reactor core cooling, various coolants used and envisioned for this function, fuel fabrication from separated materials, main equipment (pumps, valves, taps, waste cock, safety circuits, heat exchange units, etc.) have now attained maturity, sufficient to implement sodium cooling circuits. Notwithstanding, the use of metallic sodium still raises certain severe questions in terms of safe handling (i.e. inflammability) and other important security considerations.

The structural components, both inside the reactor core and outside (i.e. heat exchange devices) are undergoing in-depth research so as to last longer. The fuel cycle, notably the refabrication of fuel elements and fertile elements, the case of transuranic elements, etc., call for studies into radiation induced phenomena, chemistry separation, separate or otherwise treatments for materials that have different radioactive, physical, thermodynamical, chemical and biological properties.

The concerns that surround the definitive disposal of certain radioactive wastes could be qualitatively improved with respect to the pressurized water reactors (PWRs) in service today. Lastly, the author notes that breeder reactors eliminate the need for an isotope separation facility, and this constitutes a significant contribution to contain nuclear proliferation.

Among the priorities for a fully operational *system* (power station – the fuel cycle – operation-maintenance – the spent fuel pool and its cooling system-emergency cooling system-emergency electric power-transportation movements-equipment handling – final disposal of radioactive matter, independent safety barriers), the author includes materials

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(fabrication of targets, an irradiation and inspection instrument), the chemistry of all sorting processes, equipment "refabrication" or rehabilitation, etc., radioprotection measures and treatment for the "transuranic" elements. For a long period of time, France was in the forefront of nuclear breeder power generation science, technological research and also in the knowledge base related to breeder reactors. It is in the country's interest to pursue these efforts and this could per se constitute one of the national priorities.[1]

R É S U M É

Les connaissances acquises depuis les premières réalisations de surgénérateurs dans le début des années cinquante sont récapitulées. La neutronique, les phénomènes thermiques, le refroidissement du cœur du réacteur, les divers fluides envisagés pour cette fonction, la fabrication des combustibles à partir de corps purs, les gros équipements (pompes, vannes, purgeur, circuits de secours, échangeurs de chaleur, etc.) ont atteint une solide maturité pour le refroidissement par sodium. Celui-ci, toutefois, pose encore des problèmes pour les manutentions et leur sureté.

Les matériaux de structures, tant à l'intérieur du cœur qu'à l'extérieur (échangeurs de chaleur) font l'objet de recherches approfondies. Le cycle des combustibles, et notamment la « re-fabrication » d'éléments combustibles et d'éléments fertiles, le sort des transuraniens, etc. demandent des études d'irradiations, de séparation chimique, de traitements séparés ou non des corps ayant des propriétés radioactives, chimiques et biologiques différentes.

Les préoccupations liées au sort ultime de certains déchets radioactifs, pourraient être qualitativement améliorés par rapport aux filières de réacteurs à eau existant aujourd'hui. Enfin, les surgénérateurs suppriment la nécessité d'avoir recours à une usine de séparation isotopique, ce qui est une contribution substantielle pour contenir une éventuelle prolifération.

Parmi les priorités du système complet: centrale-cycle de combustible-maintenance-déplacements-manutentions-sort ultime des corps radioactifs, comptons les matériaux (donc un instrument d'irradiation et d'examen), la chimie de tous les procédés de triage, de re-fabrication, de corrosion, de fabrication des équipements, etc., la sûreté-radioprotection et les transuraniens. La France a longtemps été au premier rang des recherches et des acquis dans les divers domaines concernés par les surgénérateurs. Il s'agit de continuer cet effort qui pourrait être une des priorités nationales.[1]

1. Introduction

The scientific, engineering, industrial, economic, safety and radioprotection track-record success for nuclear power stations built in those countries that had the capacity to do so, is due mainly to that of a technical policy decision, to use pressurized natural water as the moderator and the coolant. A powerful technological factor was the principal lever: engineering knowledge for water circuits (pipes, taps, pumps, boiler vessels, safety circuits and devices, etc.) alloys adapted to water environments, corrosion, welding, radioprotection and control, etc. We should not forget the intrinsic properties of water, its transparency, the radioprotection screening provided by the water tanks, etc. However, these water-cooled reactors have several limitations. To begin with operational limitations: their corrosion plays a major role leading to regular *replacement* of components. (1) Limited availability of natural ores containing uranium or thorium; (2) production of certain nuclear wastes[2] the ultimate disposal of which has not yet been totally solved by the nations concerned but is feasible on scientific and technical grounds; (3) the fuel cycles for these nuclear power generation stations require

[1] Nous sommes naturellement bien conscients de l'énorme problème qui se pose au Japon actuellement comme suite au tremblement de terre et au tsunami de mars 2011 et leurs conséquences, notamment sur des installations électronucléaires. Le texte que nous présentons concerne des conditions totalement générales, indépendantes des problèmes spécifiques de sûreté qu'il faudra, de toute façon, traiter dans le cadre d'un développement éventuel de l'énergie nucléaire.

We are aware, of course, of the huge problem that Japan has to deal with the aftermath of the quake and tsunami of March 2011 and their consequences on electronuclear power plants. The text that we present here concerns general physical topics independent of the specific safety problems, general physical topics which will have to be solved in the case of a contingent development of electronuclear power plants.

[2] *Noteworthy radioactive wastes*: The neutron capture cross-sections for water-cooled reactors (where kinetic energies of the neutrons are of the same order of magnitude as that of matter agitation in the reactors, viz., 1/40 eV at ambient temperature and approximately 1/20 eV when the reactor is producing power, are much higher (by as much as a factor 100) than in the case of fast neutrons (between 0.3 and 1 MeV). This leads in water cooled reactors to the production of numerous radioactive nuclei in the reactor itself and inside it, as well the reactor vessel contamination as various corrosion products, i.e. in the heat exchange circuits but also in the vessel itself due to circulation of the coolant fluids with, in suspension, these corrosion products materials, which are, radio activated in the core of the reactor. Such activated products are circulated throughout the complete cooling circuit and contaminate it everywhere inside the circuit and the other equipment connected for other purposes. This is a very important physical-chemical process which does not happened with the sodium coolant because the corrosion is so small. The cleanest cooling circuit of France was the SUPER PHENIX sodium circuit.

uranium enrichment plants to obtain concentrations of the ^{235}U isotope from the natural uranium element, which calls for a technology that led certain countries to produce weapon-grade uranium. In a long-term vision, breeder reactors allow us to offset these drawbacks: loss of coolant accidents, loss of pressure and safety pressure circuits and devices, etc. This article aims at recalling and stressing a few basic notions needed by all the professionals involved in the design of such future breeder reactors.

2. Formation of ^{239}Pu plutonium in breeder reactors. The plutonium isotopes and other transuranic elements. Neutron-based nuclear reactions and disintegrations. The competition between the two processes, the first one being created by the neutron flux

The first nuclear reactor that actually produced electric power was code-named EBR I ("Experimental Breeder[3] Reactor" no. 1: operated during 1951–1953), assembled under the supervision of the Argonne National Laboratory, University of Chicago. Historically, it was in this university that: (a) Enrico Fermi, in late 1942, demonstrated the scientific feasibility of a nuclear chain reaction; (b) where the element plutonium was studied by the chemist Glenn Seaborg [3], who transferred to Chicago University[4] for this purpose from the University of Berkeley, CA. The high neutron flux for the production of plutonium (in mass production) was supervised and organized by the physicist–engineer Eugene (Jëno Pal) Wigner [4]. Wigner was born and educated in Hungary and came to Germany to study and obtain chemical engineering diploma; he also produced the draft plans for the reactors that would produce mass of plutonium, and presented them to the Dupont de Nemours Company. This company then constructed the reactor facilities at Hanford, in the State of Washington. The plutonium they produced was transferred to the National Laboratory at Los Alamos, New Mexico. There, Emilio Segré [5,6] discovered that the very small proportion of ^{240}Pu had copious spontaneous fission, then emitted a lot of neutrons and make the device they were preparing useless.

Since then, a first Experiment Breeder Reactor (EBR I) and a second reactor EBR II (1963–1994)[5] was built and operated, followed by "Fermi 1" (1963–1972), then SEFOR (1969–1972) and the Fast Flux Test Reactor (1980–1993), at Hanford. Today, General Electric and Hitachi design compact modular pool – with passive cooling for decay heat removal – named PRISM, with sodium coolant, at $\approx$ 300 MWe. There is an electro metallurgical reprocessing, for the fuel elements, which are unloaded every 6 years.

Otherwise, the following reactors were built:

- A breeder reactor was built on UK Scotland's North coastline at Dounreay (1969–1977);
- A series of breeder reactors, with increasing power ratings, were built in the USSR, BN-5-10 at Obninsk – 1959–1971; and after a pause 1973 – to day, BOR 60 Dimitrovgrad – 1969 – to day; BN-350; BN-600[6] Beloyarsk 3 – 1980 – to day; BN-800, Beloyarsk 4 – critical in 2014; BN-350 – 1972–1999 (today in Kazakhstan). We have to add a design of a fast neutron reactor of 300 MWe, BREST;
- In Japan, at Monju, Joyo – 1978 – to day; Monju – 1994–1996, then 2010–;
- In the Republic of India, FBTR – 1985 – to date (40 MW thermal); PFBR[7] at Palkattam 2012–;
- In China, CEFR (65 MW thermal) since 2010[8];
- In France, a complete range of reactors (RAPSODIE – 1960 1982, PHENIX – 1973–2009, SUPERPHENIX – 1985–1998, very low power reactors for neutron flux measurement experiments – MAZURKA, etc., and a series of equipment as specific

[3] The term "breeder" was translated into French as "surgénérateur". We can observe that the fuels used for these American reactors were purely metallic and not oxides, this being the case for France's PHENIX and SUPERPHENIX. The following reactors designs studied by the Argonne National Laboratory (from 1984 to 1994), then at a new conceptual level without implementation of the final design, the Integral Fast Reactor (IFR) which grouped together the reactor facility, the chemical separation, fuel rod assembly, etc., were always *fuel metal*-based. The reason lies not only in the spectrum of kinetic energies of the "harder" neutrons, but in the facilities to carry out the treatments using advanced processes of chemical separation of the irradiated fuel elements, whether they be fissile or fertile [1,2].

[4] Glenn Seaborg, chemist from Berkeley University, then transferred at the University of Chicago, had the constant help of the University of Washington [3], at Saint Louis, Missouri, USA.

[5] EBR II was the base of a complete program, reactor and all the fuel cycle in the same place, named Integral Fast Reactor (IFR). The National Academy of Sciences (NAS) of USA write that this was the highest priority research for the nuclear power. The fuel used in this facility reached, at the end of the 1980s, the *burn up fraction* of 19% compared to the 4% in PWR. The experiment had been able to demonstrate potentialities in waste destruction. But it was arrested for budget limitation, before the end of the demonstration of the destruction of neptunium and americium. This IFR was to be proliferation resistant by always mixing plutonium and other actinides during all the fuel cycle processes and the lack of transportation out of the site of IFR.

[6] The fuel of BN-350 and BN-600 was enriched uranium oxide. BN-600 had a power of 560 MWe and a thermal power density of more than 1 MW/dm^3. The plutonium cycle, i.e., his chemistry and physical chemistry, his fabrication and refabrication, with a very radioactive field of action, the manipulations, the transport, the cooling after irradiation, etc. had not been studied. The same remark can be applied to the majority of breeders, about their fuel cycle. The only work in this scope had been, for the MOX cycle, in France.

[7] The design of FBTR has some analogies with the design of the project "European Fast Reactor" (EFR), but EFR was not built.

[8] China had purchased in 2009 two BN-800 to Russian. The construction begins this year, 2011.

for sodium handling[9] (i.e. pumps, taps, waste cocks, valves, heat exchange units, etc.) were successfully assembled and tested.[10]

The basic principle of breeder reactors calls for two nuclear reactions:
Fissile reactions[11]:

(a) the fissile fuel positioned in the reactor core[12] is plutonium. Fission (total energy of a fission ≈ 210 MeV) of the plutonium produces a majority of the generated output energy, due the kinetic energy of the fission fragments (≈ 170 Mev) in the fuel elements heat them. This heat is, evacuated via a coolant fluid. Moreover, among the ≈ 3 neutrons (this number is the mean of a probability process and written ν), that result from the fission of the fissile Pu nucleus;
(b) one (on average) of the other 3 neutrons resulting from the initial fission serve(s) to continue the chain reaction.

Fertile reactions[13]: another neutron one (or more than one, on average) initiates the breeding of plutonium (^{239}Pu) by the following nuclear reactions: ^{238}U + neutron $\rightarrow$ ^{239}U + γ (half life by "beta" disintegration: 23 minutes) $\rightarrow$ ^{239}neptunium (lifetime β disintegration: 2.23 days) + electron + antineutrino, $\rightarrow$ ^{239}Pu (half life alpha: 24 100 years) + electron + antineutrino. Depending on constraints of another nature (material resistance to intense irradiation), steep temperature gradients and consequently high mechanical strain, the pressures exerted by the gaseous fission products,[14] and the helium nuclei (α particles) which contribute to command the length of time of irradiation of the fuel elements, there is also an important and deep transformation of the fuel elements: the gradual variation of the reactors reactivity[14] as the ^{239}Pu nuclei are 'burned', the gradual loss of fertile nuclei (i.e., ^{238}U), but also creation of ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu and their disintegration into nuclei, fissile or not, some different by-products, the accumulation of the fission products, which almost all are capturing neutron, etc. All these phenomena are changing continuously the economy of neutrons. Among the consequences, one is at the heart of every nuclear reactor conception. It is *the stability of the neutron flux in all the core of the reactor* (and its possible blanket or reflector), and then the *reactor power levels with respect to any degree of internal or external disturbance, which finally modify the reactivity of the breeder reactor.*[15] It is necessary that this feedback to any disturbance (as in all the operating power reactors) leads to a *negative feedback*. One of the conditions for this stability with respect to

[9] Coolant fluids such as lead and the eutectic mix lead–bismuth are currently under study in Russia. This approach would enable us to attain a fluid reactor output temperature of approx. 800 °C. But *different concepts of breeder coolants have different spectra of kinetic energy; – i.e., at high neutron kinetic energy* ($\geqslant 1$ *MeV*), *lead is very effective inelastic scattering material.* Molten salt solutions are also under study (*e.g.*, lithium–sodium–potassium fluorides). These enable temperatures up to 1000 °C to be attained. This would call for installation of refractory liners in the reactor: ceramics, graphite, one of the 22 so-called "Hastelloy" categories (the name is derived from a diminutive of the Haynes International company, which produce these very corrosion resistant alloys. The predominant metal is nickel). For the pipe work molybdenum-based alloys, or with a high nickel content (INOR-8) are under study. Lastly, the inert gas (such as helium) coolant fluids enable a reactor output temperature of 850 °C, at a pressure of approx. 70 bar (7 MegaPascal; we recall at this point that the cooling water used in PWRs and the EPR is 16 MPa for a reactor output temperature of 320 °C) using ceramics for certain cable-ways or pipe-ways, plus refractory materials for structures with the reactor vessel (some of which use the alloys and ceramics mentioned above), such as oxide dispersion strengthened alloys (ODS: alloys reinforced by numerous very small oxide dispersion). For the materials external to the reactor vessels, all sorts of nickel base super alloys are being investigated. *We note here that material choice and availability are the key factors to future breeder reactor design and their specifications form the real constraint for all designs and for the "Rankine" cycle Turbine efficiency, i.e., determine any future electronuclear power generation plans.*

[10] We could reflect in France, given the number of Pressurized Water Reactors reactor-years of experience we have gained, what component of our generation systems has proved most fragile, whether in the reactor or outside the reactor. The answer by far lies in the heat exchanger systems. The responsible factors include vibrations, strains by fatigue, welding failures, and then stress and deformations, high temperatures and thermal gradients, corrosion, etc. Consequently, these system components must be "replaceable", as on a spare parts list.

[11] A *fissile reaction* between a nucleus A and a neutron (of kinetic energy between 0.02 eV and 10×10^6 eV) is a nuclear reaction, which leads to the division of a *compound nucleus* (A + n) in two (or three, by example, the third part is a *tritium* nuclei) nuclei (total kinetic energy $\approx 170 \times 10^6$ eV), to 0 to 5 neutrons, to neutrinos ($\approx 12 \times 10^6$ eV), to prompt gamma rays ($\approx 6 \times 10^6$ eV); the fission products decay will produced gamma rays ($\approx 6 \times 10^6$ eV), beta rays ($\approx 8 \times 10^6$ eV). This nuclear reaction is written: A(n, f) its important products, for industrial energy production are fission products and neutrons. When this reaction is relatively important, we say that the *nucleus A is fissile*. For the range of incident neutron kinetic energy mentioned above, there is always the possibility of a capture of the incident neutron by the nucleus A. This nuclear reaction is written A(n, γ) $\underline{\text{A} + 1}$. The nucleus (A + 1) is always in an excited state. It comes down his ground state by emitting gamma (γ) rays and successive β disintegrations. These two categories of nuclear reactions are called *absorption* reaction of a neutron by the nucleus A.

[12] Convention: we shall use the term "reactor" for all component parts placed inside the reactor vessel; we shall likewise use the term "reactor core" for those components that contain nuclear fuel and their assemblies (of fuel rods, of control, etc.). If certain peripheral items only contain fertile assemblies, or targets of waste nuclei (for purposes of destruction; in fact, transformation in fission products or another nucleus), or components with a single function of reflecting the neutron flux to impinging on the CE components, we shall use the term of blanket.

[13] A *fertile reaction* between a nucleus B and a neutron (of kinetic energy between 0.1×10^6 eV and 10×10^6 eV) is a nuclear reaction, which leads to the formation of a nucleus B + neutron = C, this C nucleus, after some disintegrations, becoming a nucleus C which is fissile. We say then that B is a *fertile nucleus*. A nucleus can be fissile and fertile.

[14] Among the fission products, the more abundant gaseous are: xenon 131, 132, 134, 136 (15.8% mass); krypton 85 (1% mass). The volatile chemical elements which had contaminated the Chernobyl area was mainly the isotope 137 of cesium (cesium: 8.1% of the fission products). The other contaminant was the strontium 90 (2.6% of the fission product) which disappear quickly, by the run off of water. The iodine 131 (half life: 8.02 days) had a morbid effect (with 0.6% of the fission product for all isotopes of iodine, mainly isotope 131 – half life β: 8.02 days) for short term radioactivity and ^{129}I for long term radioactivity (half life β: 1.57×10^7 years) [7]. The natural isotope of iodine is 100% of 127 isotope.

[15] Definition of "*reactivity*": We define first the *factor of multiplication* of the reactor [k]: It is the *ratio* of the number of fissions in the reactor during a chain reaction (between the birth of a neutron from one fission until his mean time of absorption creates the following fission) to the number of fission of this next generation. Then, we define the *reactivity* [ρ] by $\rho = (k-1)/k$. If $k = 1$ and so, if $\rho = 0$, we say that the reactor is *critical*.

disturbances is that the breeder reactor possesses a *negative coefficient of reactivity* as in all reactors. For breeders, there is only one important possibility: It is that the concept of core of this breeder has a strong *negative Doppler* (*so it is a prompt phenomena*) *coefficient of reactivity*[16] – and the mass density – possible local 'voids' in the coolant fluid, etc., the ^{239}Pu nuclei are going to spend between 4 to 8 years in the reactor vessel, give or take a few planned fuel element displacements. Given these constraints above, the nucleus may change by all the nuclear reactions mentioned. Either, the nucleus may produce neutrons by fission or it will capture neutrons. In the latter case, the nuclear capture reactions will produce, in turn, ^{240}Pu (half life $\alpha = 6.5 \times 10^3$ years), ^{241}Pu (half life $\beta = 14$ years), ^{242}Pu (half life $\alpha = 3.7 \times 10^5$ years), which in turn lead to new fission events, or through capture of one or several neutrons leads to unstable isotopes or disintegrate into americium (^{241}Am, from the beta-γ-disintegration process, half life $\beta = 432$ years and ^{243}Am that results from the capture of a neutron to produce ^{243}Pu, followed by a β disintegration, half life $= 3370$ years) or by different possible and simultaneous nuclear reactions to ^{238}Pu (half life $\alpha = 87.7$ years, used as source of heat and subsequently electric power production for deep space probes where solar panels are no longer a viable energy source), or finally into curium or ^{237}Np neptunium (half life $\alpha = 2.1 \times 10^6$ years).

What we note here is the competition for each nucleus in the core to proceed by natural disintegration or by neutron capture, either path leading in essence to a transformation, also called "transmutation".

So-called "neutron economics"[17] – neutron flux (with their associate kinetic energy and *importance* for the chain reaction – and other desirable nuclear reactions – depends how the neutrons are physically positioned in the reactor, their directions and kinetic energy), depending how the neutrons are physically positioned in the reaction chain, spectrum of kinetic energy of the neutrons in each place of the core, etc.

Fast neutron reactors, i.e., are those in which the kinetic energy of the neutron at the start of a fission event (the average energy level at birth in a fission), is close to 2 MeV[18] (2 million electron-volts) and where this kinetic energy is the least diminished before being absorbed in another fissile nucleus. So, the design of the core has to avoid a dilution of target fissile nuclei (i.e. by incorporating material which do not reduce, by inelastic collisions with neutrons, their kinetic energy). Thereby, it leaves more neutrons available for the ensuing fission and fertile events.[19]

Compared with the case of the reactor cores of PWRs, looking, for example, at the capture ratios of neutrons in the fission products, we see that there is a reduction of neutrons, ranging from 7% of the neutrons per neutron resulting from a fission event for the PWR, to 1.5% of the neutrons per neutron resulting from a fission event for the fast breeder. For the coolant, the proportion of neutrons absorbed directly decreases from, 3.4% to 1%, etc.

This allows us to offset the loss of reactivity due to the fuel combustion irradiation "time length" duration, both: (1) a reduction in the absorption process of the fission products that tend to accumulate and by creation of new fissile material (e.g., ^{239}Pu); and (2) concomitantly those placed in the reactor core at the beginning of the operational cycle are 'burned' and therefore gradually disappear. In this way, the admissible radiation time for fissile fuel elements (FE) can be greatly

[16] *The Doppler Effect of thermal motion of fissile nuclei and fertile nuclei on the reactor reactivity*: in the formulae used to assess the cross-sections (known as "Breit and Wigner" cross-sections), it is the kinetic energy of the neutron in the center of mass of the target nucleus and the impacting neutron, which is the reference system. Thus the positions and the relative velocities are measured in this reference system. For a neutronic computation, the velocities are measured in *the motionless reactor system* (also used is the expression. *under laboratory framework*). As long as the nuclei are immobile, we can easily move from the cross-section in the mass center to the cross-section in the reactor (or the frame work of the laboratory). However, the target nuclei are not in fact immobile in the frame work of the reactor. They oscillate in all directions in the electromagnetic quantum field of the "microcrystal" and depending on the temperature, around an equilibrium position. Therefore, our neutron – that interacts with our moving nucleus – possesses a variable kinetic energy in the mass center framework system, and therefore also a variable cross-section. In another language, this so-called Doppler Effect here widens the resonance phenomena of the nuclear reaction between the nucleus and the neutron. The broadening of the resonance lines by the Doppler Effect leads to an increased fission rate and is therefore seen as positive for ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu. It qualifies as a positive, prompt, reactivity increase. But there are also absorptions in the fissile nuclei with captures of neutrons (so-called (n, γ)). So there is a portion of loss of neutrons for these fissile nuclei, so a prompt decrease of reactivity. As we are studying prompt effect on reactivity, say during a lifetime of a neutron, the effect of our neutron on a fertile nucleus, say ^{238}U gives ^{239}U (half life: 23.5 minutes; fissile), then ^{239}Np (half life: 2.26 days, non-fissile), then ^{239}Pu (half life: 2.41×10^5 years, fissile). But for the prompt effect, prompt meaning between an average life time of a neutron just born from fission and scattered, then absorbed, we have only to consider the first nucleus (less than 10^{-6} seconds), the ^{239}U (absorption of a neutron leads to fission or to capture). As this increase of reactivity is proportional to the increase of temperature of the fuel material (and also, of the fertile material) for a small modification of temperature, we name the ratio of these two modifications (i.e. temperature on the "possibly fissioned" nuclei and fission of this nucleus): the Doppler coefficient of reactivity. We see that the sign of the Doppler effect on the system reactivity is different when neutrons are being created or subtracted (between neutron pits and neutron sources). It is the difference of the two that gives the sign of the Doppler effect on the system reactivity. We have to manage the design of the core to make it negative. It is a very important phenomenon because it is a *prompt* (immediate) negative feedback, so decreasing the reactivity when the temperature of the fuel elements increase. The conclusion is that the Doppler effect on reactivity in breeder reactors is negative, if the fissile and fertile nuclei are appropriately positioned in the reactor space because the physical *importance* of a neutron for the chain reaction by fission and the creation of plutonium by capture, change with his position in the core and his direction of speed. We have to stress that this Doppler effect *changes sign* when a fertile nucleus becomes a fissile nucleus, this being one of the very purpose of the reactor. The Doppler coefficient also changes sign when a fissile nucleus is "split"! So the value and the sign of the Doppler coefficient, changes continuously during the irradiation process! Hence the importance of positioning the fuel elements and the fertile elements in a fast neutron reactor. It is possible that this phenomenon forces the breeder reactor operator to modify during a fuel length (6 to 8 years) of cycle of irradiation the way the fuel elements are positioned when being irradiated. This also stresses the importance of complying with particular safety criteria (notice that this is not the only safety criteria, i.e. the loss of coolant is also a concern) throughout the design stage of the breeder reactor, during its loading, its operation, its maintenance and its complete fuel cycle.

[17] [8].

[18] As the energy equivalent of the mass m of a neutron is $mc^2 = 939.5$ MeV, where c is the speed of life, so the speed of a 1 MeV neutron is $\approx c/30$ and for a 2 MeV neutron, $\approx c/21$.

[19] The spectrum of kinetic energy of one of these neutrons at birth in a fission is spread between 0.1 MeV and 10 MeV.

increased, compared to PWR, up to 8 years (or much more for the destruction targets). In other words, the energy one can draw from a given mass of new fuel positioned in the reactor core (so-called "burn up"[20] can be increased from ≈ 40 *in a PWR to* ≈ 200 *MW, days/kilogram of actinides*[21] ([9], p. 442) loaded in the breeder reactor.[22]

Given that the neutrons possess a high kinetic energy – in a range where iron,[23] and its alloys do not readily capture the neutron flux – extensive use can be made of stainless steels alloys (which are far better for oxidation resistance than zirconium alloys) for the fuel rod assemblies and for the internal structures for the reactor core vessel (they are also used in PWR). Steel is an alloy for which we now have accumulated an enormous degree of experience and wide set of compositions available enable us to face any of the reactor environments, whether they be chemistry, physical chemistry, physics, mechanics, thermal transmission, thermodynamic. Which is more, research and progress on steel is always active and fruitful. Steel therefore complies fairly easily with the *technical specification requirements* (so-called *terms of reference*), for a given function assigned to the material in a given setting. We can thus *capture the largest available number of neutrons* by *using fertile nuclei* such as ^{238}U and therefore generate a *largest* number of fissile nuclei compared with the number introduced at the beginning of the operational core cycle.

Let us now summarize this brief overview of *static core "neutronics"*, by stating that one neutron is 'born' for every 0.36 fission events (of which 0.04 fission events for ^{238}U and 0.03 for other actinides[24] formed in situ[25]). What we can readily note is that the number of fission events in the ^{238}U per neutron born in another fission event, is relatively small, compared with the number of fission events that take place in the plutonium. Nevertheless, we must recall that the number of nuclei present in the ^{238}U is approximately 5 to 6 times[26] higher that the number of Pu nuclei. The reason for this low number of ^{238}U is that the fission threshold for ^{238}U lies in the range 0.8–1 MeV.[27] The spectrum of kinetic energies for the neutrons, at time of birth in a fission event, is ≈ 2 MeV. However, the neutron will cover a distance of ≈ 20 cm before being absorbed by fission or by capture. In that interval, the neutron will collide with nuclei and undergo reactions (n, 2n), and these inelastic, anisotropic collisions will tend to decrease the average kinetic energy of neutrons, depending on the way the reactor core is composed and assembled. Finally the *average*[28] kinetic energy of the neutrons will be, following the design of the core and of the coolant structure, from 0.3 to 0.5 MeV.

The number of neutrons captured by ^{238}U (the fertile body), per neutron escaping the core, the blanket and reflector is a fission event, is 0.48, thus producing 0.48 ^{239}U $\rightarrow$ 0.48 ^{239}Np 239 $\rightarrow$ 0.48 ^{239}Pu.

We should, to be complete, add 0.13 sterile captures and 0.02 neutrons escaping the core, the blanket and reflector 'lost' neutrons for each neutron born in a fission event.

So, the summary of the life of a neutron is: *born from a fission event, father in the chain reaction to the next absorption of this neutron, giving a fission, with one of the neutrons emitted from this fission, following the same story of random walk and then eventually*

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[20] Another characteristic of irradiation is the "*burn up fraction*". The unit is [atom per cent]. It is the ratio of the number of actinides nuclei "fissioned" to the initial number of actinides loaded in the reactor. By example, for a PWR the burn up fraction is $\approx 4\%$ (≈ 2.5 to 3% from ^{235}U and $\approx 1.5\%$ coming from ^{239}Pu created in situ by capture of a neutron on ^{238}U). The correspondent burn up is ≈ 33 to 40 GW day/ton uranium. So the relation between these number measuring the same phenomena in a PWR is: 1 atom fissioned $\approx$ 9–10 GW d/ton of ton of actinide.

[21] This is a very important result with large practical consequences. As the reactivity of the core of a fast neutron reactor vary very more slowly than in the PWR, we can afford to burn a great fraction of the actinides loaded in the core. In place of the $\approx 3.5\%$ of uranium of PWR, we can accept $\approx$ 18–23%, of nuclei fissioned (coming from the Pu isotopes loaded in the reactor and the Pu isotopes created in situ by captures of neutrons on ^{238}U) which, with the factor 8.5, means $\approx$ 150–200 GW day/ton of oxide. Moreover, it follows that we can irradiate the fuel elements of a breeder, without unloading them, during 7–8 years (in place of $\approx$ 2–3 years of PWR). We stress that this means that the materials for the fuel elements and their structure in the subassembly, etc. had to sustain very high doses de dpa (i.e. 200 dpa/atom). It needs a fundamental research effort, in account to be safe in all situations.

[22] One of the consequences is that is that the fuel element taken out (so-called "spent fuel" element) of the reactor breeder are very much warming in the water pool than in the case of PWR. This means that one may be obliged to wait a longer time to make some operation. There is also a big difference between the residual power of a breeder fuel element made with *plutonium*, or plutonium with *americium homogenized* fuel assembly or with *americium heterogeneous*: For the first one, one has to wait 200 days of cooling time for to cool to 7.5 kW (maximal thermal power to clean the fuel assembly). For the second, 500 days of cooling time are necessary; For the last one, we have to wait 4000 days of cooling time.

[23] The capture cross section (n, γ) of iron (^{56}Fe) is around 1 barns (one barn $= 10^{-24}$ cm^2) from a neutron of kinetic energy 1/10 eV to 10^{-3} barn for a neutron of 2×10^3 eV. There is a lot of captures lines of resonances from an energy 3×10^4 eV until 6×10^5 eV. For a neutron of kinetic energy between 0.3 and 10 MeV, the capture cross section is around 2×10^{-3} barns. The case of sodium (^{23}Na) is similar in the spectrum of energy of fast neutrons, but with a lower cross section.

[24] The *actinides* (from the Greek word "actinos" = ray), are the elements that run from the actinium (Z = 89, A = 227) not including ^{227}Ac, and therefore from ^{232}Th, thorium, up to and including 260 which symbol is "La" (lawrencium with Z = 103; A = 260) that gradually fill their 5f shell (maximal number: 14) with their electrons; the thorium 5f electron shell is empty, while that of uranium has 3 electrons 5f, plutonium 6 electrons 5f, americium 7 electrons 5f, and curium 7 electrons 5f, [10].

[25] This occurs even when no transuranic elements are introduced with the plutonium and the ^{238}U. If, for example, one introduces some americium Am 241 (half life $\alpha = 432$ years) $\rightarrow$ neptunium 237 (half life $\alpha = 2.1 \times 10^6$ years), Am 243 (half life $\alpha = 7370$ years $\rightarrow$ Np 239 $\rightarrow$ Pu 239), these elements lead to additional fissions and neutron captures and residual thermal power.

[26] See below for an explanation of the number ≈ 5 to 6. It is equivalent to say that the ration of ^{238}U (85%) to the number of Pu (15%) $= 85/15 = 5.6$.

[27] The ratio of the fission cross-sections for ^{238}U and the capture cross-sections, i.e., $\sigma_f/\sigma_c = 1$ for a neutron with incident kinetic energy of 1 MeV, on the ^{238}U nucleus. and this ratio is approximately 100 for a neutron with incident kinetic energy of 2 MeV. This ratio is approximately 100 for a neutron with incident kinetic energy of 2 MeV and 0.01 for a neutron of 0.6 MeV. We must stress that the spectra of neutrons are different between sodium cooled oxide fuels, sodium cooled metal fuels, gas cooled and lead cooled reactors. Among them, the spectrum neutrons of the French fast neutron reactor RAPSODIE at Cadarache/CEA was the "hardest", with a maximum at 1 MeV.

[28] Do not forget that the transport of neutrons in the core is a process of probability. There are virgin neutrons ("Virgin neutron" = A neutron with no collision with nuclei since birth by fission) at 2 MeV and more (the fission spectrum follows also a probabilistic law) and lower kinetic energy neutrons (roughly from 0.1 MeV to 10 MeV).

absorption and fission, with descendant, itself creating a fission event and more neutrons, all this chain reaction with further neutrons descendants from the single father.

However, the absorbed neutrons of these successive fissions can be also captured by the ^{238}U, which we call the fertile material.

Finally, the ratio of the number of fission[29] events in the ^{239}Pu to the number of neutron captures in the ^{238}U, is ≈ 0.8. If we take in account the fissions in ^{240}Pu, ^{241}Pu, ^{238}U, ^{235}U, this ratio is ≈ 1.1. In a PWR, the number of fissions divided by the number of neutron fertile captures in the ^{238}U is ≈ 1.5 [12].

When all the design of the reactor is fixed, there remains a possibility of *trade-off between the absorption leading to a fission, and the absorptions creating a fissile nucleus from a fertile nucleus* (*and different categories of losses*: *the neutron flux level* [*number of neutrons*/*cm*^{2} *second*]).

Over and above the relative proportions of the fissile, the fertile material, the structures, the control systems, etc., that are introduced at the beginning of an irradiation sequence that is going to last several years, what are the other factors that can have an influence on the figures above? The fast neutron flux[30] determines the number of nuclear reactions per second, therefore in fact all the nuclear reactions mentioned earlier for a given breeder reactor.

But the disintegrations we mentioned (α and β disintegrations, etc.) have *fixed probability distributions* (*independent of the neutrons flux, for a given nucleus*) *hence also with a fixed half life*. So their proportion per neutron born in a fission does not depend on the neutron flux or the spectrum of kinetic energy of the neutrons in the core.

It therefore becomes possible, by deciding on the level of neutron flux, to give an advantage to one or another nuclear process. We shall see later the margins that limit these degrees of freedom here. To be more precise, for a given reactor core, if one wants to multiply, for example, by a factor, say N, the neutron flux, then one increases the ^{239}Pu (isotope 239 of plutonium), production per unit time, the power output of the reactor by the same factor N increases the production of ^{240}Pu by a factor N^2, and the production of ^{241}Pu by a factor N^3. The process of production of transuranic is more and more non-linear, promoting the production of transuranic nuclei of the highest atomic mass.

Thus, the process whereby the transuranic elements are formed is not proportional to the neutron flux level. In another language, it is not linear. The quantity of transuranic elements formed in situ increases at a far faster rate than the neutron flow, in *all* nuclear power generation reactors. However, given that the capture cross-sections are much smaller when the energy levels approach approx. 1 MeV than for the thermal neutrons, the number of neutron captures per fissile nucleus 'burned' is far smaller (roughly, a factor "thousand"[31]) in breeder reactors than in thermal neutron reactors (PWRs etc.). Moreover, this is where other processes occurs, the disintegration ($^{241}\text{Pu} \rightarrow {}^{241}\text{Am} + \beta +$ neutrino), which is a process *independent of* N. There is therefore a competition between the two processes (absorption of neutrons and disintegration β or α) or spontaneous fission (in this case, with emission of neutrons), the outcome of which depends on the scale of the neutron flux.

3. How does one design a useful fast neutron reactor core? What are the constraints? Useful for which optimal objective? What are the special constraint for a breeder? And with what optimal objective?

A first fundamental result: compared with thermal neutron reactors, the number of the fissions per new born neutron are decreased while the number of the captures increase. The reactor can no longer reach criticality in the same proportion of fissile to fertile material as the PWRs. We can no longer content ourselves with 3 to 4% ^{235}U in the heavy metal ^{238}U. The ratio needed for fissile to fertile matter attains 15 to 30%, not only at the start of the chain reactions, but *over the operational years* that allow us to draw several MW/day per kilogram of heavy matter.[32] If we wish to use the reactor to "burn" various actinides (over and above those produced in situ by neutron capture and disintegration), operating time of heating them in the reactor will be much longer than the fuel elements and the fertile elements. Moreover, the length of time of heating for each nucleus before destruction will be different for each nucleus. This concentration will cost much more per unit volume of core than the fuel used in PWRs.

A second fundamental result: The conversion of the ^{238}U nuclei into ^{239}Pu nuclei, multiplies the potential number of fission events, conditioning in like manner the amount of energy that can be extracted from a given mass of ^{238}U by a factor $99.27/0.72 \approx 140$. If we include all forms of possible loss, the multiplication factor for a given mass of uranium is approx. 60.

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[29] In a PWR, most fissions occur in the 0.1 eV thermal peak [11].

[30] All these numbers related to neutrons economy come from the design of a example of 1200 MWe fast *breeder* reactor [12]. They may change with another design. But the orders of magnitude remain the same.

[31] This big difference between the cross section for nuclear reaction between the MeV and the tenth of an eV of kinetic energy E of the neutron comes from the fact that the cross section for nuclear reaction, at low energy are proportional to $\lambda^2\Gamma$ (formula of Breit–Wigner where Γ is the width of the resonance line shape and λ is the probability for decay of the compound nucleus per unit time). λ^2 is proportional to $1/E$. Γ is proportional to $E^{1/2}$. So between 1 eV and 10^6 eV $= 1$ MeV, there is a factor 1000. In casual language, a slow neutron spends a larger time at the proximity of the nucleus.

[32] Or in equivalent terms, to fission $\approx 70\%$ of the ^{235}U in a PWR.

3.1. What are the limitations for fast neutron flows and their kinetic energy spectrum?

The quantity of fissile matter (^{239}Pu with a high proportion of isotopes present[33]) implies that the cost of fissile matter (always expensive, whatever the choice, ^{235}U in PWRs and ^{239}Pu in the breeder reactors) as0. needed to be cost effective per cubic decimeter [dm^3], provides the highest possible amount of thermal energy, at the highest temperature compatible with the materials that contain the coolant fluid.

3.2. The thermodynamics of the contents of a breeder reactor and possible consequences. Thermal density in the reactor and its evacuation

In PWRs,[34] the power transfer rate from the core via the coolant fluid is approximately 100 to 170 kW/cubic decimeter (dm^3). Since the start of studies on breeder reactors, in the nineteen-fifties, attempts have been made to extract 3 to 5 times that amount, viz., from 300 to 500 kW/dm^3.[35] This approach calls for a choice of liquid metal coolants given its high thermal specific capacity at, the operating core temperatures (i.e. sodium-high boiling point: 893 °C; but which burns when meeting water:

$$2Na + 2H^2O \rightarrow H^2 + 2Na^+ + 2OH^-$$

followed by explosion if there is oxygen, sodium–potassium, lead or eutectic[36] mix, etc. which are at ambient pressure for the temperatures under consideration, or of an inert gas such as helium,[37] with the additional difficulty of the high pressure needed to obtain a high caloric transfer rate, and therefore a high mass density and, lastly, molten salts).

3.3. Materials temperatures and their limitations

The other significant characteristic of these "nuclear heaters" is the coolant output temperature. The best result to date, for the materials mentioned is to move up form the 320 °C we have for PWRs to approximately 550 °C (this increasing the PWR turbine efficiency factor from 34% to more than 40%) with an improved steam circuit in the turbine system.

One of the consequences of having *a much higher power transfer density in breeders reactors compared with PWR*, for a given reactor core thermal power output is that the core itself must be much smaller in volume. To evacuate the heat production from the reactor core, one needs to have a much higher temperature rise: the input and output temperatures respectively for a PWR and a breeder reactor are: PWR 290 °C/320 °C and for a breeder reactor 370 °C/550 °C; the cladding of the fuel elements, at the exit of coolant from the fuel assembly[38] can be as high as 620–630 °C. This of course implies a high temperature gradient for the materials at the core coolant input and output that the materials specifications must take into account.

We must be aware that the temperatures and the choice of coolant fluid, *for example, sodium*, had required a complete development of a new technology: pumps, valves, pipe work, heat exchangers sodium/sodium[39] and sodium/water, wash-out devices, waste cock for the sodium, free surfaces of sodium treatment, fuel assemblies, fertile assemblies, control rods and other, objects manipulation in an *opaque* environment, fabrication processes with radioactive materials emitting neutrons, welding, pipe work, shaping, corrosion, proofing techniques, *absolute separation principle for sodium, water and air, etc.*

To summarize, a technology and associate know-how are needed appertaining to heat transfer and evacuation, power densities starting at the fuel cell surfaces, through the heat exchangers[40] and leading to high temperature, high pressure

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[33] Cf. the example of the isotope compositions of plutonium coming from radiation of the MOX fuel elements used in PWRs that are authorized to do [13] pp. 109–110. The proportion of the 239 isotope is ≈ 38%, the ^{240}Pu is ≈ 32%, the ^{241}Pu is ≈ 14%, the ^{242}Pu is ≈ 13%. Another number useful for simple computations, is the neutronic reactor economy of theses isotope: If we fix the importance for the chain reaction of 239 Pu at 1, the "equivalent" of the other isotopes of plutonium are, respectively: ^{238}Pu = 0.43; ^{239}Pu = 1; ^{240}Pu = 0.13; ^{241}Pu = 1.5; ^{242}Pu = 0.03; ^{241}Am = −0.35. All these numbers relate to a given core, a local neutron flow, a given spectrum, temperature, length of time of irradiation, initial isotope composition of the fuel, rearranged fissile and fertile fuel elements, etc., data. These are only reference values for the reader. We have retained 2 significant figures to stay coherent with the reference values.

[34] [14].

[35] By way of a comparison: in a modern aircraft, the average power delivered by the combustion chamber of the turbine of the engines is approx. 4 MW/dm^3; for a road car the value is approx. 2–3 MW/dm^3, [15] for the projects of magnetic fusion tokomak, 1 kW/dm^3, or with the same units, 0.001 MW/dm^3. This is why the tokomaks experiment volumes are so big.

[36] Russia has forty years of experiment with lead–bismuth coolant on his submarines, but without know-how in France.

[37] For comparison of the spectra of fast reactor concepts, and for neutrons, under the kinetic energy of neutrons of less than 0.1 MeV, the gas cooled lead to more captures in the low part of the energy spectrum [11]. The advantages of helium stems from its inertness in that it is a single phase coolant. But it needs pressure of ≈ 90 bars.

[38] The main characteristics of a fuel assembly of a breeder electronuclear plant is, taking the example of a real designed (European Fast Reactor: EFR) electro power plant: Total length: 5.4 meter; Weight: 580 kilogram; Core height: 1 meter; Axial blanket height: 0.3 m. For SUPERPHENIX, the number of sub assembly was 364 fuel and 233 fertile subassembly. The number of so-called pins (we use also the word "rod") per subassembly is 271 [9].

[39] The material which seems today the best tradeoff between all the mandatory is steel 316 L(N) with 9% chrome, 1% molybdenum, small amount of niobium (L for low carbon; N for controlled nitride).

[40] For the different categories of heat exchangers (sodium/sodium; sodium/water, etc.), thermal conductivity for the steels used must be as high as possible. Experience tells us that it is one of the more fragile components of a power station, and they must be dismantled and changed throughout the operational life-time of the station.

steam, heat source entering the turbine, going through in the best thermodynamic cycle and heating up the cold source, a condenser unit, and above all, redundant safety circuits and devices, including electric power circuits.

3.4. What are the real limits for all these factors?

What "signal values" can be used? As an example for a signal value, we can point to the requirements for material radiation resistance: *"fluence" of neutrons* [number of neutrons crossing a unit surface of the material during the length of irradiation inside the reactor] [number of neutrons/cm^2] or *the number of displacements per atom* [*dpa*][41]? Among the materials specifications[42] used for such equipment and fabrication processes, we can cite the radiation resistant factors. But what are the good indicators or signal values for possible radiation and thermal damage situations? We could perhaps refer to the fluency of neutrons at a given place in a given material sample; or to the radiation energy level (neutrons? gamma rays? etc.) that crosses the material, or the energy gradients observed. To test the relevance of a fluency factor, let us briefly examine the case of a 316 stainless steel sample and measure the change in the yield stress [in MPa = megapascal] as a function of the neutron fluency[43] somewhere between 10^{17} to 10^{20} [number of neutrons/cm^2 with its kinetic energy in excess of 0.1 MeV. Three measurements with variable neutron spectra, carried out in three independent laboratories give highly different results, with a factor close to 10! The same experiments based this time on the "displacements per atom" [dpa] give three superimposed plots between 10^{-3} dpa and 10^{-2} dpa.[44,45] Displacement per atom control the number of point defect created by irradiation which in turn, will control the density of structural defects (i.e. dislocation loops), which will contribute to hardening. In materials such as stainless steels, this contribution is dominant.

4. What breeder reactor equipment can be considered as new technology and difficult to maintain?

4.1. The pumps and all rotating machines, the valve, seals and tightness proofing, the pipe work joins and connectors, etc.

Let us take a technological example: development:

- of pumps and all turning machines, valves seals and proofing, pipe work joins and connectors, etc., the state of the inert gases overlying the free sodium surfaces;
- sodium purification;
- the two types of heat exchanger vessels (sodium/ sodium and sodium/water and generated steam);
- identification of positions in the reactor full of sodium of fissile and fertile fuel elements (FEs);
- the machinery used to handle the FE into and out of the reactor core, either for the purpose of changing the current neutron flow, distribution, hence the power output of the reactor, hence the core temperature, or to remove them completely from the reactor vessel, continuing the sodium cooling function and placing them in an intermediate storage system for cooling and wait for the disintegration of highly radioactive substances in the fuel elements and the fertile elements and other materials in sodium;

[41] "The *dpa* is the number of collisions that each atom of a sample of solid material, completely defined had been displaced from his site in the material sample by the action of one and only one piece of nuclear ray t (i.e. one neutron of de 1 MeV). For the ejection of one atom from his site in the material sample, this atom needs a to absorb from the collision with our neutron an energy greater that a threshold energy, called threshold energy of displacement, called the *threshold energy of displacement* E_d. This threshold energy is a function of the direction of our neutron and the direction taken by the displaced atom, compared with the nature and the direction of the crystal lattice of the material sample, by example, fcc for iron material. In this case, $E_d \approx 40$ eV. Generally, with our neutron of kinetic energy $E = 1$ MeV, many displacements of atoms in the sample of material will occur. As a first approximation, the number N_d of displaced atoms is: $N_d = E/2E_d$. One part of the displaced atoms will find a new equilibrium site in the sample of material after some lattice vibrations and will not participate to the damage by irradiation. Some other atoms displaced from their site will let a *vacancy* in the lattice and will stop as an atom distorting the lattice in a place that they created and that is called "*interstitial*". For the kinetic energy 1 MeV of our neutron quoted above, there is roughly 40 pairs of vacancy-interstitial, called Frenkel's pair. The distance between a vacancy and its interstitial is calculated by molecular dynamics simulations that gives roughly 100 interatomic distances" [16].

[42] [17], pp. 649–655.

[43] "*Fluency*": number of neutrons crossing a surface of 1 cm^2. At a given structural position, chosen by a metallurgist, we measure the number of neutrons crossing the section chosen for a given time, i.e. the length of irradiation of the fuel elements. For example, in a PWR, the resulting thermal neutron flow, over a two year time interval, is $\approx 10^{14}$ neutrons/cm^2 second – the measurement point being a position chosen on a fuel assembly cladding, for example, giving a fluency of $3 \times 10^{14} \times 3.15 \times 10^7$ seconds/year $\times$ 2 years $= 6 \times 10^{21}$ neutrons/cm^2. In a breeder, the neutron flux is $\approx 6 \times 10^{15}$ neutrons/cm^2. The length of irradiation may be longer to burn efficiently the transuranic nuclei. So the fluencies at different positions are in the several $\approx 10^{22}$ n/cm^2.

[44] [18] Fig. 1.1, p. XII and pp. 112–113. "An example applied to a structure with an iron content in a breeder reactor is as follows: we are looking for damage to the iron content due to the impacting flow of kinetic energy neutrons, 0.5 MeV, in the heart of the core of a breeder reactor; the mass density of the iron is $N = 0.85 \times 10^{23}$ iron atoms/cm^3. In the fast neutron spectrum of a breeder, the diffusion cross section is $\approx 3 \times 10^{-24}$ cm^2. The neutron flux in a breeder is several $\approx 10^{15}$ neutrons/cm^2 seconds. The number of iron atoms displaced by the neutrons is 350 (cf. Ch. 2) "displacement of atoms" in [18]. These figures imply that the number of iron atoms displayed per cm^3 and per second is 9×10^{16}, viz. by dividing by N, approx. 10^{-6} dpa/s. This in turns implies, in a breeder, that the iron crystal network, for a given atomic site, that this iron atom is displaced once every 12 days. By an analogous calculation, in the case of a thermal neutron reactor, *we deduce that a structural atom will be displaced once every* 6 *months*" (from [18]).

[45] We quote here two characteristics of irradiation in a reactor. To be complete, let us add for the fuel elements: (1) The energy released during the period of irradiation per unit mass of fuel element. It is expressed in Mega Watt day per kilogram [MWd/kg] of oxide for fast reactor; (2) The percentage of heavy atoms (uranium + plutonium) "fissioned" in the fuel element during the irradiation. It is expressed in [%]. The relation between these two quantities, for a given irradiation is 1 fuel atom "fissioned" % $\approx$ 8.5 MWd/kg produced.

- the reactor vessel;
- the safety circuits and devices;
- the system output through the civil engineering interface (the containment building which is the concrete barrier that ensures system integrity and overall safety should a reactor accident occur), via the sodium pipe work, etc., called for enormous efforts in terms of systems design and technical solutions. All that gave positive results in France if we judge by our knowledge on reactor physics, sodium circuit technologies, on the reactor vessel, the containment building, the safety equipment the fission fuel handling devices.

For the rest of the *fuel cycle* (chemistry separation of each actinide element, "refabrication" of fuel elements with plutonium resulting from the previous irradiation cycle, refabrication of fertile elements using the already radiated uranium existing in the breeder core, for possible radiation of spent wastes, etc.), in short, for the chemistry,[46] the physico-chemistry, the materials under radiation and high temperatures, substantial work remains to be done. To proceed we need a new set of tools (such as irradiation facilities, hot laboratories, instrumentation, etc.). A first iteration of this important program is to define its terms of reference.

4.2. This previous object-oriented analysis should be crossed with a scientific analysis

Fundamental of radiation science of the materials, whether the functional (materials of fuel element, fertile element, control rod, etc.) equipment or the structural elements (at 180–200 dpa) is still a domain that calls for further investigations.[47] In particular, the plasticity of materials under irradiation and post irradiation, the strain localization induced by irradiation and its relevance to embritlement are of utmost importance.

Instruments to measure radiation appropriately are needed as well as specific analytic laboratories.[48] The last system that met this requirement was the PHENIX Reactor at CEA/Marcoule (France). A new project is under way. It is primordial to avail of such a new radiation measurement facility.

- *Corrosion* in sodium transfer pipes is far less severe[49] than for water circuits, in the PWRs and the associate steam generators. Therefore, there is a sharp drop, compared with water reactor and their steam generators. In this case, the corrosion products, created in the heat exchangers, as they pass into the reactor inside circuit, and became radioactive, will continue to cycle endlessly through these circuits, depositing radioactive corrosion material on all parts of the sodium circuits (including pumps, valves, etc.). The fact that sodium is only slightly corrosive (compared to water) with respect to the material used in the cooling circuits is therefore a significant advantage for sodium circuits.
- Thermal aging of Fe Cr alloys for fuel cladding and irradiation accelerated aging [22].
- Mechanical parts resistance, to stresses, creep damage,[50] material fatigue, torsions, buckling, bowing, vibrations, etc.[51]

Currently accumulated experience: there are now ≈ 390 "reactor years" experience in the world.[52] The breeder reactors functioning today are in Russia, Japan, India and in PR China. The faster breeder reactor operators in France have decommissioned their facilities, for political reasons.

5. The intrinsic stability of the total power of the reactor with respect to probable disturbances

We have addressed various functional aspects of a breeder reactor, working in a *continuous operating mode*.

If *a planned or unplanned disturbance occurs in the core of the reactor, or in any factor entering or leaving the reactor* (*in this case we talk about "control-command"*), *what will happen?*

The feedback "triangle" is formed by neutronic reactivity, thermodynamic power level of the reactor, temperature and – mass density cross-section per unit volume reactivity.

This perturbation of the reactivity of the core of the reactor sets in motion three[53] categories of phenomena that interact:

[46] The chemical properties of plutonium are set out in [19], pp. 29–50.

[47] To give a frame of reference, the main irradiation characteristics of fast reactor fuel sub assembly, was, in dpa/atom%, a: RAPSODIE: **4**; PHENIX: **7**; b: SUPERPHENIX: **9** [9].

[48] Among many possible examples, we can cite [20] and [21], pp. 28–34.

[49] The austenitic steels have a good resistance to corrosion by sodium.

[50] "The cladding must withstand the internal pressure of the fission gases. The Wrapper tube must resist to the difference in sodium pressure between the inside and the outside of the tube, which is ≈ 3.5 bars at sodium entry in the pin bundle" [9], p. 36.

[51] All of this constitutes a high-precision device; the French nuclear scientists and engineers have followed the scientific, technical, industrial and operational paths, and were "winning the day" until a High Authority decision stopped the main experiment and had the site placed under demolition orders, with no scientific, technical, industrial financial or safety factor forthcoming to justify the decision.

[52] For water cooled and/or moderated reactors, such as the PWRs, and others categories we now have fifteen thousand "reactor years" experience (World Nuclear Association).

[53] This is why we name it the "triangular" feedback.

(a) *Variation of reactivity*[54] $\boldsymbol{\rho}$ of the reactor core modifies the *neutron flux* (noted $\boldsymbol{\Phi}$ neutrons/cm^2 second) for any given point M in the reactor core. The phenomenon includes birth of neutrons per fission, absorption of neutrons by nuclei to give fission – and then neutrons – or capture of neutrons, diffusion (elastic and inelastic, of neutrons, etc. All these phenomena are called transport of neutrons inside the reactor vessel. This field of studies is known by the name "*NEUTRONICS*". The rate of change (length-of-time between a perturbation on the reactivity ρ of the core and its consequence on the change of neutrons flux). command the rate of increase of the chain reaction. In more physical terms, it depends on the length-of-time, in a given chain reaction, between the birth of a neutron by fission (or emission of a neutron by a fission fragments[55] in an excited state) and its absorption by another fission nucleus, if it leads to a fission. This length-of-time,[56] therefore, represents the flight time, including free flights elastic and inelastic collisions, up to the point of absorption. In PWRs the time is $\approx 10^{-3}$ seconds. In the breeder reactors, it less than $l \approx 10^{-6}$ second: this is one of the fundamental difference between thermal neutrons reactor like the PWR and the breeder.[57] A positive perturbation on the reactivity ρ increases the neutrons flux exponentially, through a neutron transport phenomena with a period of approx. l seconds, hence an increase in neutron flow Φ.

(b) This in turn, where the number of fission events occurring per unit time in the core of the reactor, increases the thermal power created in the core (i.e.) per dm^3. Hence, it results from the neutronics of the core, that there happened a fast increase in the total thermal power generated **P** in the core [in MW = megawatt].

(c) The increased power **P** in the reactor core leads inevitably to a rise in temperature of all the inside of the reactor vessel, an increase in volume by dilatation and consequently a *decrease in the number of fissile nuclei* (*noted FN*) *per dm^3*. Both these phenomena belong to the field of *THERMODYNAMICS of the inner reactor vessel.*

(d) *A prompt feedback and the strong Doppler effect on reactivity*: (for Doppler feedback, see more details in footnote 9).[58] These phenomena in all the fuel elements, the fertile elements, etc. constitute the third part of the feedback[59] to the initial perturbation. If we want the nuclear chain reaction to spontaneously return to its initial null value, viz., the value it had before the perturbation occurs, then the feedback t must be negative vis-à-vis the initial perturbation of reactor reactivity.

(e) The difference between a PWR and a Breeder is that the PWR had a powerful feedback in the dilatation of water, which is always great and negative. So the Doppler effect in PWR is of second order. In a breeder, there is no high density of the number of hydrogen and oxygen with a neutronic importance. The sodium is quasi transparent. There remains only the Doppler effect, but strong and prompt. Moreover, the sign of the *Doppler effect of a given nucleus* changes when as a fertile element nucleus capture a neutron and then become fissile. So the management of the places of each fuel element and each fertile element must be a careful handling during all the length-of-time of irradiation.

– *Fraction of slowed neutrons and the period for flux variations*: in a PWR, in order to cancel, other than by a prompt feedback, the effects of a perturbation of the reactor's reactivity (i.e. via thermodynamic considerations related to the coolant fluid, deformation of a fuel element or of an internal core structure, etc.), there is a mechanical device used to lower the number of nuclear reactions, following suit to a perturbation; the neutron flux growth rate (first side of our feedback triangle) must not exceed the exponential increase of the period which is the lifetime ($\approx 10^{-3}$ seconds) of one average neutron. In a breeder reactor, this period is $\approx 10^{-6}$ seconds. It is too fast for mechanical device like "control rods".[60] But the fission process enables a great increase of this lifetime of the average neutrons naturally. When a ^{239}Pu nucleus undergoes fission, certain fission nuclei – not counting the prompt neutrons – produce successive β[61] disintegrations, moving into a nucleus which is a neutron emitter. The proportion of these neutrons emitted and delayed with respect to the initial fission event,

[54] The reactivity ρ of the reactor core is the difference between the multiplication factor K of the chain reaction and the value 1 (corresponding to the stable operational status, known as criticality), divided by K. In the normal operation of a breeder, the reactivity is less than 0.01.

[55] We shall see later in this paper that the proportion of these neutron emissions, compared with the neutrons born simultaneously with the fission events, is approx. 1%. The time lapse, between the separation of the two fission products and the possible emission of a neutron by one of them, is approx. 10^{-17} second. The time between the capture of a neutron and the emission of both fission products is approx. 10^{-14} seconds. This is the time during which the nucleus, composed of a fissile nucleus and an absorbed neutron evolves through various excited states among which one event of division between 2 fragments of fission (but some times three, conducting to tritium), among two among ≈ 160 different fragments of fission (each one with his own disintegration chain of up to six successive disintegrations β) and 0 or 1 or 2 or 3 or 4 or 5 neutrons will be emitted. One of these t is the fission under consideration [23].

[56] This time-lapse is a random variable, and possesses a mean value, noted 1 [second], and called the half life of the neutron.

[57] The shortest length-of-time of the process of the chain reaction is a metallic sphere of pure fissile matter, and in this case the time is $\approx 10^{-8}$ second.

[58] Another effect of increased core temperature and depending on the nature of the coolant fluid, a variation in the density of this fluid.

[59] The three sides so to speak of the triangle of the feedback: first side: Δ reactivity ($= \Delta\rho_1$) $\rightarrow \Delta$ flux of neutrons ($\Delta\Phi$) and ΔP (thermal power); second side of the feedback triangle $\Delta P \rightarrow \Delta$ mass density $= \Delta$ number of nuclei per unit volume and $\rightarrow \Delta t$ (temperature); third side of the triangle feedback: $\Delta T \rightarrow \Delta$ "broading" of the absorption lines resonance (Doppler effect) $\rightarrow \Delta$ absorptions in fissile and fertile elements $\rightarrow \Delta$ reactivity ($\Delta\rho_2$) $\rightarrow$ add to $\Delta\rho_1$. If the fissions absorb more neutron than the captures, the gain in reactivity $\Delta\rho_2$ is positive and then also the feedback. In the contrary, if the captures are more important, the gain in reactivity $\Delta\rho_2$ is negative., and so the feedback. There is a stability process.

[60] The complete process of detection, signal, movement, efficient effect, etc. of the control rods last ≈ 2 seconds.

[61] Therefore controlled by weak interactions, hence the relatively long delay of these nuclei (0.2 to 54 seconds for ^{239}Pu instead of 10^{-17} seconds for the prompt neutrons).

is approximately 0.6% for ^{239}Pu[62] and 1.5% for ^{238}U[63] (in a PWR, this proportion event for the ^{235}U is $\approx 1.58\%$). A relative abundance in the breeder reactor core is, on average of time, 0.8%. The period of the exponential rise of the neutron flow is $\approx$ some tenths of seconds. This period is compatible with the measurements of the phenomenon and the mechanical introduction of an object acting as a absorbent of neutrons in the core of the reactor,[64] the so-called "control rods".

– *Degrees of freedom* required to improve the intrinsic stability of neutrons flux: the stability of the reactor core neutron flow, vis-à-vis all the perturbation sources, is obviously *a priority for the operator of a breeder reactor*. The degrees of freedom to act on this stability factor are, notably:

(1) the way the fertile materials and fissile materials are placed in the reactors (and other possible material one wishes to destroy when the total cycle of his destruction, "from birth to grave" is possible and more advisable than other processes;
(2) the combustion rate for the fissile and the fertile elements placed in the reactor core;
(3) the initial percentage for the fissile materials with respect to the capturing materials among which certain nuclei, born in situ or not, one wishes to destroy);
(4) the neutron flux;
(5) the neutron spectrum;
(6) the nature of the neutron reflector placed round the reactor core;
(7) the nature and the quantity of the radioactive materials introduced into the reactor core or the reactor cover, so that they can be destroyed by fission, or the capture of a neutron or any other process, etc.

A *trade off* between these considerations have to be given *a priority in the specification for future nuclear power generation stations* (*the so-called "terms of reference"*).

The thermal power dynamics of a reactor:

– *The relevant unit of time*: we saw, above, that the relevant time unit for the dynamics of a reactor is the sum of the in-flight length-of-times between the birth of a neutron by fission, mainly fission of ^{239}Pu, till absorption by a fissile nucleus (^{239}Pu or ^{238}U or ^{233}U, etc.), or by a fertile nucleus (^{238}U, etc.), by capture in a structural reactor component or finally by total loss, i.e., by the neutron leaving the reactor volume inside the reactor vessel: in terms of prompt neutrons, we saw that the flight time was in the order $\approx 10^{-6}$ seconds and in the case of delayed neutrons, $\approx$ some tenths of second.

– *The unit for the chain reaction*: we also saw that the proportion of the number of delayed neutrons with respect to the prompt neutrons coming from the same fission events is much smaller for ^{239}Pu than for PWR core reactions and fission of ^{235}U. Consequently, there is a far "smaller margin (≈ 0.006 neutron per chain reaction)" in which to adjust the reactivity factor in a shorter period, with therefore a possible exponential rise in the chain reaction? This is one of the reasons for the care with which *the safety equipment*, mechanisms and devices for breeder reactors are implemented. Experience in France shows that safety aspects of fast neutron reactors have been satisfactorily mastered at various levels of power and physical dimension (RAPSODIE, PHENIX, and SUPERPHENIX).

– *Unit of length*: a useful unit for length is that of the average path of a neutron born in a fission event that experiences elastic and/or inelastic collisions in flight before being absorbed by some other nucleus, fissile, fertile or other. These phenomena being random by essence, they can only be defined in average value, viz. ≈ 20 cm in a breeder reactor (compared with ≈ 2 cm a PWR). As a result, there is a coherence in fission events for a volume associated with this order of magnitude. This helps the process for control-command of the reactor as a whole. Action via a limited number of neutron absorbing devices[65] has an impact on most of the breeder reactor core volume. The relatively long path (≈ 20 cm) of a given neutron leads to high loss (i.e. escaping neutrons from the core or a possible blanket) rates during the fission of the core, hence the importance of having an efficient neutron reflector. In one sentence, a 'smoothing' of the heterogeneous events that change the number of neutrons, per unit volume, is necessary. So, one has to be very cautious before introducing in the core or in the blanket special material that one wishes to "burn" there.

– *Unit of detriment*: residual power of a fuel sub-assembly in order to be able to clean it before entering spent fuel pool (approximately 8 kW) or to bury it in a waste disposal (less of 1 kW).

6. Material properties, mechanical structures and characteristics. Degrees of freedom

We saw that a relevant indicator for changes by irradiation in the mechanical characteristics of the structural materials for the entire duration of their operational life in the reactor system, and the reactor vessel, is the number of displacements

[62] For the fission of ^{239}Pu by fast neutron, the delayed neutrons may be divided in 6 groups of energies, their lifetimes and relative abundance being respectively: 1: 54 seconds, 0.013; 2: 23 seconds, 0.03; 3: 5.6 seconds, 0.124; 4: 2.13 seconds, 0.325; 5: 0.62 seconds, 1.12; 6: 0.26 seconds, 2.7.

[63] For the PWR reactors operated today in France, the corresponding number – taking into account the fact that a little more than 2/3 (56% from ^{235}U and 7% from ^{238}U) and of the fission events come from the uranium and that around 1/3 (36%) *come from the* ^{239}Pu (*and* ^{241}Pu, 4%), *created in situ* – is 0.65%. It is one of the advantages of the ^{235}U chain reaction.

[64] An analogue result for the delayed neutrons is true for the PWR, but so necessary for the control command of the reactor dynamics.

[65] Among the *absorbent nuclei* used, we can cite the element boron, in its compound boron carbide. Natural boron, symbol B, contains 80.1% ^{11}B and 19.9% ^{10}B. The effect of isotope 10 of boron come from the two nuclear reaction: ^{10}B + neutron $\rightarrow$ ^{4}helium + ^{7}lithium + 2.6 MeV and: ^{10}B + neutron $\rightarrow$ 2 ^{4}helium + tritium (^{3}H). This last reaction needs a neutron of kinetic energy $\geqslant 1.2$ MeV.

damage in the alloy-created by the neutron flux (one displacement per atom in the solid = 1 dpa) per atom.[66] For PWRs, this number lies between 10 and 30 "dpa",[67] depending on the initial position of the atom in the material in the structural system. For breeder reactors, the equivalent number lies between 100 and 200 dpa, or even high values.[68] Consequently, the structural engineers have to develop suitable materials. To increase the efficiency of the turbines, the temperatures will rise from 300 °C to 550 °C (and more if we increase the limits of temperature of the concerned materials. One of the most important parts concerned here is the cylindrical sleeve that contains the fissile or fertile MOX[69] cylindrical fuel "pellets".[70] For this purpose, studies and tests are made using ferrite martensitic stainless steels (with between 9% to 12% chrome) and so-called Oxide Dispersion Strength steels[71] [26] – which have also ferrite martensitic materials as matrix.

The materials used for the sodium circuit pipes are also ferrite martensitic stainless steels, 316 stainless steel grids inside the reactor and for the external sodium circuits (secondary circuits), using ferrite and austenitic steels. It seems that, although there is some studies of plasticity of irradiated materials, and some of plasticity of materials under irradiation, the exact relation with fracture behavior is still basically not understood. This is the real engineering problem at least for long live structural component. For short lived components such as fuel cladding, dimensional stability (irradiation creep, irradiation growth) together with induced brittleness or creep fracture are of relevance. Again, as material science issues are concerned, one has to make clear the difference between irradiation damage and thermal aging: irradiation damage is fast less understood, difficult to investigate experimentally and likely to be quite different for the high dpa situation of fast breeders. Thermal aging will be some how similar for fast breeder and classical PWR (although likely to be faster).

The whole system of reactor, coolant circuits, etc. requires that the plastic properties for irradiated materials under these conditions, that are not fully understood yet, should lead to studies into the basic properties of materials used, such as hardening[72] (primordial to comprehend how stainless steel internal structures react and behave) likewise for the reactor vessel for creeping and metal fatigue under radiation conditions.

7. Fuels cycles and material cycles. Separating fissile, fertile matter and wastes. Degrees of freedom. Raw materials used as fuel and those considered fertile

In order to fuel possible future breeder reactors in France, there must first be a mechanical and chemical separation of the plutonium contained in irradiated spent MOX[73] elements, during their cooling process, on the storage sites of the reactor operator, who is the owner of the elements or plutonium extracted from spent UOX fuels that had not been transformed into MOX fuel. The mass of plutonium in these stocks can be reckoned at several hundred tons. For fertile fuels, we would have a choice between using hundreds of thousand tons of depleted uranium (containing approx. 99.7% ^{235}U) as separated at the Tricastin isotopic separation factory (in the south of France) and the tens of thousands of tons of spent UOX, from

[66] Cf. [18], Chapter 2: The displacement of atoms, pp. 73–124.

[67] The definition for "*dpa*": "We consider the case of crystals. We suppose that a projectile strikes an atom in the crystal lattice and ejects this atom, creating a vacancy. The atom that was ejected can move to another position in the lattice and stop; in this case we talk of an interstitial atom. The vacancy and the interstitial atom play an important role in regard to the effects of radiation on the solid. We use the expression a Frenkel pair [defect or disorder discovered by the Soviet physicist Yakov Frenkel, 1926]. The pair creates mechanical stress and stores energy in the solid. The so-called radiation damage stops increasing when the atom stops moving and repositions itself in an interstitial status. The phenomenon lasts approx. 10^{-11} seconds. Many other physical and mechanical effects follow. These are called "the physical effects of radiation": crustal expansion, phase change, segregation, etc. When the crystal is subjected to mechanical stresses during or after irradiation, the effects of these Frenkel pairs can be quite considerable: creation of vacancies, dislocations of the microstructure, fracture, metal becoming fragile, creeping, fatigue cracks, etc." Cf. pp. X and XI in [18]. *The initial creation of a Frenkel pair is known as a dpa* (*displacement per atom*).

[68] [24], pp. 10–19, as well as the articles in the same issue of "MRS Bulletin" authored by Jean-Pierre Bonnal, Hélène Burlet, Céline Cabet, Nathalie Chauvin and Etienne Vernaz [CEA]. We can quote Stéphane Gin [CEA] on the question of glasses [25], the numerous publications that followed and his contribution to the seminar, Dec. 15–16, 2010 at the Academy des sciences, Institute de France.

[69] As mentioned in the text, the oxide of actinides are shaped in small cylinder, of a diameter of ≈ 1 centimeter, height of some millimeters. The hottest point of an oxide pellet, i.e., in its center can reach 90% of its melting temperature. The operational margins are very slim and we readily see how important it is to correctly control the distribution of the neutron flow in the breeder reactor core. To avoid this hot point in the center of the pellet, they may be with a small cylindrical hole in their center. But we mentioned earlier that there are other fuels that are studied in Europe and India, i.e. thorium.

[70] The small thermal conductivity of oxide induce a temperature of 2000–2400 °C in the center part of a fuel element pellet, for a surface temperature of ≈ 550 °C. The melting temperature of the oxide is ≈ 2700 °C. So the pellet are sometimes, annular.

[71] "The ODS are special materials used to increase the mechanical performances at high temperatures. The metallic properties are characterized by the behavior of the *dislocations* that exist in the *crystals*. When a material undergoes all sorts of mechanical, physical, thermal, chemical, etc., disturbances, the dislocations themselves can move, and this determines the *plastic* properties of these ODS materials. Engineers have imagined *dispersing* "nanometric" impurities in the *metal* or *metal alloy* crystals, in order to block the dislocations, etc., by introducing structural faults. Metallurgists and black-smiths, empirically, have been doing this for centuries. In the case of steels, the materials introduced are transition metal oxide powders (yttrium: Y_2O_3 in the proportion 0.2 to 2% and chromium, for example) dispersed in a chrome-steel lattice. The operational temperatures of the ODS family can attain a temperature in excess of 1150 °C." These materials are used industrially, for example, for turbine blades in aircraft engines. Fabrication is usually made by sintering. These materials however are not without drawbacks: their creep behavior changes very abruptly from a low creep rate to an accelerated tertiary creep, and the processing and joining of these materials are far from obvious.

[72] An example of basic research here is in [22].

[73] MOX is a mixing of uranium oxide (uranium which can be natural or depleted as a waste at the enrichment factory or extracted from fuel elements reprocessed at the reprocessing factory like La Hague) and oxide of plutonium (plutonium separated from the irradiated enriched uranium irradiated in one of the nuclear plants of UOX fuel) which is mixed and used to make new fuel elements for the water reactor. This process has been used in Belgium, Germany and France. It is now in project in some others countries, like USA (to destroy the military plutonium). The complete MOX fuel element is in France at a well industrial level.

the Hague reprocessing facility, with a ^{238}U content slightly in excess of 98% ^{238}U, approx. 1% ^{235}U and even less ^{236}U (the half life of which is 2.3×10^7 yrs). France therefore can disposes of sufficient raw material fuel sources to supply the first fuel charges of possible future breeder reactors in France or elsewhere. Over and above these choices, France's high level echelons (Administration and State) would also have to decide whether other transuranic elements (extracted[74] from the spent MOX fuels or the Irradiated UOX that had not been reprocessed) could be used as fuels or targets to destroy.

Another degree of freedom comprises the precautions (including time scales involved in burying nuclear wastes in a repository, with due preparatory thought given to the specifications for doing so-the so-called "terms of reference", taken in respect to safety considerations for all forms of radioactive materials in a context of possible malevolence, while awaiting for the High Public Authorities concerned by these topics make their decisions as to the disposal procedures to be implemented.

8. Mechanical, chemical engineering and physicochemical considerations, related to these fuel cycle processes

- *Design and assembly of fuel elements*,[75] fertile elements, mixed elements (with both fertile and fissile properties), material to be destroyed, control-command on radioactive elements already manipulated and placed in storage facilities, such as the ^{239}Pu (available in large quantity) in the MOX facility etc.
- *"Refabricate" fertile, fuel elements and control elements.*

Degrees of freedom: however, the highest density of ^{240}Pu (cf. above) and in ^{242}Pu, will increase the neutron flow emitted by spontaneous (i.e., natural) fission.[76] The radioprotection from neutrons[77] places very stringent constraints on the fabrication processes, even impossible to meet given the industrial quantities needed for certain chemical elements containing special isotopes. Their relative density depends on the *duration of the radiation received by the MOX fuel elements and their length-of-time of cooling*. This constitutes a new degree of freedom for the fabrication methods and the radioprotection of the personnel handling such components.

8.1. Results, targeted and envisioned. Our experience. Comparison with PWRs

Finally, in the case of a breeder reactor, as indeed for any other nuclear power (with the temperature of the coolant leaving the reactor vessel, so with a common yield of the turbine, between 34 and 40%) generating reactor,[78] if we take as an example a power output of 1 GW electric, the amount of fissile matter needed is $\approx .1$ ton per year, or a little less if the fuel is plutonium (including notably fast neutron fission of the ^{238}U and other actinides formed in situ) and if we can operate the reactor with a higher temperature (it is again a problem of material).

- Let us now summarize the case for fuel and fertile matter: France has enough material on his territory to provide for a first load operation of both ^{285} uranium and plutonium for a number of nuclear generation facilities that would cover the current total energy requirements of the country for many centuries to come, even millenniums.
- A second method to calculate the fissile and fertile element mass flow is as follows: let us take as our reference, the electrical energy output rather than the reactor fuel consumption and time scale. Here consumption reads:
  Plutonium mass[79] (existing today because it has been produced by the PWR of the electronuclear facilities), consumed per TW. Hour. electric: ≈ 120 kilograms Pu/TWhe;

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[74] We have to be careful to state where are the difficulties of this assumed destroying process. There is a creation in situ of practically all these transuranics also in the breeder, but in very smaller quantity. A lot of them will be destroyed in situ. But for the assumed external transuranics coming from past irradiations in PWR, we have to add the problems of separation of each chemical element, of fabrications of targets, of a different length-of-time (more than 15 years, and this will have to be endured by the structure material of these targets) of irradiation in the breeder, because the isotopic concentrations in the chemical element will be different from the transuranic created in situ, etc., and which is more than all that, the safety of the breeder with these perturbations.

[75] [9].

[76] The neutrons of *spontaneous fission* of isotopes like ^{240}Pu (half life for spontaneous fission $= 10^{11}$ years $\rightarrow$ 950 neutrons emitted/gram second) are not a problem *inside the reactor* or in the pool for cooling, or for fabrication of fuel elements or targets. We can say that the emission of neutrons by all the other transuranic nuclides are also not a problem *inside the reactor*. But, these emission of neutrons are a important constraint if we want *to handle them* for fabrication of targets, for transportation, for *manipulation* and even to storage and final disposal (i.e. the example of the isotope 244 of curium: Half life for spontaneous fission $= 1.3 \times 10^7$ years $\rightarrow 1.2 \times 10^7$ n/gram second. But half life $\alpha = 18$ years).

[77] [27].

[78] We have already seen in the case of PWRs that a little more than 2/3 of the consumption come from burning the ^{235}U and a little less than 1/3 from the fission of the ^{239}Pu created *in situ* by capture of neutrons by the ^{238}U. Interested readers will find the relevant figures for so-called neutron economics in [12].

[79] The plutonium that comes from the spent UOX [Uranium OXide] irradiated fuels from thermal neutron reactors contains is at most 53% of the isotope ^{239}Pu. In the case of MOX [Mixed OXide of uranium and plutonium] irradiated fuel elements, the ^{239}Pu isotope mass density is less than 40% of the whole chemical element plutonium. As we make progress in radiation levels and length-of-time of irradiation (in one word, "burn up" in [Megadays per ton]) in PWR, the plutonium 239 isotope density decreases. Moreover, the higher the burn up of fuel elements in the PWR, the higher is the proportion of energy of the PWR coming from plutonium fission (this plutonium being created in situ by capture of neutrons by the ^{238}U). But the smaller, is the proportion of isotope 239 of plutonium among all the other isotopes of plutonium at the end of the irradiation in the PWR. On the contrary, the plutonium produced in the breeder is composed mainly of isotope 239, because the capture cross sections are so small in the 3 MeV–1 MeV domain of kinetic energy of the neutrons.

Plutonium Mass produced per the possible future breeder per TWhe: approx. (or slightly in excess of) 120 kilogram/TWhe; let us refer to the case of combustion of plutonium from MOX Fuel elements in the reactors of France. In order to produce 1 TWhe in a PWR of the PWR French reactor, we need to consume ≈ 226 kg of plutonium coming from the reprocessing of spent UOX irradiated fuel elements. When spent irradiated MOX FEs are removed from this PWR there still remains some ≈ 170 kg/TWhe of plutonium and also, some transuranic nuclei). However, we have to stress that what varies at the same time of unloading the irradiated fuel from the reactor is:

(1) the isotope content of these two categories of plutonium – the input in the reactor and the output from the reactor[80] and
(2) the isotope content of the transuranic elements (^{241}americium, ^{242}americium, ^{237}neptunium, ^{244}curium, ^{245}curium, etc.) that were formed in the irradiated MOX from transmutation of the plutonium isotopes and the other transuranic created in situ the breeder.

- The non-fissile and non-fertile transuranic elements: all transuranic elements are fissile[81] with relatively high kinetic energy neutrons, approx. ≈ 1 MeV. We refer to these transuranic nuclei as having a fission threshold. The threshold differs from one isotope to another, over a range from 0.3 to 1 MeV. We saw that the neutrons from the fission events have a kinetic energy spectrum centered on the number ≈ 2 MeV. Depending on the physical composition of the 20 cm that surround these elements, the kinetic spectrum is more or less high. The "hardest" spectrum, viz. with highest kinetic energy is found in the case of sodium cooled breeder reactors such as EBR I and EBR II.[82]

For this reason, certain Authorities and scientists are exploring the possibility of destroying some of the transuranic elements by fission, or their descendants by disintegration of the short life elements produced, or by neutron capture.[83]

In this light, further studies are needed to analyze the output of the fuel cycle of the breeder reactor (fissile fuels elements and fertile fuel elements), the associate mechanical, chemical and physical-separation[84] processes, and those used to "re-engineered" new fuel loads for the breeder reactor core. Currently, we do not have enough data to determine what to do exactly with those transuranic elements present in the fuel and in the fertile Elements at the end of an operational cycle.[85,86]

Another utilization has been envisaged for high kinetic energy neutrons: the separation process for certain transuranic materials coming from the future irradiated (spent) fuel elements used in the future PWRs, destroying them by the high flux of fast neutrons of the breeders radiation in special targets that would be placed in the breeder reactor for this purpose. This idea presupposes that we wait till the end of the initial cooling length-of-time after extraction from the PWR reactor core, a decrease in the ambient radioactivity such that manipulation of the fuel elements becomes possible, then proceed with their separation either by chemical or by "physical chemistry" means and in all cases, mechanical processes, fabricating new targets with some parts of these same elements. Then, we would have to decide on the length-of-time (≈ 14–15 years) which is more than necessary under the high flux of fast neutrons of the breeder reactor radiation so that a significant (but never entire), fraction is indeed *destroyed* (= *changed into fission fragments*). It could be necessary to move them and re-move them again to adapted position in the reactor core where the spectrum is optimal. We will have to minimize the perturbation to reactor physics whenever we have to change a subassembly (fissile or fertile) or a target to a new position. The change of position of a target or a fuel element or a fertile element is indeed a complicated operation

In short, it will be a situation where, for each phase, operational, maintenance, incident/accident, etc., we shall have to weight up the pros and cons. The experiments run in PHENIX and other flux of fast neutron, and those that will be conducted in any future nuclear facility[87] in France should allow us to assess the scientific, engineering, industrial and safety-related feasibility of attaining these aims (destroying some transuranic or other radioactive materials?). On the basis of these results, adding the interest of these processes both for the reactor and for the complete fuel cycle (including the final disposal sites chosen), safety, radioprotection of personnel of the power generation operating company,[88] the

[80] One can find the detail numbers in [13], pp. 109–110 and [19].

[81] Detailed properties and qualities for the transuranic elements are set out [19].

[82] Reminder: EBR = "Experimental Breeder Reactor".

[83] For those transuranic elements formed in the breeder reactor cores, a significant fraction will be destroyed in situ by fission, neutron capture, disintegration, etc.

[84] We stress that, today, the scientific feasibility of chemical actinides in the irradiated fuel, fertile and target is demonstrated to the scale of *the laboratory*. Moreover, there exist several options and so the possibility to choose following the terms of references of the future industrial fuel cycle. This industrial process of the complete fuel cycle of breeder, is, in the complete world, an open problem, even at the pilot plant level. But certain steps of this process have reach the industrial level.

[85] We can say the same statement for the transuranic material from the fuel cycle of the PWR, which are to day cooling in water pools and are accumulated each year [13,19]. We stress that the transuranics which will also accumulate in the fuel and the fertile elements of the breeder reactor and will be unloaded with these breeders fuel and fertile elements are of the same physical and chemical kind as the transuranics accumulated, for as an example, by the irradiated MOX fuel elements. But the difference is not in the physical or chemical nature, but in the processes to manipulate these highly radioactive materials and to use them to fabricate targets to place in the breeder. Once more, the big problem, now, is not only inside the reactor, but the cycle outside the reactor concerning the fuel and fertile elements and eventually the targets to destroy. This outer cycle is so important that we will go deeper in the next paragraph.

[86] As well of inside as we mention before.

[87] For example, there is a test reactor whose construction is ending and which be operated soon: the "Jules Horowitz test reactor" in Cadarache/CEA.

[88] The company producing the electricity.

costs (in safety, radio Protection, manipulation, complication, maintenance) we can draw up complete and specific technical specifications for future breeder reactors and their fuel and fertile processes, for these particular functions, and for each of the transuranic elements considered one by one (neptunium, americium, curium, etc.). This constitutes an important topic that will have be included in the general specifications for future nuclear power generation and their complete fuel cycle facilities. One remark coming from 50 years of experience on nuclear reactor and their fuel cycles in all the industrialized countries. Until now, one of the reasons of their quick technical and industrial success had been. the simplicity of realizations of these power reactor operation and of their fuel cycle. Are we not leaving this simplicity? So, the main advice to the future generation of engineers who will design these breeders: if you want to have a robust installation and fuel cycle process, keep it simple.

9. Possible orientations taking into account past experience. Reactor operation and maintenance

Experience in breeder operation, worldwide, has taught us that overall, it has been possible to obtain a perfect control of operational phases, maintenance and element handling. But we also learn that the minor incidents or accidents on record were due:

(a) to manipulation of various components in the sodium circuits. Such possible occurrences were satisfactorily solved in both PHENIX and SUPERPHENIX[89];
(b) to lack of definition of the objective of the breeder facility realization.

A prototype of power electronuclear plant is not the same object, operation, safety, radioprotection, fuel cycle, than a reactor to learn about all the function of cooling, loading, unloading, etc. or a material test reactor or a reactor to destroy certain waste elements.

So the *objective* of a nuclear power generation facility must be very carefully defined and documented by his "*terms of references*". The technical literature must be carefully written and edited and made readily accessible to all staff, the missions of which, are the design of handling of reactor or control components and equipment and fuel element cycle. The result is a *reactor operation specification* that details and lists all the functions expected from a given facility and also the means available to enable staff to follow up (either through annual-at least-technical reviews, the contents of which are transformed into sets of operational orders, procedures and system status acknowledgments).

10. Containing, preventing and reducing nuclear weapons proliferation

An idea that prevails, but which is inaccurate, is: with some small masses of the metal plutonium, it is easy to assemble a nuclear weapon. However, nuclear weapons with just plutonium require numerous test explosions to be fully operational. The failures noted in this area are those from nations who have produced plutonium with their uranium fuelled reactors and have generally only succeeded in getting a quasi chemical poor energy yield. They will not admit it, but United Nations watch dog observations prove it. The only criticism leveled at breeder reactors had been that they do produce plutonium that can be adjusted to have weapon's grade characteristics. However, the reader will have noted that when there is a breeder reactor, then the fuel cycle doesn't need an uranium isotope separation facility. The only way, at present, to produce a nuclear weapon is to start with ^{235}uranium. The Los Alamos laboratory proved this in 1945 by releasing and exploding a nuclear weapon over Hiroshima, without engaging in any preliminary test, but being obliged, for the plutonium device to make the so-called "Trinity" nuclear test in a desert place near Los Alamos and from this a weapon as big as a car [6], which could not fit in a missile head or in a normal plane.

Moreover, all thermal reactors needed nuclear fuel using uranium in enriched ^{235}uranium. International treaties on United Nations as they stand today, confers the right on all nations which had signed these treaties to buy natural uranium.[90] They can also build and operate an uranium isotope separation facility to enrich the natural uranium in ^{235}uranium up to the 3.5–4.5% grade as required for nominal reactor operation. But we stress that for somebody who want to produce weapon's grade uranium,[91] the hardest path of the process is to reach $\approx$ several% of ^{235}U. Half of the thermodynamic work has been accomplished at this stage in the enrichment process. The mass flow of uranium into the separation process, when one begins the enrichment at 4 or 5%, is divided by between a factor between 7 and 10 (compared to the case of flow with natural uranium). So, it is a small building compared with those of natural uranium enrichment, easy to hide. With only a few circuit rearrangements in the separation process facility, and without changing the facility as a whole, the uranium output at weapon's grade can be achieved.

[89] The technical incidents that marked the beginning of operation stages of SUPERPHENIX breeder reactor were all solved and repaired technically. But the "cost" was a series of stoppages and outages, aggravated by the delays to obtain the administrative permission to restart the nuclear chain reaction in the reactor. No serious technical incident was recorded for the PHENIX reactor.

[90] (And also reactor grade enriched uranium) and as much to fabricate their uranium fuel domestically, provided that they accept the conditions of the treaties, and among them, the inspection of their facilities by the UN (United Nations) "watch dog", the Atomic Energy Internal Agency [AEIA].

[91] This uranium, in an enrichment factory is a gas, usually, a fluoride of uranium.

A gradual changeover to breeder reactors would on the contrary reduce *the danger of production of weapons grade enriched uranium*.

11. Radioactive wastes from a breeder reactor cycle

The nature and the quantity of radioactive wastes of the breeder fuel cycle and operation of the plant have to be compared[92] with those of other electronuclear production methods with thermal neutrons: to the extent that the uranium irradiated in the PWRs or analogue facilities, separated out in the nuclear waste reprocessing units, and also the depleted uranium from enriched uranium are re-used in the fertile elements and in the fuel elements for the breeders reactors constitutes a first advantage.

The plutonium necessary for the fuel elements of the breeders exist today in all the irradiated MOX elements cooling in the pools of the French operators there is already enough plutonium for fuelling a complete nuclear park during ten complete loading fuels, and so to wait to draw out from the fertile elements, the plutonium necessary for the future fuel elements.

The ratios between the cross-section for neutron capture and the fission cross section being far smaller, than in thermal neutrons of the PWR and all the present types of electronuclear plants, there are relatively less transuranic elements created in a breeder reactor core.

Given that the spectrum of neutrons is very close to the average fission thresholds, many of the transuranic fissions can take place during the irradiation of the fissile and fertile FEs.

The final comment here is that the mandatory waste mass come from the fission events, which in the case of the PWRs or breeder, have approximately ($\leqslant 2\%$ of the neutrons) the same order of magnitude,[93] with slight variations in the compound compositions. In the electronuclear stations of the main electricity producer in France, this quantity is approx. 55–60 *tons of fission products per year, i.e. approx.* 0.1 *ton of fission products per year per terawatt.hour* (1 *terawatt* $= 10^{12}$ *Watt*) *or* 1 *gram/year of fission products for each French national.*[94]

To be complete, we must add to these wastes in the structural materials that are also irradiated. However, the capture cross-sections for these materials to the impacting neutron kinetic energies which lies between 0.1 MeV and 1 MeV and are far smaller than in the case of the kinetic energy in the domain of the thermal agitation energy of the matter around its equilibrium points.

12. Conclusion

What remains to be done and decided before we can have a demonstration of the feasibility of an industrial electronuclear breeder reactor?

Since the mid 1950's, our knowledge base about breeder reactors has progressed sufficiently to ensure that such a reactor, with its specific fuel cycle,[95] is a real technical and engineering feasibility in France. The stumbling block for a long period was the high cost of building a breeder reactor, leading consequently to a high cost per kWh electricity price tag. Recently the cost of energy (all sources) has risen to a point that this argument against the electronuclear breeder solution is countered. Several installations are already up and running in Russia, Japan and in India (using a fuel derived from natural thorium). If France wishes to participate efficiently in this international effort, then we must rapidly and without delay define an action plan, presenting the decision-choices made, placing them in a time schedule for the major milestones to come.

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[92] How to compare detriment from wastes? (1) Is this *radioactivity variation* factor a good indicator of the damage potential detriment attached to such fission products? We mention this only as a reference. What then are the other possible risks? They all relate to a fictitious scenario for which *we* carry out *a risk assessment* and a *management risk assessment.* (2) For the case of deep geological repositories, *the migration duration* through the various rock layers, through what we call the *geological barrier*. This leads to giving a preference to providing protection against certain fission products i.e. cesium; (3) For *the size of these underground repositories*, the *volume* of ultimate wastes is the criterion. In this case we privilege high combustion rates. (4) For *precious materials* (*i.e. platinoids*"), there is temptation to seek them out; (5) For scenarios in which we imagine wastes migrating via water passages, taken up in water plants, food chains, according to a given calendar and for a chosen population ("a cohort" in the language of radioprotection), the test bench mark is the radioactive products *ingested and inhalated by the cohort*. This indicator reveals the *radio toxicity* of the wastes and is measured in "Sievert"/person (definition in [28]: The average permit standard for population is 0.1 μS for the non-official International Commission of Radioprotection and 0.3 μS for the International Health Organization of UN) in the cohort. This scenario is broken down into two sub-scenarios: (5a) we count just what migrates and we find the material mentioned above, (5b) we suppose that the cohort has access directly to the underground waste, the indicator here being the same for all actinides, since they emit alpha rays which once ingested have a very serious effect on health. To be precise, if a person ingests (or inhales) an actinide, the damage to vital organs is approx. 10 000 times more serious than for other radioactive wastes.

[93] A important fact between the fissions of ^{235}U and ^{239}Pu, concerning the chain reaction, and so, all the economy of neutron of these two cases, is the average number (quoted ν) of neutrons per fission: For ^{235}U, $\nu = 2.43 \pm 0.2$ at thermal energy of the neutron and for ^{239}Pu, $\nu = 3.1 \pm 0.15$ at 2 MeV. [Argonne National Laboratory; Reactor physics constants.]

[94] The composition of the fission products, for a uranium fuel enriched with the isotope ^{235}U in level of radioactivity, i.e., for a radiation level of 30–40 GW day/ton of heavy metal (uranium and plutonium), from a 3.5% enriched ^{235}U base, for use in a PWR – is set out in Fig. 57 [13], p. 244. We note that in this figure *the radioactivity of the fission products returns to the 'natural' level of extracted ore of uranium that produces the fission products in about* 2 *centuries.* In the case of fast neutrons ^{239}plutonium fission events, one must increase the relative fraction of the fission mass centered on the atomic mass of ≈ 120 (for example, palladium and the other so-called "*platinoids*", and by a factor in excess of 10).

[95] [30].

Materials and mechanics, chemistry and physical chemistry: all forms of activity that relate to building and operating a series of electronuclear breeder plants will call for equipment, reactors and pilot factories to test and assemble fuel elements and deal satisfactorily with the question of final disposal of nuclear wastes. This implies an important industrial sector dedicated to these activities and an in-depth understanding of the physical chemistry of materials involved, both at atomic level, molecular bonds, crystal phases with surface and dislocations, their level of chemical reactivity and possible toxicity.

The structural materials [31] and the functional materials – the latter including in particular the actinides and their compounds, are under continuous investigation [29], pp. 841–875. The important role of fuel chemistry [32] and physical chemistry was stressed at the December 15–16, 2010 meeting organized by the French Academy of Sciences.

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