Transmutation: the state of the art after the Fourth Framework Programme

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Abstract

In the present paper the state-of-the-art of the transmutation technology is sketched based on the results of projects performed in the specific programme on Nuclear Fission Safety of the Fourth Framework Programme (1994-1998) of the EU.

1. Introduction

Partitioning and transmutation (P&T) of long-lived radionuclides is a possible means for the management of radioactive waste [1]. Its goal is the improvement of the long-term safety of disposal of highly radioactive waste by minimising the amount of radiotoxic and hazardous elements that need to be disposed of. This can be achieved by partitioning (separating) specific nuclides from the spent fuel, and transmuting (converting) these separated nuclides into short-lived or stable ones by means of nuclear reactions. It is thus evident that P&T is an extension of the conventional fuel cycle with reprocessing, as is applied in several European countries.

In the recent years, research programmes to investigate the possibilities and benefits of P&T have been initiated in many European countries. The French SPIN programme [1] is certainly the most prominent and most ambitious of these and it has become the origin for many collaborative studies in Europe. The importance of P&T has also been recognised by the Commission of the European Communities (CEC) and the CEC has sponsored a number of projects on P&T in the specific programme on Nuclear Fission Safety of the Fourth Framework Programme (1994-1998). In these projects, which include theoretical as well as experimental studies, various aspects of the

transmutation of actinides and long-lived fission products, and the incineration of plutonium have been investigated, both for critical reactor systems as for accelerator-driven systems [2].

In the present paper the state of the art of the transmutation technology is described taking into account the results of the projects performed in the Fourth Framework Programme, which are summarised in Table 1. The state-of-the-art of the partitioning technology is subject of the paper presented by Madic at this conference [3]. It is beyond the scope of the present paper to discuss the results of the studies in detail, also because they do not address one specific P&T strategy but explore different options or address fundamental aspects. In the next sections, the major achievements will be described in the context of the global developments. The reader is referred to the final reports of the projects for details. However, for the sake of clearness, we will make a division in three areas: (i) P&T strategies and technologies based on the conventional and fast reactor systems, (ii) P&T strategies and technologies based on accelerator-driven systems, and (iii) the thorium cycle.

Table 1: Overview of the shared-cost actions and concerted actions in the field of transmutation, as performed in the specific programme on Nuclear Fission Safety of Fourth Framework Programme (1994-1998).

Title	Objective
Evaluation of possible P&T strategies and of associated means to perform them.	Give indications on what can be expected from P&T strategies by evaluation advantages and drawbacks and assessing technical feasibility of the required operations.
Supporting nuclear data for advanced MOX fuels	Update nuclear data libraries that are relevant to advanced MOX fuels and to assess the accuracy of the strategy studies
The joint EFTTRA experiment on americium transmutation	Analysis of the transmutation americium in a $MgAl_2O_4$ matrix by means of an irradiation experiment in the HFR-Petten
The impact of accelerator-based technologies on nuclear fission safety	Assessment of the possibilities of accelerator-driven hybrid reactor systems from the point of view of safe energy production, minimum waste production and transmutation capabilities
Neutron-driven nuclear transmutation by adiabatic resonance crossing (TARC).	Develop theoretically and experimentally the method of adiabatic resonance crossing which enhances strongly the capture rate of neutrons of radionuclides to be transmuted
Physical aspects of lead as a neutron producing target for ADS	Evaluation of lead as a spallation target for the production of high-energy neutrons
Thorium cycles as a nuclear waste management option	Assessment of thorium cycles from the point of view of limiting the actinide production and of the prospective of plutonium incineration

2. Background

The most relevant radionuclides to be considered for P&T scenarios are the actinides and some long-lived fission products. The actinides dominate the radiotoxic inventory of the spent fuel after about 250 years of storage, when the majority of the fission products have decayed. Plutonium is the most important actinide element that contributes to the radiotoxic inventory. But because plutonium is a fissile material, its separation and re-use in MOX fuel are already well developed. Nevertheless, the management of plutonium in a P&T scenario is also subject of further studies since multiple recycling of plutonium is far from trivial.

Much attention in P&T research is focussed on the minor actinides americium, neptunium and curium, which presently are included in the high level waste. The radiotoxicity of the long-lived fission products is small compared to that of the actinides. However, some long-lived fission products (^{135}Cs, ^{129}I, and ^{99}Tc) contribute significantly the radiological dose to the population in the long term, as follows from performance assessment studies of concepts for geological storage. Therefore, the objective of most of the P&T programmes is the reduction of the inventory of the radiotoxic actinides as well as of the long-lived fission products.

Implementation of P&T in the management of nuclear waste will not only affect a number of operations in the conventional nuclear fuel cycle but will also extent it with new operations (Figure 1). At this moment, different P&T strategies are being explored, but, anyhow, it is evident that special facilities for the separation of elements and the fabrication of targets and fuels are required, as are new reactor types to incinerate and transmute the separated isotopes. As a result, a multidisciplinary approach is required to answer the question whether P&T are a feasible option in the nuclear waste management.

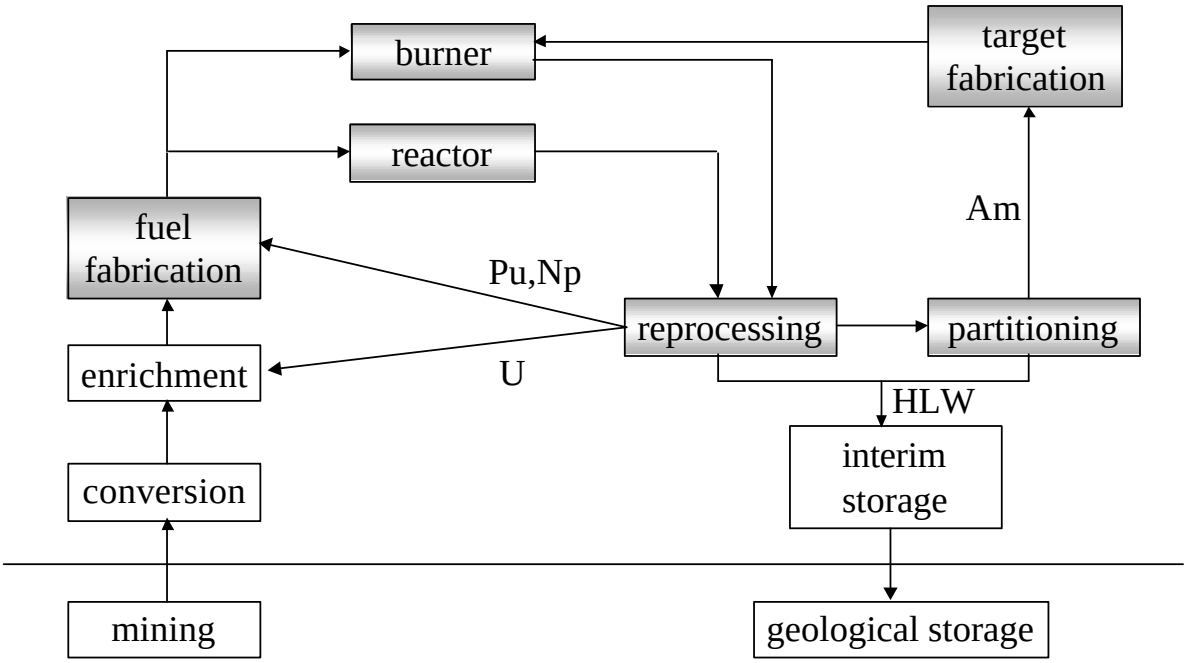

Figure 1: A schematic view of the nuclear fuel cycle with recycling of plutonium and the minor actinides. The shaded boxes represent the stages that are relevant to P&T.

3. P&T strategies and technologies based on conventional and fast reactors

Strategies for P&T always consider an evolution of the present situation (partial recycling of Pu in MOX-fuelled PWRs) to a scenario in which a complete recycling of Pu is achieved, and, ultimately, to a scenario in which Pu, the minor actinides and some specific long-lived fission products are recycled. To achieve the complete recycling of Pu, conventional PWRs are not suited due to their limited capabilities to incinerate plutonium. The quality of the plutonium deteriorates with each recycling step. The incineration capabilities of highly-moderated PWRs are better, but still insufficient to attain an equilibrium situation for the recycling of plutonium. In addition, the high plutonium content in the MOX fuel that is required has a negative impact on the safety of highly-moderated PWRs. When special burners like fast-neutron reactors (FNRs) are used, the recycling of

plutonium and the minor actinides seems feasible from the point of view of reactor physics. Plutonium can be recycled in FNRs in advanced MOX fuels (high Pu content) and separated americium can be irradiated in a once-through scheme in special targets. However, (very) long residence times are needed for the Am targets.

Analysis of different scenarios for plutonium and minor actinide recycling shows that the reduction of the radiotoxic inventory of the cycle waste is a factor 3-5 after 150 years of operation when only plutonium is recycled [3]. When also americium is transmuted this would improve to a factor of 10-20, depending on the strategy (once through or multiple recycling) and the transmutation yield of the targets. To further increase this performance, it is necessary to also recycle curium and enhance the transmutation yield for americium. Also the fraction of FNRs in the reactor park has to be increased [5].

Very little experimental data exist on the feasibility of special americium targets. In a pilot experiment in the Fourth Framework Programme, an americium target for an once-through scheme (Am in a magnesium aluminate spinel) was prepared by a dust-free infiltration technique. The target was irradiated in the HFR in the EFTTRA-T4 experiment [6]. In this experiment the extent of transmutation of americium achieved was 94% and the extent of fission was 28%. The results of the post irradiation examinations revealed a significant swelling of the target (See Figure 1) which is explained by the accumulation of helium, which is produced by in the transmutation scheme by decay of ^{242}Cm, in gas bubbles.

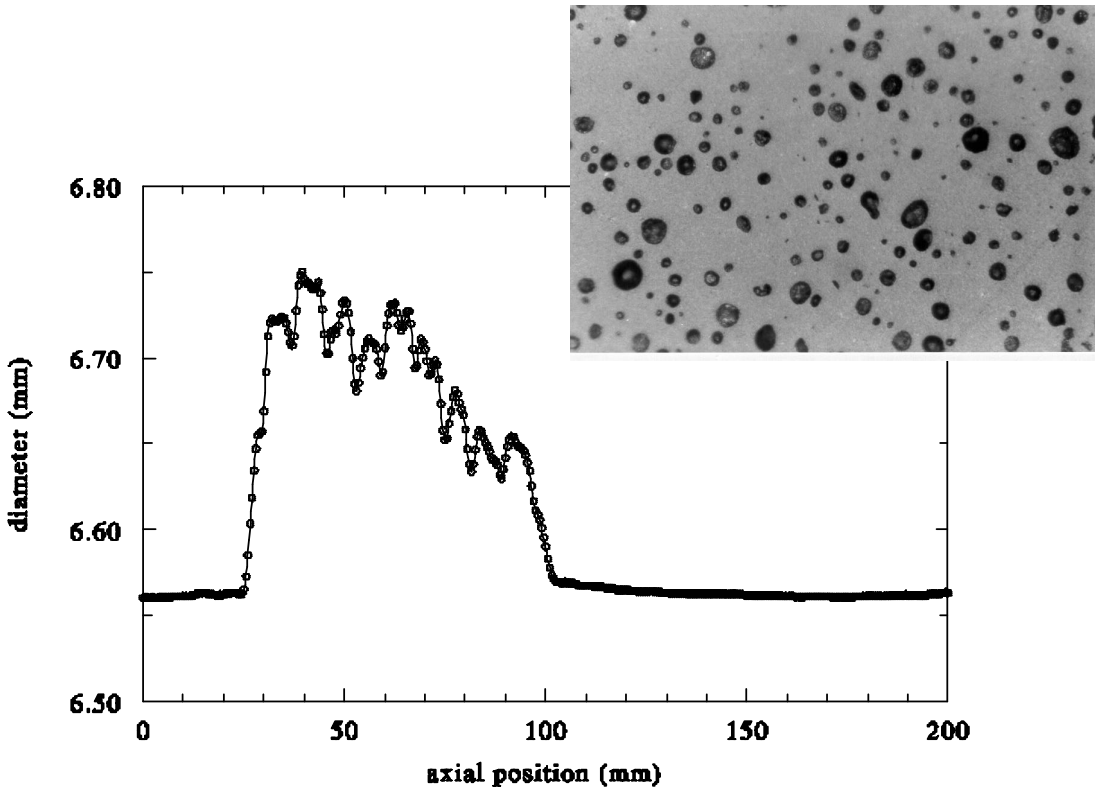

Figure 2. The change in the diameter of the cladding of the EFTTRA-T4 target showing the expansion of the cladding in the area where the target pellets were located. The insert shows a micrograph of the target in which the helium-containing gas bubbles, which caused the swelling of the target pellets, are clearly visible [6].

Neutronic analyses show that multiple recycling reduces the plutonium masses but leads to an increased production of minor actinides. However, the results of these analyses depend very much on the data for the cross-sections of the actinides. These must be checked and assessed very carefully since small errors, when accumulated in subsequent calculation steps, may result in unacceptable uncertainties. Careful analysis of the isotopic composition of the actinides in high burn-up MOX fuels in LWRs [7] indicated that the plutonium isotopes are predicted satisfactory with the exception of ^{238}Pu (which is partly produced by decay of ^{242}Cm). Also for the americium isotopes ^{241}Am and ^{243}Am the agreement is acceptable but the measured concentrations for the isotopes ^{242m}Am, ^{243}Cm and ^{245}Cm agree poorly with the calculations (Table 2). The same exercise has been made for the fast reactor TRAPU experiments in Phénix [7], which give comparable results (poor agreement between the calculated and measured concentrations for ^{237}Np, ^{243}Cm and ^{244}Cm).

Table 2: The calculated to experimental (C/E) ratios for Np, Pu, Am and Cm isotope masses as derived from integral analysis of MOX fuels irradiated in Saint-Laurent (SLB1), Beznau and BR3 using and … using JEF-2 nuclear data file [7].

Isotope	Thermal reactors			Fast reactors		
	SLB1	BEZNAU-1	BR3	TRAPU-1	TRAPU-2	TRAPU-3
^{237}Np	-	-	-	0.78	0.77	0.75
^{238}Pu	0.91-094	0.94-0.97		0.99	1.01	1.01
^{239}Pu	1.01-1.05	1.02-1.06	0.99	1.02	1.00	1.00
^{240}Pu	0.99-1.02	0.98-1.00	1.04	0.99	0.97	0.99
^{241}Pu	0.97-0.99	1.00-1.02	1.05	1.04	1.01	1.03
^{242}Pu	0.94-0.98	1.02-1.03	1.02	1.11	1.05	1.03
^{241}Am	0.98-1.01	1.12-1.15	1.00-1.10	0.98	0.98	0.98
^{242m}Am	0.6-0.8	0.6-0.7		1.03	1.06	1.02
^{243}Am	0.93	1.00-1.01		1.07	1.03	1.06
^{242}Cm				1.02	0.99	0.99
^{243}Cm	0.70-0.84	0.80-0.89		-	0.71	0.70
^{244}Cm	0.92-0.94	0.93-0.95	0.84-0.89	0.98	1.09	1.10
^{245}Cm	0.87-0.94	0.88-0.94				

In the short- to medium term, the implementation of P&T can leads to a small decrease of the collective dose resulting from front-end operations, and to an increase of the collective dose resulting from back-end operations. However, the overall radiological impact is not changed significantly. The benefits of P&T on the long-term collective dose that is the result of geological storage, are most relevant for human intrusion scenarios, in which the radiotoxic actinide inventory is predominating. In normal evolution scenarios, the long-lived fission products like ^{129}I, ^{135}Cs and ^{99}Tc contribute most significantly to the collective dose. However, the transmutation of these fission products in critical reactors is difficult. The transmutation efficiency is relatively low and, as a result, the residence time in the reactor is long.

4. P&T strategies and technologies based on accelerator-driven systems

Instead of critical reactor systems, accelerator-driven systems (ADS) have been suggested for the transmutation of actinides and fission products, operated in symbiosis with conventional reactors. In these systems a proton accelerator, which produces neutrons in a spallation target, is combined with

a sub-critical reactor. In addition, ADS in combination with the thorium cycle, is offering a flexible means of generating nuclear energy with minimum plutonium and minor actinide production (see section 5).

The technology for ADS can be divided in several parts: the accelerator, the spallation target, the reactor and the fuel. For the accelerator, linear and circular devices have been proposed. Circular devices, such as separated orbit cyclotrons, have the advantages with respect to power and space requirements. However, limited experience exists with such devices, especially in continuous operation. For the spallation target, heavy elements (high Z-number) are considered and lead is attracting most attention. The design and optimisation of the target (with respect to neutron production, shielding requirements, activation, radiation heating, and material damage) is being done using high-energy transport calculations, for which accurate cross-section data are required. Efforts to evaluate and compile cross-section libraries for the intermediate and high-energy range for lead are therefore underway [].

The reactor can be a thermal or a fast system, cooled by liquid metal (lead, lead/bismuth) or gas (helium) and fuelled with molten salt, metal or ceramic fuel. Although these different options lead to a wide variety of systems, there is general consensus at present that fast systems cooled with liquid metal offer the best possibilities, preferably as dedicated burner in a so-called double strata option. Scenario studies show that recycling of plutonium and the minor actinides in dedicated ADS will be more efficient compared to the recycling in critical FNRs [5].

Figure 3: TARC figure

One promising aspect of ADS compared to critical systems is the fact that efficient transmutation of fission products seems to be more feasible as significant amounts can be loaded as a result of the potential large subcriticality margin. Also the possibility of applying the adiabatic resonance crossing (ARC) method offers great benefits. ARC is based on the fact that in lead neutrons lose their energy in small decrements as a result of which access to the resonance region can be achieved. In the resonance region, the neutron capture cross sections of many isotopes is generally much larger than for other energies. The feasibility of ARC has been demonstrated at CERN, where experiments on the transmutation of ^{99}Tc were performed [8]. It was also shown by calculations that large amounts of impurities such as ^{99}Tc or ^{129}I can be introduced in the lead volume of an ADS in a parasitic mode, without affecting the operation in a significant way.

5. The thorium cycle

The thorium cycle offers some advantages over the uranium cycle with respect to the long-term hazards, depending on the strategy chosen. Natural thorium is not fissile and the fuel cycle should be started up and/or maintained by addition of a fissile material such as ^{235}U, ^{233}U or ^{239}Pu. The choice between these materials represents different strategies. The use of ^{233}U, which implies that reprocessing of thorium fuels must be done in a Th/U cycle, is an option for the continuation of nuclear energy in the long-term. The use of ^{235}U (highly enriched, HEU, or medium enriched, MEU) in a once-through scheme can be considered as an alternative for the U/Pu once-through scheme, as less actinides are formed. Lastly, Th/Pu fuel can be considered as an option for the incineration of excess plutonium in PWRs, but also as a first step towards the Th/U cycle.

Self-sustaining fuel cycles based on thorium (i.e. enough ^{233}U is produced to fuel to maintain the cycle) can only be obtained in fast reactor systems (critical or accelerator-driven). Reprocessing of the fuel to recover the ^{233}U using the THOREX process or pyrochemical techniques should then be applied with an efficiency similar to the PUREX process (major actinides with an loss fraction of 0.1%, other actinides with 1% or better). In that case, the radiotoxic inventory of the waste to be disposed of will be smaller than in case of the U/Pu cycle. The benefit is the largest (a factor 15-60) during the first 30.000 years of storage. The best results are obtained for thorium-fuelled accelerator-driven systems.

In PWRs, the use of HEU or MEU as fissile phase of the thorium fuel seems technically feasible, although the use of HEU requires measures to avoid a positive moderator temperature coefficient. In principle, the uranium produced in such cycles could be recycled, but only in the case of HEU this makes sense with respect to the radiotoxic inventory of the waste as only small quantities of plutonium and minor actinides are formed. For MEU, which contains at least 80% ^{238}U, this is not the case. MEU-based thorium fuel offers some benefits with respect to the radiotoxic inventory compared to uranium fuel, but much less than HEU. However, the use of HEU is difficult as a result of its proliferation risks.

Thorium-based mixed oxide fuel (Th-MOX) has the potential to incinerate plutonium efficiently. The consumption of plutonium in Th-MOX in a PWR (60 MWd/kg HM) is twice as high as in U-MOX fuel for a full core loading (see Figure 3). In such as case, almost 100% of the fissile plutonium will be destroyed, whereas some ^{233}U will be produced. As a result, Th-MOX is very well suited for once-through incineration of plutonium. The remaining Pu can be recycled with the ^{233}U, but this will have only minor impact of the transuranium content at the end of cycle. A more

logical improvement would be the extension of the burnup of the once-through case to 100 MWd/kgHM.

The radiological impact of direct storage of thorium-based fuel does not differ from that of uranium-based fuel. It is determined by the geo-mobile fission products like ^{129}I and ^{135}Cs in the normal evolution scenarios.

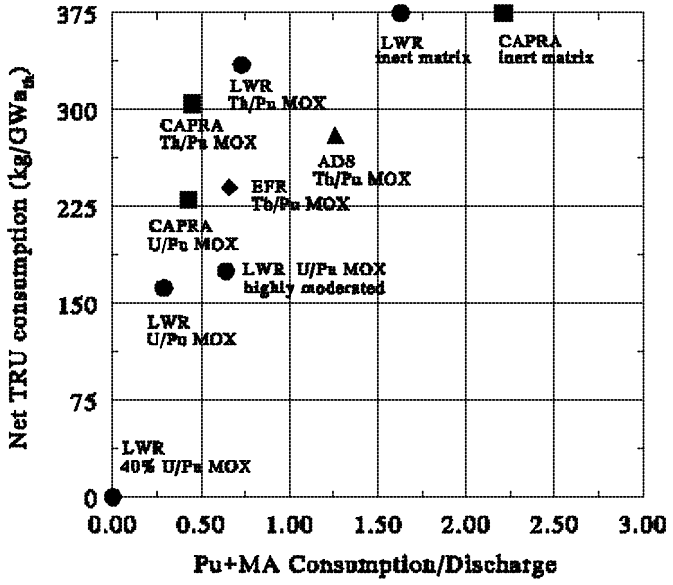

Figure 3: Compilation of net consumption efficiency of transuranium elements in different reactor types [10,11] showing the much better performance of thorium-based and inert matrix fuels compared to MOX. The theoretical maximum TRU consumption is about 375 kg/GWa$_{th}$.

6. Conclusions and outlook

In the previous sections the state-of-the-art of the transmutation technology is sketched, mainly based on the results of the studies performed in the frame of the EU Fourth Framework Programme. It leads us to the following conclusions:

- The recycling of plutonium in mixed-oxide fuel in LWRs is already practice. Improvement of the efficiency can be achieved by use of highly-moderated PWRs or FNRs. In addition, the use of thorium-based mixed-oxide fuel offers the possibility to increase the plutonium consumption rate by a factor of about 2.

- The results of the scenario analysis (calculations) clearly demonstrate that transmutation of neptunium and americium, in combination with plutonium recycling, is a means to reduce the radiotoxic inventory of the generated waste by a factor of 10-20. An equilibrium situation is achieved after 150 years of operation using fast-neutron reactors [4]. However, such P&T scenarios do not affect the long-term radiological impact of geological storage. The radiological

impact is only reduced when other nuclides such as certain fission products (Cs, I, Tc) and curium are transformed into short-lived ones.

- Transmutation of long-lived fission products (Cs, I, Tc) and curium, together with the further improvement of the efficiency of transmutation of americium, neptunium and plutonium, seems to be possible with accelerator-driven systems. The technology of accelerator-driven systems is, however, complex and much less developed, with emphasis on the accelerator, the target and the fuel. ADS therefore only offers a solution in long-term.

- An ADS operated with a thorium fuel cycle, offers the possibility of spent fuel with a very low radiotoxic inventory.

- The technical feasibility of transmutation has only been demonstrated on an experimental scale. The transmutation of americium in inert matrix targets has been tested, but the results of this experiment have shown that further development of the target design and fabrication methods are needed. The experiment supplements the findings of the SUPERFACT experiment on transmutation in a UO_2 matrix [12]. Also the feasibility of technetium transmutation by the ARC has been demonstrated, extending the earlier studies on transmutation of Tc in reactors [13]. Such experiments are the first step toward the technical realisation of fission product transmutation.

- The nuclear fuel cycle has a very long time constant. The development and licensing of new systems or new fuel takes a long time. Changes can only be implemented gradually and they become fully effective only after they will have been pursued for several decades.

Based on these conclusions it is recommended to focus future research on fuel cycle options for transmutation of americium and curium in critical and subcritical systems and on the development of the technology for accelerator-driven systems. By nature, this research must be multidisciplinary encompassing fundamental research (nuclear data, materials science), applied research (irradiation experiments, materials testing), design and scenario evaluations. Proposals covering these fields have been prepared for the 5th Framework Programme.

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