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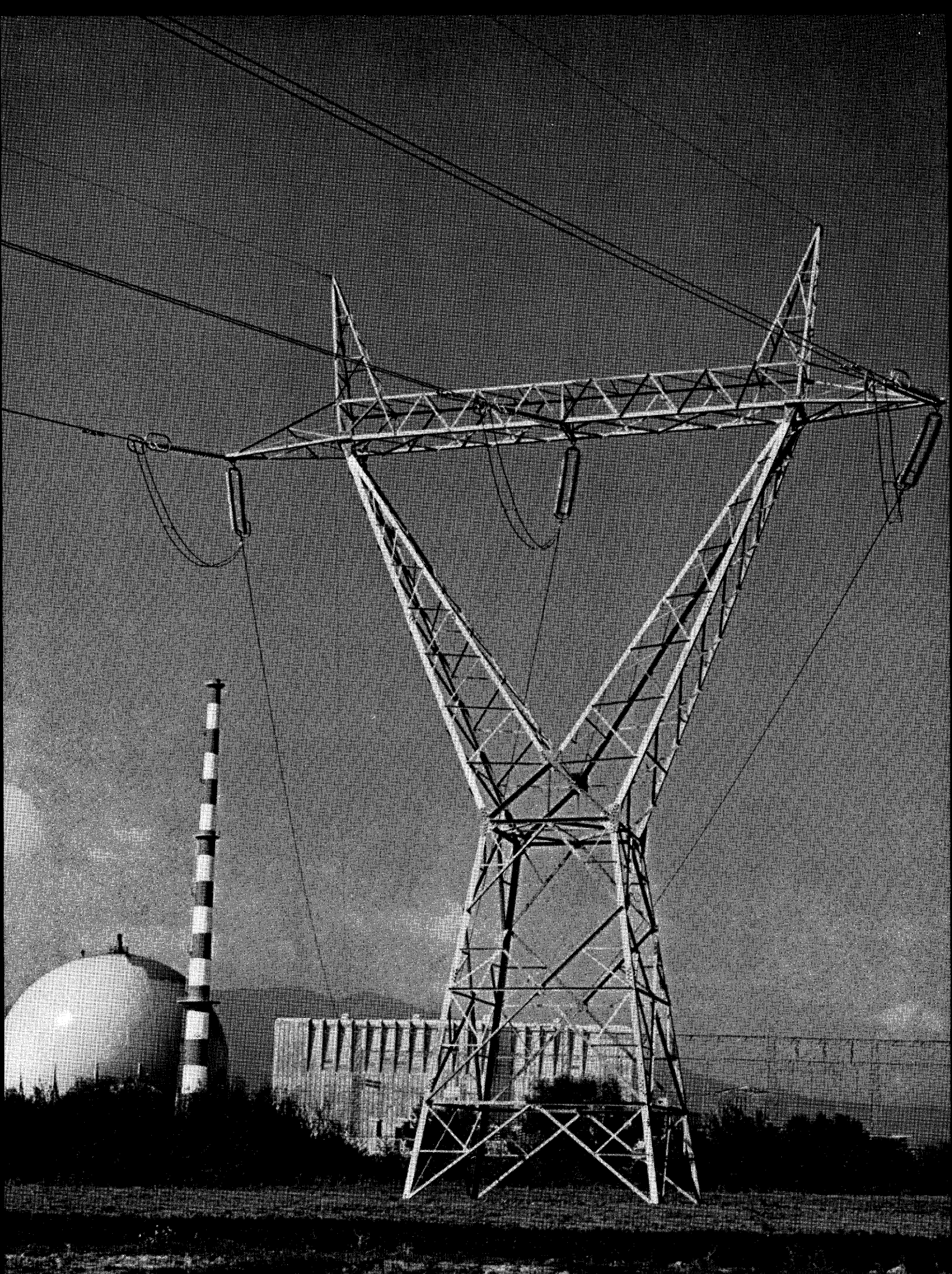


Information Bulletin of the European Atomic Energy Community

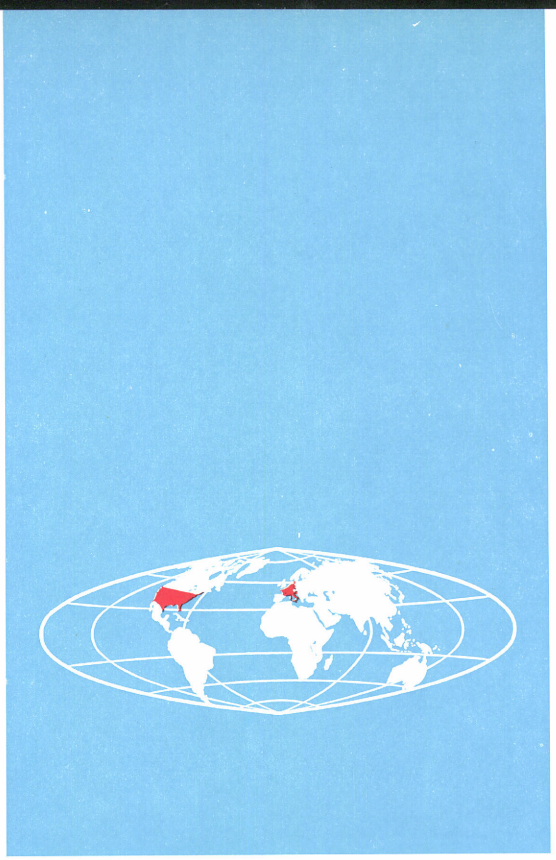
1963 - 1

The Community's mission is to create the conditions necessary for the speedy establishment and growth of nuclear industries in the member States and thereby contribute to the raising of living standards and the development of exchanges with other countries (Article 1 of the Treaty instituting the European Atomic Energy Community).

During the first year of Euratom's life, 1958, an Agreement for Co-operation with the United States of America was negotiated and signed.

"Co-operation" is a term which is widely used in international affairs nowadays, and it is perhaps not unfair to say that it can describe anything from the most tenuous of ententes to the most effective of partnerships. In spite of the vicissitudes which have attended its implementation, it is undoubtedly at the businesslike end of the scale that the US/Euratom Agreement can be placed: it has led to the building of several nuclear power plants, one of which will shortly be commissioned; it has meant the launching of a joint programme of research and development which has already begun to yield its fruits.

In the following pages, most of which are devoted to the US/Euratom Agreement, an attempt has been made to bring out its significance by showing its aims and achievements and by spotlighting some of the fields of inquiry with which it is directly concerned.



The US/Euratom

FEDERICO CONSOLO

Adviser to the Euratom Commission

The Agreement for Co-operation between Euratom and the US Government was signed in Brussels on 8 November 1958, i.e. during the first year of Euratom's existence. In addition to its intrinsic value from the scientific, technical, industrial and financial standpoint, the Agreement was of considerable political importance, since from the very outset Euratom was able to benefit by the assistance of the world's major nuclear power.

The Agreement was based on two well-defined and closely related programmes:

(a) a programme for the construction of power reactors of a proven American type, for a total of 1000 MWe, to go into operation before 31 December 1963, with the exception of two power plants scheduled for operation by 31 December 1965;

(b) a joint research and development programme on the types of reactors accepted under the power reactor programme, the budget for which, in the first five years, would total 50 million dollars for each partner (Euratom and US), each in its own territory.

Under this agreement, the US Government undertook to supply Euratom with 30 tons of contained uranium 235 (29 tons for the reactor programme and one ton for the research and development programme). Deferred payment facilities were granted for the first time, and favourable conditions were also offered for processing of irradiated fuel and for the repurchase of a certain amount of the plutonium produced. It is worth recalling that, for the period extending from then up to the end of 1961, the US Government had a maximum total of 50 tons of uranium 235 for its "Atoms for Peace" programme, under which the Agreement with Euratom was drawn up.

Furthermore, the US Government undertook to grant Euratom a long-term line of credit, via the Export-Import Bank, of 135 million dollars, which Euratom could in turn loan to participants in the power reactor programme.

Finally, the US Government recognised Euratom's value as an organ of inspection

Agreement for Co-operation

by ceding to it the right of control which it had exercised over the strictly peaceful uses of fissile material in the Six, as provided for under the bilateral agreements concluded with Euratom member countries.

The Agreement set up standards and procedures for the launching of both the reactor programme and the research and development programme, which led to the establishment of two joint boards by the Euratom Commission and the US Atomic Energy Commission (USAEC), its counterpart in the US:

(1) *The Joint Reactor Board*, which examines the proposals submitted for the construction of power reactors and then submits a report embodying its recommendations to the two Commissions. The Board is presided over by a chairman (Euratom's Director General for Industry and Economy) and vice-chairman (The Head of the Nuclear Section of the US Mission to the Communities) with voting rights, and an equal number of Euratom and American experts with no voting rights.

(2) *The Joint Research and Development Board*, which studies proposals for research under the research and development programme and takes the appropriate decisions. The Board consists of two executive members (Euratom's Director General for Research and Training and the Head of the Nuclear Section of the US Mission to the Communities), who both possess voting rights, and an equal number of Euratom and American experts, who do not.

Despite the complications which arose during the implementation of the Agreement and the difficulties encountered as the work progressed, these two joint boards have functioned very satisfactorily thanks to the comprehensive and cordial attitude on both sides.

It should not be forgotten that this Agreement was negotiated in an atmosphere of concern as to the possible consequences of the Suez crisis at a time when the illusion was prevalent that it would not be long before nuclear power could compete with conventional sources of energy. It is therefore not surprising that,

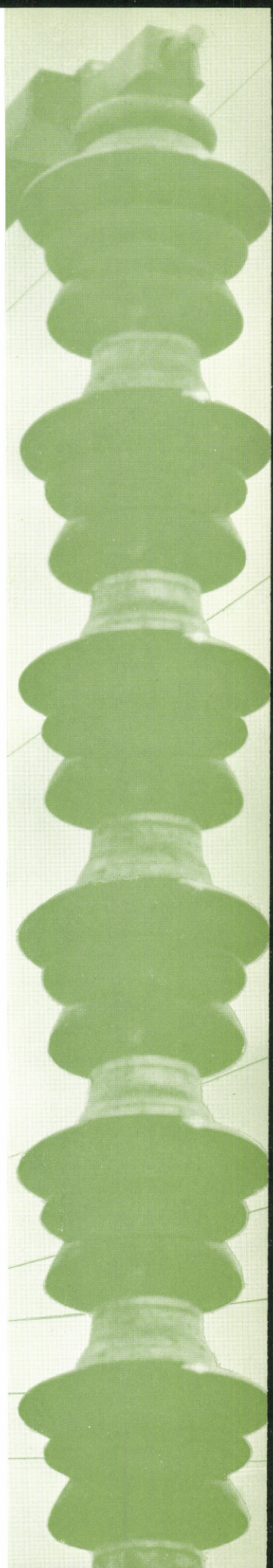
once the initial upsurge of enthusiasm had waned, and after the situation with regard to conventional power supplies in Europe had returned to normal, it was realized that the Agreement had overreached itself on the industrial side, at the same time being too limited from the research angle. These considerations sparked off criticism in Europe, the Agreement being accused of both subordinating Euratom to US policy and of investing it with excessive powers in its own sphere of influence. Notwithstanding this criticism, and in the light of its achievements during the four years of its existence, the Agreement can now be regarded as a success.

The research programme has progressed satisfactorily and the reactor programme will give rise to a total capacity of 700 to 800 MWe, as against the 1000 MWe initially contemplated.

As the ties between Euratom and the USAEC developed, the Americans realized that these relations ought to be extended and made somewhat more flexible. It was with this aim in mind that, at the end of 1961, an important "supplementary agreement" to the Agreement for Co-operation was negotiated, which freed Euratom from the restrictive interdependence which had originally existed between the reactor construction programme and the research programme.

In line with the terms of this supplement, the USAEC is now supplying Euratom with up to 3000 kg of uranium 235 for its own research programme. Moreover, the balance of the 30,000 kgs of uranium 235 made available under the Agreement which is not utilized for either power reactors or for Euratom's own research purposes can be used by Euratom to supply reactors which do not come under the joint programme.

In conclusion, taking stock of what has been achieved under the US/Euratom Agreement for Co-operation, it may be stated quite objectively that, after initial difficulties and results which were at first apparently disappointing, Euratom has managed to break through the narrow framework of the original Agreement by endowing it with a flexibility which is of benefit to both parties.



U S / E U R A T O M A G R E E M E N T

Power Reactor Programme

Garigliano, Chooz, Gundremmingen.

These three places form on the map of Europe a triangle which is symbolic of the United States/ Euratom Joint Power Reactor Programme, under which three nuclear power plants are being constructed.

— The first, on the banks of the river Garigliano, will within a few months be supplying electricity to the new industries of Southern Italy. Construction and operation of the plant are being undertaken by the Società Elettronucleare Nazionale (SENN).

— The second is under construction at Chooz, near the Franco-Belgian frontier, the enterprise concerned being the Société d'énergie nucléaire franco-belge des Ardennes (SENA). Whereas the Garigliano Power Plant, which is a clear landmark for miles around, has a shape which unequivocally betrays it as a nuclear installation, the Chooz Power Plant will not be so easily identifiable, since the reactor and several of its allied facilities will be concealed in one of the slopes of the Meuse valley. The reactor is due to reach criticality at the end of 1965.

— The third plant, whose start-up is also scheduled for the end of 1965, is to be set up at Gundremmingen, in the heart of Bavaria. It is designed on the same lines as the Garigliano Power Plant, i.e. it will be equipped with a boiling-water reactor. The constructors are the RWE/Bayernwerk G.m.b.H. group.

The following two pages contain a description of the main features of these three power plants.

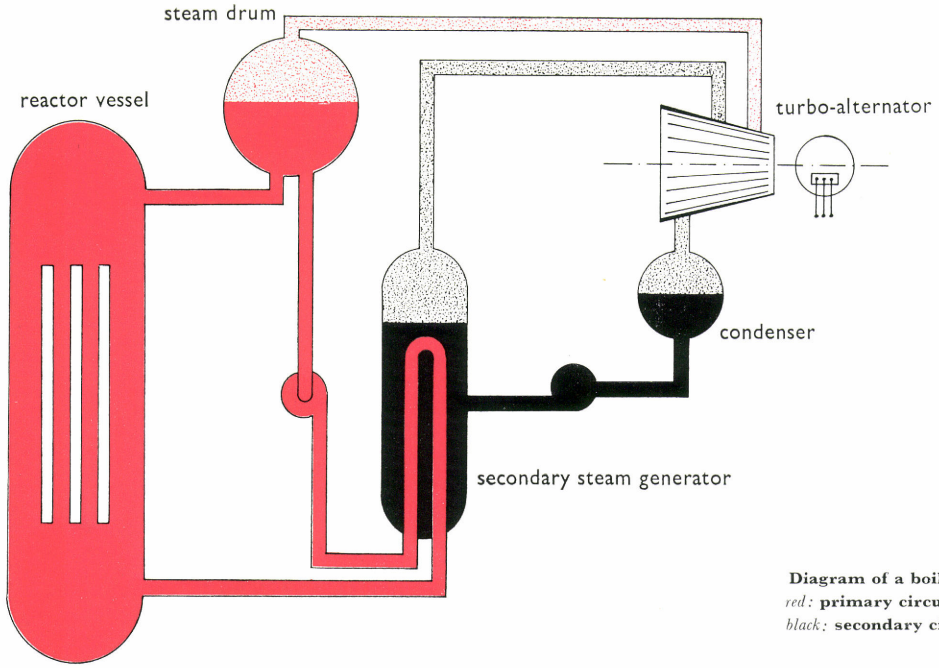


Diagram of a boiling water reactor (BWR)
red: **primary circuit (water/steam)**
black: **secondary circuit (water/steam)**

The Garigliano Nuclear Power Station (SENN)

The power station is equipped with a dual cycle forced circulation boiling water reactor, and with a group of turbo-alternators of 160.000 kW. No heat-exchangers are required since the one water/steam circuit serves both as reactor coolant and working fluid. The coolant also serves as moderator.

A large sphere made of steel contains all the nuclear parts, i.e. the reactor itself, the two secondary steam-generators, the various auxiliary systems and the fuel handling equipment.

The turbo-alternators, the condenser, the feed-water system and the power station's auxiliaries are housed in a separate building to which the office-block and control room are attached.

Liquid and solid wastes discharged in course of operation will be diverted to a neighbouring building for treatment. Waste gases will be channelled to the 100 m high stack, and thence diluted and dispersed in the atmosphere.

The RWE/Bayernwerk Nuclear Power Station (KRB)

The design of this power station is similar to that of the Garigliano plant but incorporates certain improvements. It is to be equipped with a dual cycle forced circulation boiling water reactor and with a group of turbo-alternators of 250 000 kW.

	SENN	KRB
Net electric power of plant	160/150	250/237
Reactor		
Type of reactor	BWR	BWR
Thermal power of reactor	506.3 MW	801 MW
Reactor vessel — inside diameter	3.580 m	3.710 m
Reactor vessel — outside diameter	11.306 m	15.875 m
Reactor operating pressure	71.3 ata	71.3 ata
Fuel		
Fuel material	UO_2	UO_2
Initial enrichment	2.1-1.6%	2.72-1.86%
Total weight of UO_2 in reactor	46.96 t	57.8 t
Cladding material	Zr-2	Stainless steel
Number of fuel channels	208	368
Number of fuel rods per channel	81	36
Total number of fuel rods	16,848	13,248
Coolant/Moderator		
Feedwater inlet temperature	190.5° C	188° C
Total coolant flow through the core	8, 348 t/h	12, 250 t/h
Steam production in reactor	700 t/h	999 t/h
Steam pressure at reactor outlet	69.6 ata	69.6 ata
Steam temperature at reactor outlet	284° C	284° C
Number of secondary steam-generators	2	3
Steam production in one generator	109 t/h	145 t/h
Secondary steam pressure	35.2 ata	35.2 ata
Secondary steam temperature	242° C	242° C

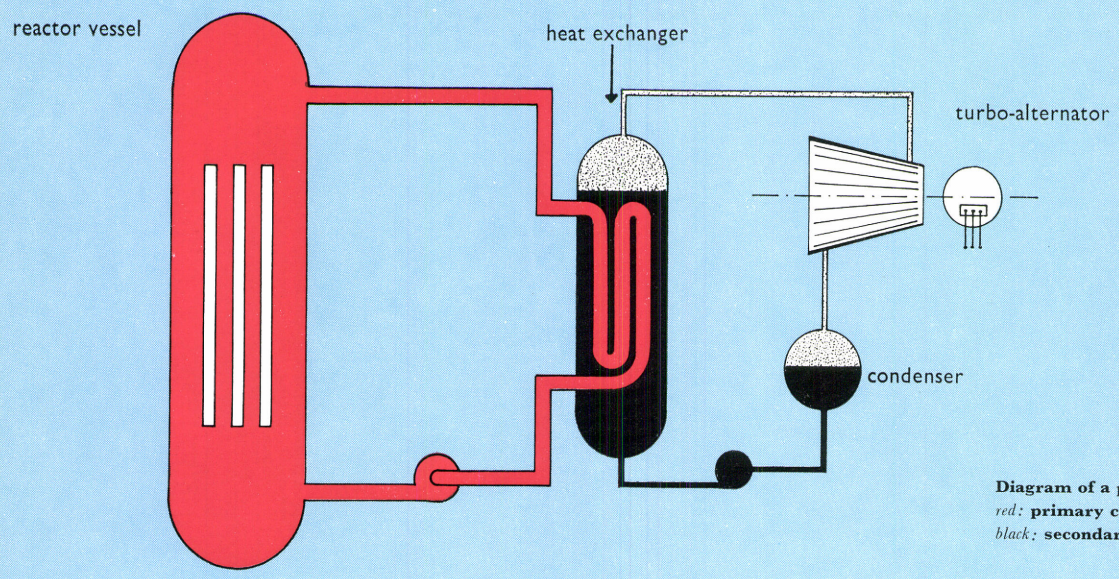


Diagram of a pressurized water reactor (PWR)
red: **primary circuit (pressurized water)**
black: **secondary circuit (water/steam)**

The Ardennes Nuclear Power Station (SENA)

The power plant is equipped with a pressurized-water reactor and a 258,000 kW turbogenerator group. The pressurized water circuit is used solely to cool the reactor. The heat generated is passed through a set of exchangers into a second circuit which supplies steam to the turbines. A notable feature of the plant is the fact that some of the main parts will be located underground. One cavern will house the reactor, the heat-exchangers and the pressurizer, while a second contains auxiliary equipment, such as the deactivation swimming-pool.
The turbogenerators, control room, offices and waste processing section are all located above ground.

Main Characteristics of the Plant	
Net electric power	258/242 MW
Reactor	
Reactor type	PWR
Thermal power	825 MW
Internal diameter of pressure vessel	3.20 m
Overall height of pressure vessel	11.90 m
Fuel	
Fuel	UO_2
Initial enrichment	3.5%
Cladding material	Stainless steel
Number of assemblies	120
Number of fuel element tubes per assembly	208
Total number of element tubes	24,960
Total weight of fuel	44.5 tons
Coolant/Moderator	
Volume of water in primary circuit	142.7 m^3
Normal pressure in normal operation	150 ata
Lower temperature	265.7° C
Upper temperature	298.7° C
Number of exchangers	4
Steam pressure at full power	34 ata
Steam temperature at full power	241.7° C
Total throughput	1,430 t/h

The Joint US/Euratom Research and Development Programme

PIERRE KRUYS *Directorate-General for Research and Training, Euratom*

Historical background

Mr. Consolo recapitulates in his article the circumstances in which the Agreement for Co-operation between the European Atomic Energy Community and the US Government was negotiated. Too much stress cannot be laid on these circumstances, for they have left their mark on the Agreement to such an extent that they go a long way towards explaining the rather chequered history of the Research and Development Programme.

The Agreement for Co-operation, signed with the US some months after the birth of Euratom, was indeed a proof of US support for the Community organizations as the corner-stones of the new united Europe. It also paved the way for the establishment of the first contacts of the technical departments of the Euratom Commission with nuclear centres and industrial concerns in the Community countries, as well as for the achievement of a certain number of concrete aims. Although there is a slight tendency at the moment to lose sight of this fact, the Joint Programme did play a vital role, enabling the Commission to select its targets without undue haste on the basis of medium-term projects such as Orgel or on long-term schemes such as the fast reactor programme.

Furthermore, the Joint Programme suffered from the "nuclear recession", the first signs of which appeared in 1958, the year in which Euratom got under way. Several considerations on which the Three Wise Men had based their short-term forecasts did not materialize. It is probable that if things had turned out differently, the

Signature of the US/Euratom Agreement for Co-operation, on 8 November 1958. From left to right: Enrico Medi, Vice-President of the Euratom Commission; John A. Mc Cone, then Chairman of the USAEC; Heinz L. Krekeler, Member of the Euratom Commission.

need to resort to nuclear energy as a source of power would have emerged much earlier and with a great deal more urgency.

Does this mean that under these circumstances the year 1958 might have marked the beginning of a boom in nuclear power plant construction? We do not think so, for the complexity of the problems involved in adapting prototype power plants to industrial use (the large plutonium-producing piles built in the US after 1945 may be disregarded, for military requirements have obviously ousted economic considerations) has been seriously underestimated, so that the initial forecasts of the cost price of the nuclear kWh have proved highly optimistic.

However, a protracted crisis on the conventional fuel market would have placed the Community in a situation similar to that which faced Great Britain in 1955 and would have necessitated the adoption of an appropriate industrial action policy. This seems to have been the basic idea behind the Agreement for Co-operation: to enable the Community to acquire the means of satisfying its new power requirements and at the same time to make it possible to speed up the development of a European nuclear industry with the assistance of American know-how.

This brief summary shows that the Agreement for Co-operation had a definite *industrial* aim which, the original motivation having been temporarily invalidated by the course of events, acted as a serious stumbling-block to the further development of the Joint Programme in which both European and American industrial firms had displayed a most lively interest.

The initial stages of the Joint Research and Development Programme

Ample evidence of the existence of this interest is provided by the fact that, during the year in which the Programme was launched (1959), the Joint Board received 115 proposals from research and industrial bodies in Community countries, 79 from the US and 82 research projects which were submitted jointly by American and European firms.

In view of this, why did the Joint Programme progress at a much slower rate than had been anticipated?

The reason resided in the fact that this programme is closely linked to the construction of six or seven nuclear plants of a design proven in the US (BWR, PWR and OMR[1]). It will be recalled that the aims fixed for the first stage of this programme (the construction and start-up of four or five reactors by 31 December 1963) were not achieved, for only the BWR reactor, proposed by the SENN (Italy), was actually given final approval[2].

Since that time (early 1960), in spite of the wish of the Euratom Commission, the Joint Programme was confined to research relating to the development of BWR-type plants, with a consequent reduction of its original scope. This was certainly not due to an act of petulance, and probably only partially the result of a feeling of disappointment: the chief cause, in our opinion, is likely to be a political conditioned-reflex—one might even say self-defence.

During this *first stage*, the Agreement for Co-operation did not in fact result in *one single* new project for the construction of a nuclear power plant, the SENN having already decided back in 1958 to erect a 150 MWe BWR plant. The failure of this first stage was felt all the more severely in view of the great hopes which had been pinned to it, while it caused a definite wavering on the part of American political and industrial circles. Tribute should be paid to-day to those who, under these difficult conditions, managed to safeguard the principle of close collaboration between the US and the Community. It is likely that the breakdown of the Agreement for Co-operation would have had serious repercussions on *all* our relationships and the question might well be asked whether, in such an event, co-operation on as vast a scale as that at present envisaged for the development of fast-neutron reactors could have materialized at all. It is misleading to imagine that an effective scheme of technical collaboration can be brought into being overnight on the basis of a mere agreement, for the process involved is a slow one, during which the partners must first get to know each other. Here, too, the Joint Programme served as an important stimulus.

It is gratifying to note that the Joint Programme was only briefly shelved and that the liberal interpretation applied since the second half of 1960 has enabled a series of concerted projects to be embarked upon, both in the Community countries and in the US. Evidence of this is provided by:

—the increasing from 1 to 9 kilograms of the quantity of plutonium put at the disposal of the Joint Programme for research undertaken in the Community countries;

—the organization of working meetings in order to bring

1. Organic moderated reactor

2. The second stage of the construction programme, aimed at the commissioning of two nuclear power plants by the end of 1965, was, on the other hand, fulfilled, owing to acceptance of the Franco-Belgian SENA (150 MWe PWR plant) and the German KRB (237 MWe BWR reactor) projects.

together the technicians employed by the European and American contractors and the Euratom and USAEC experts;

—the opening of the periodic meetings organized by the USAEC on specific subjects (uranium carbide, fabrication of UO_2-based fuel elements by certain methods, etc.) to European firms and institutes holding contracts under the Joint Programme.

These meetings are extremely useful, for they enable direct contact to be established between the laboratories, and do in fact constitute a valuable source of original information for the people engaged in research on the European side. Moreover, by linking up the research work carried out under the Joint Programme with that performed under its national programme, the USAEC showed that it did not affix a "European" or "American" label to the work of the Joint Programme according to the nationality of the contractors.

Figures on the Joint Programme

The following table gives the total number of research proposals submitted to the Joint Board on 31 December 1962 and for which contracts were concluded.

Table

Category	Community proposals		American proposals		Joint proposals	
	sub-mitted	ap-proved	sub-mitted	ap-proved	sub-mitted	ap-proved
BWR	44	14	47	17	18	4
PWR	6	—	5	1	16	1
OMR	16	—	2	—	21	—
Nuclear superheating	2	—	3	—	7	—
General interest	104	31	100	17	29	3
Total	172	45	157	35	91	8

A close analysis of these figures makes it possible to single out the following points:

1) For the reasons given above, the approved projects relate mainly to research into the development of boiling-water reactors and research of general interest.

2) The number of research proposals which are eligible under the PWR programme is still limited. This is due to the fact that such studies have only been admissible since July 1962, when the Franco-Belge des Ardennes plant (SENA) was incorporated in the power reactor construction programme under the Agreement for Co-operation.

3) The research on the development of the OMR reactor string was put forward in response to the first invitation for proposals issued by the Joint Board and could not be considered in view of the fact that construction of a plant of this type was not approved.

4) Research into nuclear superheating is eligible in view of the fact that this system represents a natural development of light-water-moderated reactors. However, so far, the Joint Board has been hanging fire on projects of this kind.

5) The heading "proposals of general interest" covers basic research into materials (fuels, structural materials), irradiated fuel processing, the processing and storage of radioactive waste, etc.

The total credit appropriated under the Joint Programme both in the United States and in Europe comes at the moment to about $ 28 million. At the present rate it can be estimated that the total will come to about half of the 100 million which were earmarked originally. In view of the Joint Reactor Programme's evolution, this is a natural development.

Broad outline of the Joint Programme

It is not possible within the scope of this article to give even a brief enumeration of all the research work covered by the total of more than 75 contracts. Specialized readers can obtain more detailed information from the "Joint Research and Development Programme Quarterly Digest", published jointly by Euratom and the USAEC.[3] We have restricted ourselves to providing a general outline of the main activities.

Development of fuel elements and cladding materials

The studies on the development of fuel elements are centred on uranium oxide. As can be seen from the article of Mr. Fernet de Boisdeffre (pages 14-21), the bulk of the effort is focused on the finalizing, from the technological standpoint, of the most promising fabrication processes, the principal objective being to enable the industries of the Community countries to obtain the experience required in the manufacture on an industrial scale of the charges for uranium-oxide-fuelled power reactors.

This does not mean that the studies on the irradiation behaviour of ceramic fuels are being neglected, and a research project is now in progress aimed at establishing the limit conditions for the use of uranium-oxide-based fuels in light-water reactors. The principal objective is to

3. All enquiries concerning this publication should be addressed to Euratom, Directorate for Dissemination of Information, 51 rue Belliard, Brussels.

ascertain whether it is possible to permit the partial melting of the uranium oxide in the fuel element.
Basic research into the properties of ceramic fuels is also being carried out. Under this heading a method has been developed for the rapid and easy preparation of uranium oxide monocrystals large enough for the intrinsic properties of the material to be determined.
The research on cladding materials, which bears mainly on zirconium-based alloys, has led to the development of a new alloy (zirconium/niobium 3%, tin 1%) the mechanical properties of which at high temperatures seem substantially better than those of zircalloys.

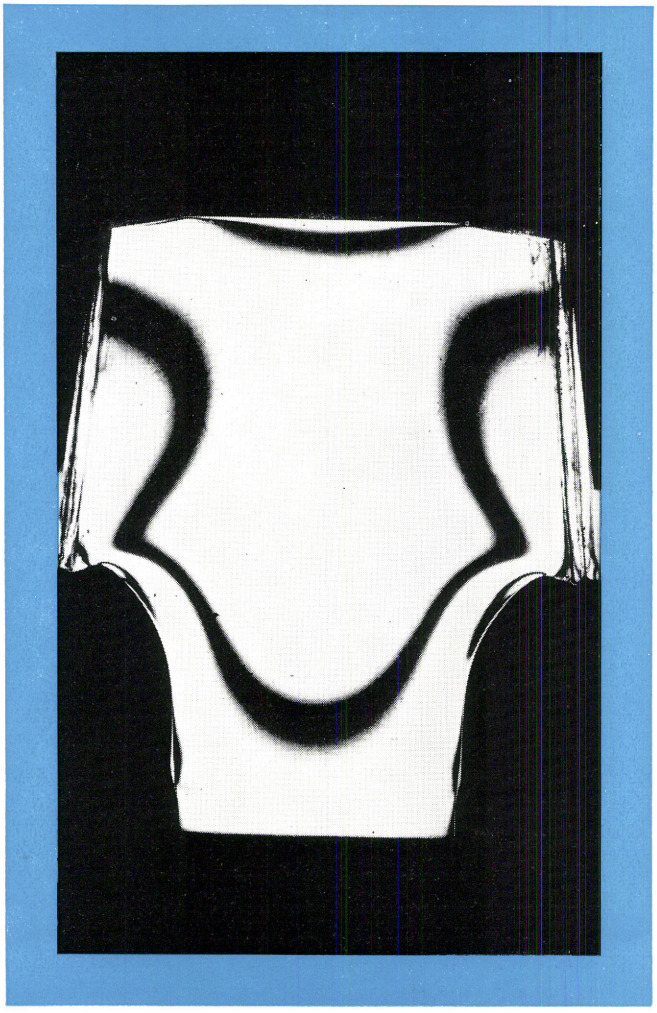
Plutonium recycling in thermal neutron reactors

A concerted effort has been made in this branch of research and work is being devoted to both the neutronics of mixed uranium/plutonium fuels in different moderating media and the technological development of the fabrication of plutonium-containing fuel elements. Mr. Vanden Bemden points out (pages 22-29) the value of using plutonium as a fissile material, stressing at the same time the complex nature of the problems to be solved.

Studies on structural materials

This section of the Joint Programme is known as the "Steels Programme" due to the marked preponderance of steels among the materials used for the pressure vessels and primary circuits of water reactors.
Roughly speaking, the problems to be overcome can be grouped under the following three headings:
a) Selection of materials;
b) Fabrication and utilization;
c) Behaviour in a particular environment.
The problems of selection have led to a series of studies bearing mainly on the classical mechanical properties of interest to the designer and the constructor. It became obvious, however, that the nature of the construction itself, such as a very thick pressure vessel, for example, could give rise to doubts as to the validity of the normal breaking strength standards. This explains the importance of brittle fracture studies in the current programme, where parallel use is being made of normal impact samples (local tests) and samples taken from the entire thickness of the plates (overall tests).
Numerous problems which crop up at the fabrication stage relate to operations such as stamping, chamfering, hot and cold bending, welding, heat-treatment, etc.
In particular, the overall behaviour of the assembly will be to a considerable extent contingent on the choice of the welding method and the conditions in which it is carried

Picture of the stress distribution in a plastic mock-up (study in photoelasticity). This method makes it possible to estimate the distribution and nature of the stresses which would obtain in a metal section of the same dimensions (Photograph: Strength of Materials Laboratory of the University of Ghent).



out, factors which largely determine the quality of the welded seams. Several research studies are therefore devoted to the suitability of various welding methods, some of which are innovations in the field of nuclear construction (electroslag, for example). The problems relating to welding also cover questions concerning the internal plating of vessels by means of surfacing techniques.
Quality specifications also entail the application of stringent non-destructive tests which are frequently very difficult to apply in view of the thicknesses involved. It was therefore thought advisable to compare the most suitable methods of control, carefully weighing the pros and cons and determining their respective limitations, in order to establish a "manual of operations".
On the other hand, numerous research studies have been found to be necessary in order to gain an insight into the behaviour of materials under specific operating conditions, involving various combinations of factors such as high temperatures and pressures, a corrosive medium and neutron irradiation.

Studies on the corrosion resistance of nuclear materials

The corrosion problems encountered in water reactors have almost all been surmounted, but the solution arrived at is frequently a costly one. Furthermore, additional problems will be raised by the development of higher neutron fluxes and more severe temperature conditions, as well as the use of new water-steam mixtures, and it is from this twofold perspective that the aims of the corrosion and oxidation study programme must be viewed.

The basic data relating to the corrosion mechanism are insufficient to obviate the need for systematic, lengthy and expensive tests, and it is for this reason that a major part of the programme has been earmarked for basic studies.

(a) Study of growth and formation of oxide films

In one of the laboratories dealing with this basic research, the oxidation speed of various materials is being studied with extremely sensitive micrometer balances capable of detecting weight changes of a millionth of a gram and thus revealing any quantitative adsorption by the surface studied or any oxide formation. The electron microscope employed in other laboratories is also an indispensable instrument if we wish to gain an understanding of the growth processes of these oxide films, which do not develop in the normal way but from nuclei, as Professor J. Bénard showed some years back.

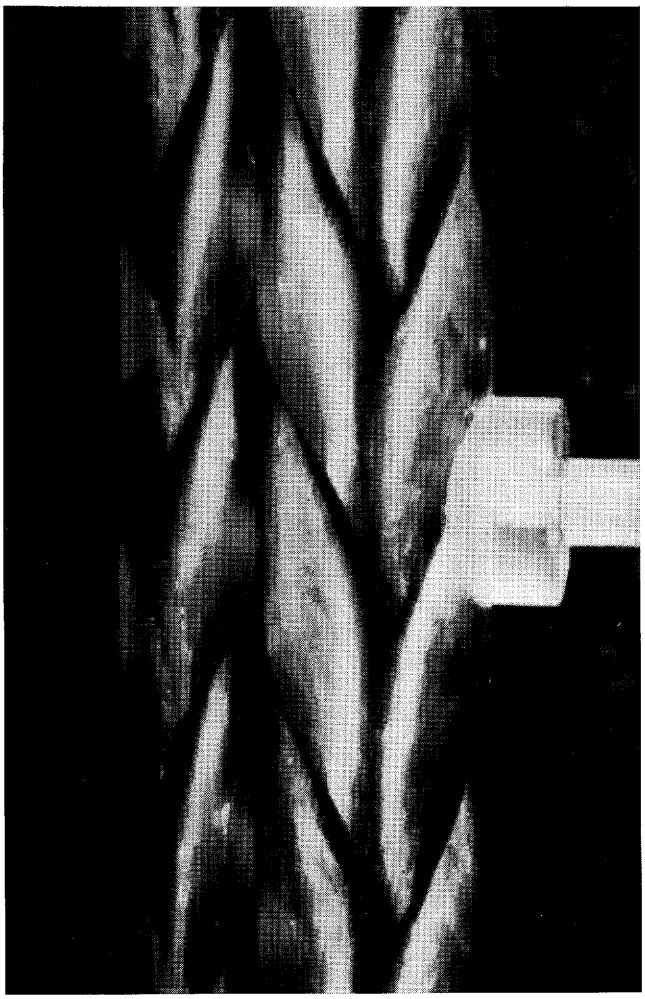
The electron micoscope ia slso used, but in this case by direct transmission over a very thin sample, for studying lattice flaws and their connection with what may be called internal oxidation, i.e. the precipitation of a gas-metal compound within the lattice. This phenomenon may have a considerable influence on the mechanical properties of metals.
Special importance is attached to examining the effect of ionizing particles on oxidizing media, the object being to obtain a more thorough grasp of the manner in which in-pile irradiation can modify the mechanism governing the formation and growth of oxides.

(b) Properties of oxide films

Modern electrochemical methods are particularly useful for studying the protective effect of oxide films.
Measurement of the films' electrical capacity yields valuable information concerning their homogeneity. The plotting of the potentiodynamic curves also makes it possible to assess the resistance of metals to particularly insidious forms of localized corrosion, such as pitting, intergranular or galvanic corrosion. Exactly the same tests are conducted both without radiation and in the CEN's BR1 reactor at Mol (Belgium), in order to see how these phenomena can be modified by radiation.

(c) Factors which are inherent in metal and which affect corrosion

The part played by additives and impurities in corrosion is of major significance. It is for this reason that research is now being conducted into the properties of steels made from high-purity metals, the aim being to ascertain the rôle of impurities in all types of corrosion and to improve the characteristics of the materials by making appropriate adjustments in the criteria adopted for their selection and manufature.
Under this head may also be classed the important project which has been launched to determine the influence exerted by the surface state of carbon or stainless steels on corrosion in water at high temperatures.
A 300° pressurized-water circulation loop, which is under construction, should make it possible to define the degree of corrosion and erosion, as well as to determine the properties and detect deposits of the corrosion particles on various surfaces.
Efforts are being made to secure co-operation between various laboratories engaged on programmes which are frequently similar or which overlap. In the same way, all the laboratories working on stainless steels use as controls standard commercial steel samples taken from the same



Assembly of six spirals surrounding fuel elements and inducing vortex movements in the coolant (Photograph: SNECMA)

plate; this enables a comparison to be drawn between the results obtained by different techniques.

Improvement of heat exchange between fuel and coolant

Although water has outstanding properties as a coolant, the fact that we still have insufficient knowledge of certain phenomena, notably burn-out, necessitates the adoption of arbitrary and pessimistic safety factors, which at present limit the efficiency of light-water-cooled reactors. Hence the reason for the sustained co-ordinated efforts of the US and European laboratories to fathom this highly complex phenomenon.

Research is also being conducted into the development of methods for improving heat extraction in reactor cores. Among these methods, the use of non-isothermal surface fuel cladding and the application of vortex movements in the coolant are of particular interest from both the fundamental and the technological standpoint.

But the most important part of that section of the research programme which is devoted to heat-exchange is undoubtedly the study of the thermodynamic and hydrodynamic properties of water/steam mixtures, with which it is possible to take advantage of the useful properties of both the water (high latent heat) and the gases (low density). The results obtained to date are promising.

Research linked with the development of power plants under construction

The Joint Board has recently authorized the execution of a programme of research on the SENN power plant, which is scheduled for commissioning in the near future. This programme has a two-pronged objective, i.e.:

(a) to amplify the general information on the BWR string by systematically measuring the salient core and primary-circuit parameters, both during start-up and in full-power operation;

(b) a technical assessment of the possibility of stepping up the plant's rated power from 150 MWe to 225 MWe.

This research is being carried out jointly by the SENN and the firms under contract to build the plant, and it is planned to bring the Community's industries into the scheme through the participation of their specialists.

It is hoped that equally valuable research will be able to be conducted on other reactors when they have been commissioned.

Irradiated fuel processing

Research in this field is concerned mainly with the properties of halogenation volatilization. This technique consists in converting the fuel, usually uranium oxide, into a volatile compound, uranium hexafluoride, which is easily separated from the bulk of the fission products. The fissile material thus recovered may be sent, without further processing, to an enrichment plant. However, recovery of the plutonium present in the irradiated fuel raises difficult problems to which solutions remain to be found.

The results obtained have made it possible to undertake the construction of a small plant capable of processing several grams of uranium oxide which has been very slightly irradiated and is therefore virtually plutonium-free. The purpose of this plant is to solve the problems of remote control and maintenance and of satisfactory uranium decontamination, the technological study of the plutonium-recovery conditions being left to a later date.

Uranium Oxide UO_2

PAUL FERNET DE BOISDEFFRE,

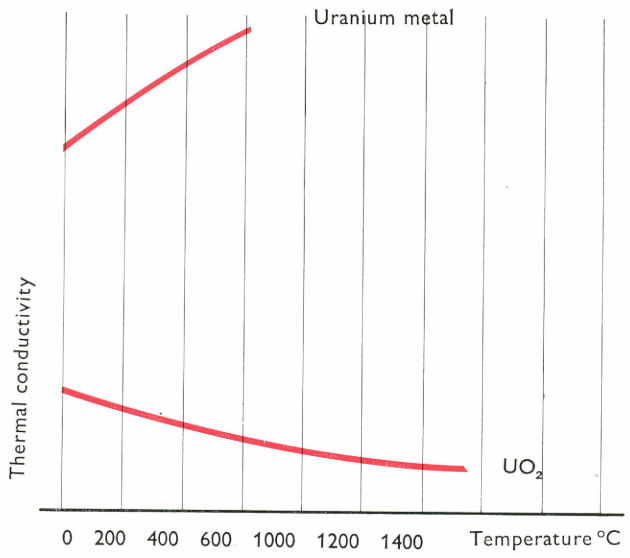
Directorate General for Research and Training, Euratom

Uranium oxide was once a poor relation of uranium metal as a nuclear fuel. It has now made a spectacular come-back and is powering reactors in many parts of the world. Its properties make it the favourite fuel of water-cooled reactors.

The first type of fuel used for the first atomic pile in the world to operate, the Chicago squash court reactor which went critical on 2 December 1942, uranium oxide (UO_2) was later superseded by uranium metal. It has, however, made a remarkable come-back since then and is now in the forefront of fuels used in large-scale nuclear power plants for electricity production.

There is an explanation for these vicissitudes in the fortune of uranium oxide. To be strictly accurate, it has to be acknowledged that the reasons which dictated the use of UO_2 in Enrico Fermi's CP1 reactor, and later also in the first French reactor "Zoé", were not identical with those which have restored this material to its present pre-eminence. In those epoch-making days, the pioneers of the new nuclear science employed uranium in the form developed by metallurgists—i.e. an oxide whose characteristics would appear to us very second-rate today. The rapid advances made in technology soon put into the hands of physicists a fuel in the shape of uranium metal which fully met their requirements from a nuclear standpoint without, however, combining all the qualities desired. By a strange turn of events, not a few examples of which are to be found in the history of modern technology, there has been a gradual rehabilitation of this compound, improved by means of protracted and painstaking research, and today, leaving aside the gas-cooled natural uranium power plants developed in Britain and France, uranium oxide is the fuel used by practically every power reactor in the world. Noteworthy in this connection are the US Navy's submarines, the surface vessels "Savannah" and "Lenin", and the major power plants, more particularly Shippingport, Dresden and Yankee in the US, BR 3, SENN, SENA and RWE in Western Europe and Vver and Ulyanovsk in the USSR. In addition to numerous research reactors, small generators which have been set up in the polar regions and also critical experiments are still fuelled by uranium oxide elements.

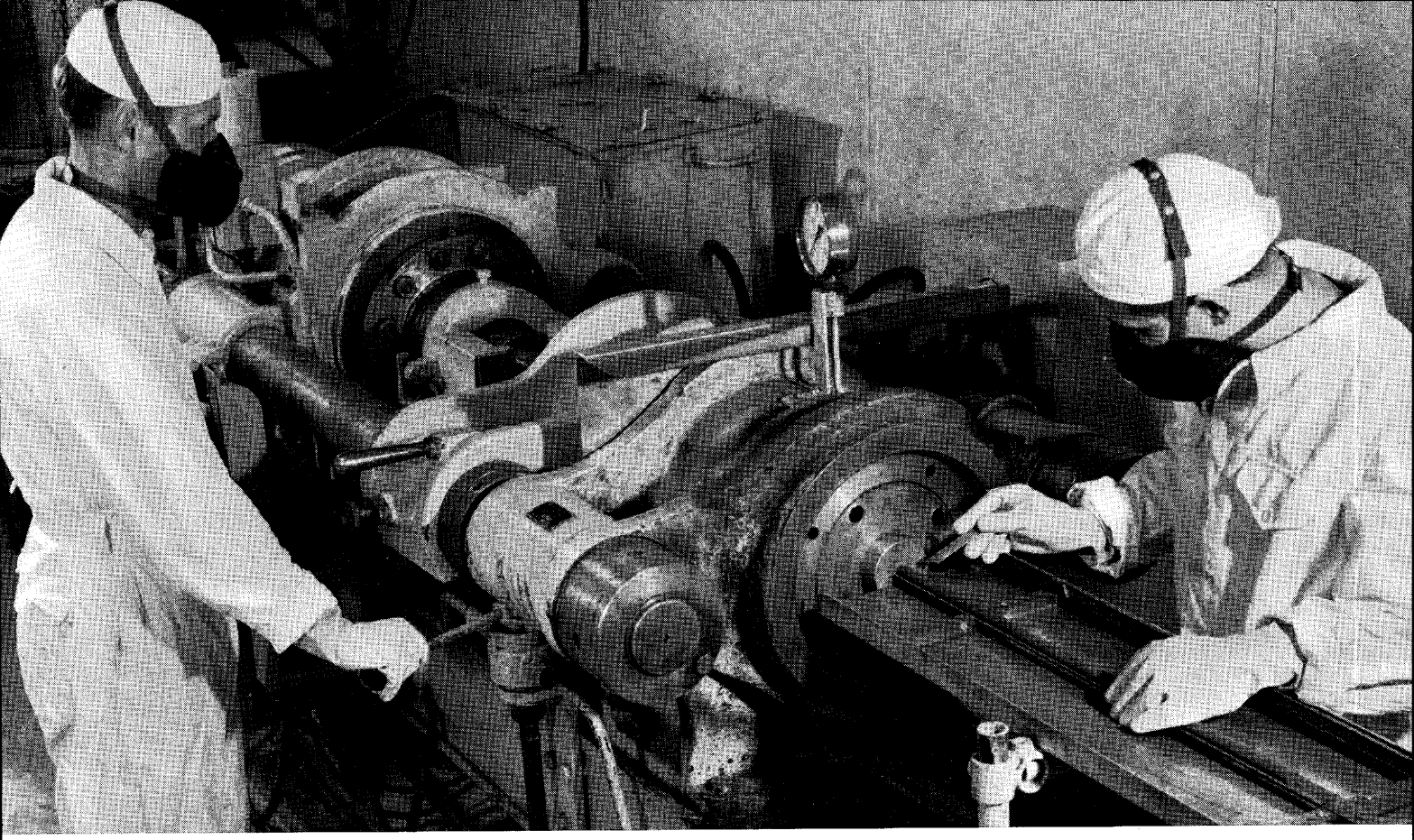
Figure 1
Comparison between the thermal conductivity of uranium metal and uranium oxide.



Why is the use of uranium oxide so widespread?

While it has some undesirable properties, UO_2 combines a number of substantial advantages:

— It has a high melting point, i.e. 2,800°C, as compared with 1,130°C for uranium metal, and its structure is stable



Extrusion of uranium oxide rods (photograph by courtesy of CSF-Georges Bru)

up to this temperature; that of uranium metal, on the other hand, is appreciably modified at 660°C and again at 770°C, which makes it difficult to use for practical purposes above 600°C.

— It is compatible with most cladding materials, which obviates the need to insert between the fuel and its can intermediate layers which not only involve additional expense but which are also awkward to construct.

— It does not, in the event of a cladding failure (an event which rarely occurs but is nevertheless possible, in spite of the precautions adopted in the element fabrication process), react with water or the majority of other cooling liquids used or planned for use in nuclear reactors, even at high temperatures. In the event of more serious incidents or accidents, this constitutes a fundamental safety factor which amply offsets any drawbacks involved.

— The various metallurgical and ceramic processes—described below—required in the manufacture and shaping of uranium oxide fuel elements are relatively easy.

— Its in-pile behaviour, under intense bombardment by neutrons, radiations and various particles, not to mention the fission processes which take place within it, is excellent, even at high temperatures. It is capable of sustaining, without any degradation or appreciable modification of its properties, far higher burn-ups[1] than the pure uranium metal. In particular, the crystal lattice's capacity for accommodating the solid and gaseous fission products, without any variations in the initial dimensions, or at the most with a very slight swelling which the cladding can easily withstand, is a vital factor from the standpoint of intense utilization.

— Its reprocessing, after use in the reactor, in order to recover the residual fissile uranium or the plutonium which has formed and to separate the fission products, raises no more problems, than the re-processing of pure or alloyed uranium metal.

After listing this impressive range of qualities, which militate in favour of the large-scale use of uranium oxide in nuclear reactors in general, and warrant its exclusive use in water-cooled reactors, attention should be drawn to some of the specific problems with which reactor designers are confronted as a result of the material's adverse properties. The chief of these are:

— Its low thermal conductivity which, at ordinary temperatures, is less than a third of that of uranium metal and which, in contrast with the latter, decreases still further as the temperature rises (see figure 1). It must, however, be pointed out that this drawback, although serious, is less significant than it was long believed to be; recent measurements have shown that the thermal conductivity of uranium oxide rises appreciably beyond 1,500°C, in atemperature range which is quite admissible for elements of this type.

1. burn-up = the fraction of uranium atoms in the fuel which has undergone fission.

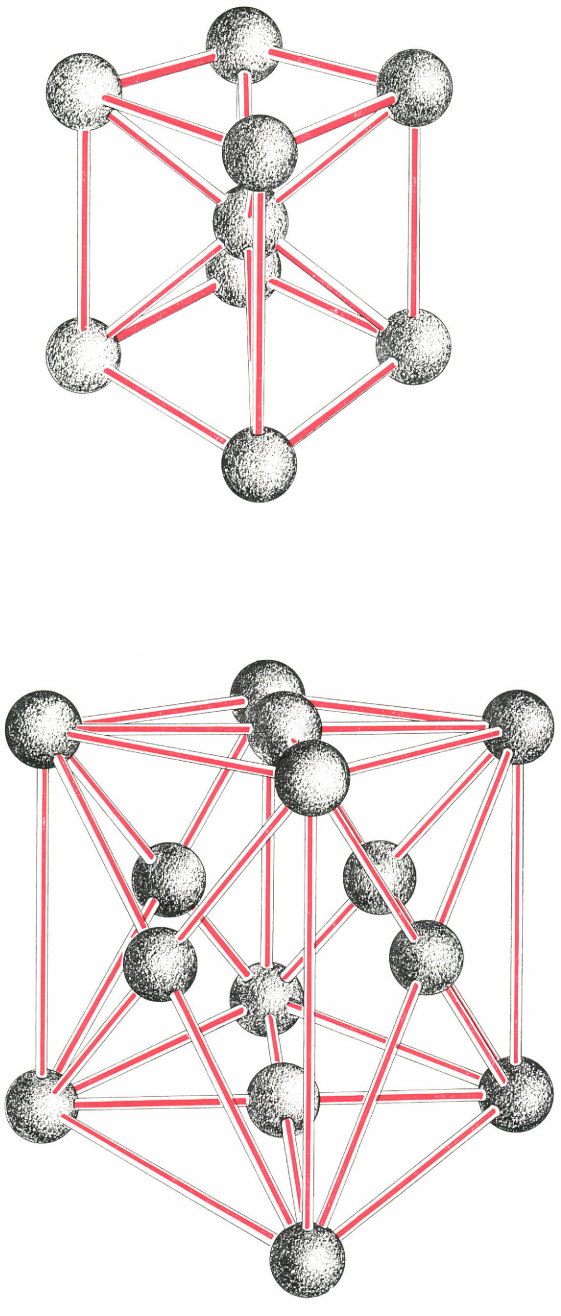


Figure 2
Diagram representing the crystal structure of uranium oxide (below) and uranium metal (above)

— Its unsatisfactory resistance to sharp temperature surges, owing to its low thermal conductivity coupled with the fact that it is a ceramic.
— Its uranium atom concentration per unit volume, which is barely half that of uranium metal and therefore faces reactor physicists with a number of awkward problems connected with neutronics. These can be mitigated by using enriched uranium, which accounts in particular for the success of this type of fuel in the United States. The use of enriched uranium is, moreover, indispensable if the possibility of achieving high burn-ups is to be turned to advantage.

What is uranium oxide?

Uranium oxide falls under the general heading of "ceramic" materials, like graphite or simply fire clay used in pottery. Factors common to these materials are that they have to be produced by high-temperature processes, such as sintering, and that they are refractory-type substances.
The uranium oxide used in reactors has the chemical formula UO_2, although there are several other stable forms in the oxygen/uranium system: U_4O_9, U_3O_7, U_3O_8 and UO_3. However, we are concerned here solely with the UO_2 form, or at least that having the more general formula UO_{2+x}, in which x is lower than 0.15.
The crystal of this oxide has a face-centred cubic lattice (see Fig. 2). Its theoretical density is 10.96 g/cm^3.
From the results of work being carried out in many laboratories on monocrystals of large dimensions (up to 10 mm in diameter), it will undoubtedly be possible to obtain a more thorough knowledge of the true properties

of this material, which hitherto had to all intents and purposes only been studied in the less homogenous state obtained by sintering. Paradoxically enough, it is only in the last few months that physicists have had monocrystals at their disposal in quantities which, albeit still limited, are adequate for carrying out the precision tests without which it is impossible to obtain a complete and reliable definition of their properties. Under one of the research contracts which it has concluded, Euratom is participating in the production of these monocrystals by an original deposition process starting from the vapour phase, and has been able to help to supply the laboratories concerned with the samples they require.

A great deal of headway has already been made with these monocrystals in the field of basic research. It has been established, for example, that the monocrystal's thermal conductivity at any temperature is roughly double[2] that of sintered UO_2.

Studies of the electrical and optical properties are in progress on this ideal structure in which the influence of impurities, grain boundaries and structural defects is to a very large extent ruled out.

Fuel elements

The oldest, and still the most common, form in which UO_2 is used in reactors is that of sintered cylindrical pellets. The most usual dimensions are from 7 mm to 15 mm in diameter, with about the same height.

2. The interpretation of this result is still under discussion, some laboratories being of the opinion that the variation observed might be due to a difference in the monocrystal's oxygen content compared with the sintered product.

These pellets are inserted one on top of the other in cans, mostly of stainless steel or zirconium alloy, which act as the protective cladding. A number of these cans are grouped in bundles to form a "sub-assembly". Not infrequently, the fuel charge of a power reactor comprises several millions of such pellets, several tens of thousands of cans and several tens of sub-assemblies. By way of example, the core of the Yankee reactor in the United States contains over 3,400,000 pellets distributed in the proportion of 150 per can, and more than 23.000 stainless steel cans, grouped into 684 sub-assemblies, which themselves are batched in 76 main assemblies.

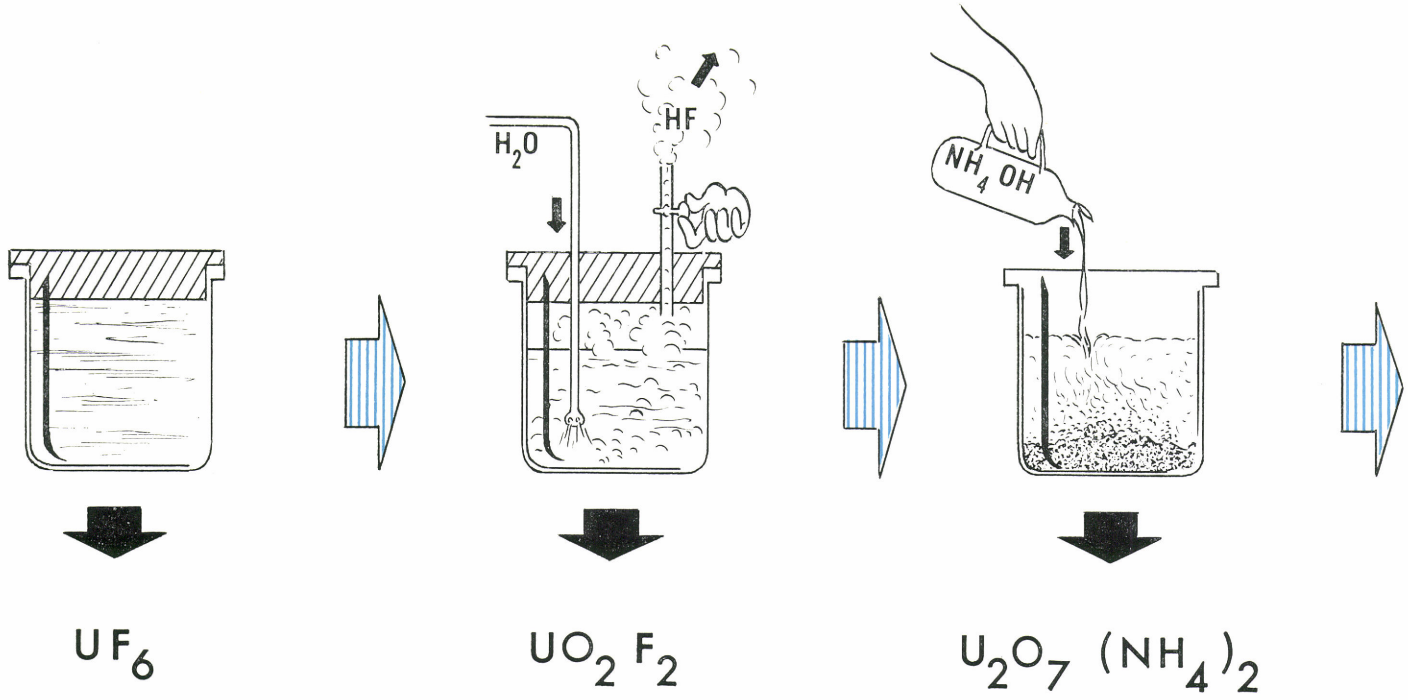
The fuel element fabrication cost, which accounts for nearly 30% of the fuel cycle costs and about 12% of the unit cost of the electricity produced, is an important item in nuclear energy economics. It is for this reason that, while complying with the essential quality requirements, efforts are being made to cut down the fabrication cost, either by simplifying the pelleting process or by developing new techniques.

During the uranium ore processing operations, after many intermediate stages involving extraction and purification, a uranium nitrate solution of very high purity is seen to form. This, on addition of ammonia, becomes transformed into ammonium diuranate, which through heating followed by hydrogen treatment, yields uranium oxide powder, the source material for the fabrication of the fuel elements.

The process for enriched uranium is virtually the same as that for natural uranium, the hexafluoride obtained in the gaseous diffusion plants being converted either into diuranate or else directly into UO_2 by the simultaneous action of steam and hydrogen (see Fig. 3).

For the fabrication of pellets, this powder is usually crushed, mixed with binders—the most widely used of which have a polyvinyl alcohol base—, cold-pressed and then sintered by powder metallurgy techniques (see Fig. 4). Sintering is carried out at temperatures between 1,350°C and 1,750°C, according to the nature and state of the starting powder, and usually in a hydrogen-reducing or cracked ammonia atmosphere. During the sintering, considerable densification of the pellet takes place (from 5-6 g/cm^3 to 10-10.7 g/cm^3), being accompanied by a corresponding reduction in size, for which, of course, allowance must be made in the fabrication cycle. After cooling, the pellets are machined over their entire surface and carefully checked before insertion into the cans.

This series of operations, some of which are protracted, delicate and expensive, has in some cases been simplified.



By preparing powders with well-defined characteristics, it has been possible to dispense with or reduce crushing, eliminate the binder and use relatively low sintering, temperatures of 1,300—1,400°C. Many research projects, some of them under the sponsorship of Euratom, are still being carried out with a view to the improvement of this method.

Among other processes which have been conceived, some have not got beyond the laboratory stage and have in fact virtually been abandoned. This is the case with hot-pressing and slip-casting.

Other methods, however, have proved their worth, in some cases actually enabling reductions to be made in the fabrication costs in relation to pellet-type elements.

Among these methods, mention may be made of the extrusion shaping of long rods (see Fig. 4), which are then sintered in the same way as pellets (a process which has also been studied under a Euratom contract), and the development of vibration and swaging techniques, on which large-scale studies are under way in both the United States and Europe under a number of USAEC and US/Euratom Co-operation Agreement contracts.

While rod extrusion calls for the use of exactly the same type of uranium oxide powder as that required for the preparation of pellets, the powders used in vibration and swaging have quite different characteristics, since the particles, while having a grain size distribution calculated to ensure optimum filling of the cladding tube, must at the same time be dense enough to allow the highest possible overall density to be produced by simple shaking. For instance, while it is now common practice to sinter pellets or rods with a density of 10.4 g/cm^3 or more, i.e. 95% of the theoretical value, it is not possible to exceed 88%-90% of this value by vibration alone, or, when a combination of vibration and swaging is used, 90%-92% in the case of cold swaging, and 92%-94% in the case of hot-swaging. If these processes are the focus of so much attention and effort (with which Euratom for its part is associated on a large scale), it is because they hold out the hope of appreciable savings in fabrication costs.

However—and this is a point to which we shall revert later—, the irradiation behaviour of the vibrated or swaged elements is not as well known as that of pellets, and it is as well to refrain from drawing over-hasty conclusions as regards the imminent supplanting of pellet-type elements by vibrated or swaged elements.

For more special applications, such as nuclear superheating or gas-cooled reactors, fuel element concepts have been evolved in which uranium oxide is no longer utilised in massive form but dispersed in metallic substances, e.g. stainless steel, or ceramic materials, such as zirconium, or beryllium oxide, or in graphite.

From the nuclear standpoint, it is then necessary for uranium to be highly enriched, and certain precautions have to be taken to avoid the release of fission products outside the element. For instance, UO_2 particles of some tens of microns are sheathed in a layer which is impermeable to fission gases and which may be *metallic* (chromium, niobium), *ceramic* (aluminium oxide) or *carbonaceous* (carbon derived from the pyrolysis of methane).

Other reactor types are designed for UO_2 dispersion in thorium oxide or of plutonium oxide in UO_2. These oxides are mutually compatible and can be mixed in any

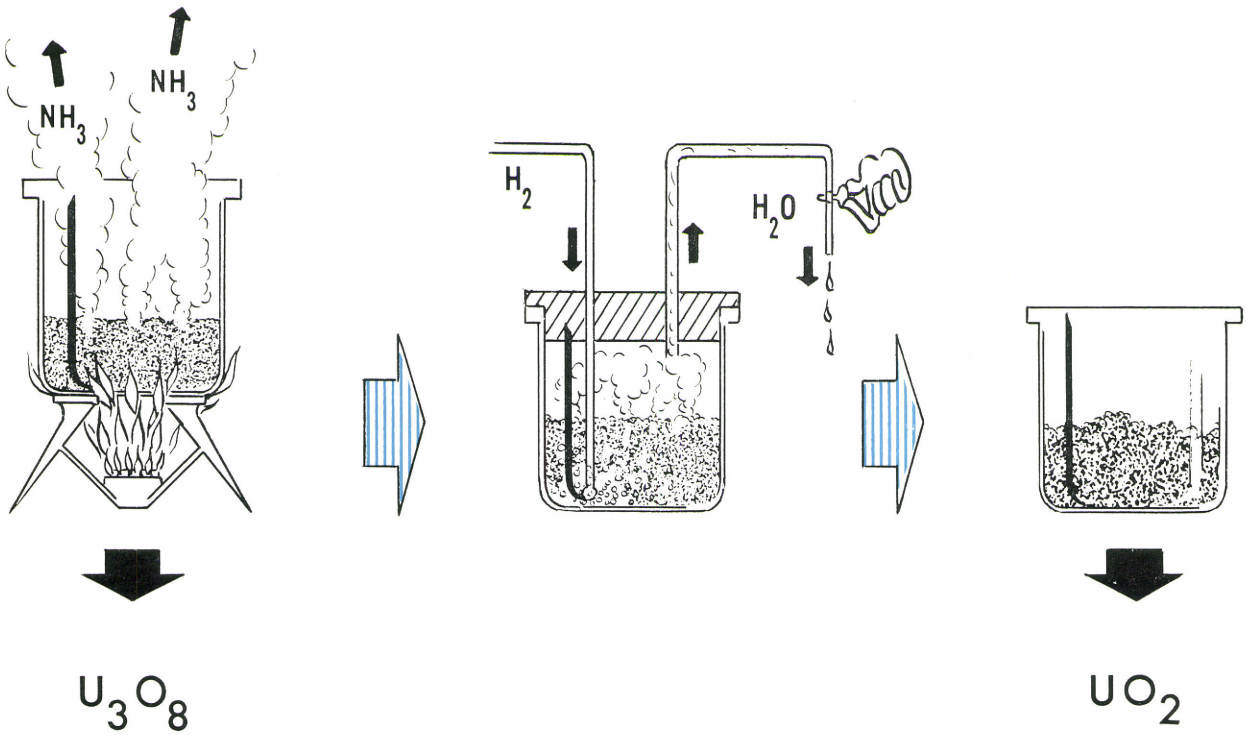


Figure 3
Production of uranium oxide powder from uranium hexafluoride by the ammonium diuranate method

Furnaces used for the sintering of uranium-oxide rods (photograph by courtesy of CSF-Georges Bru)

proportions. They have been and still are the subject of research contracts under the US/Euratom Agreement.

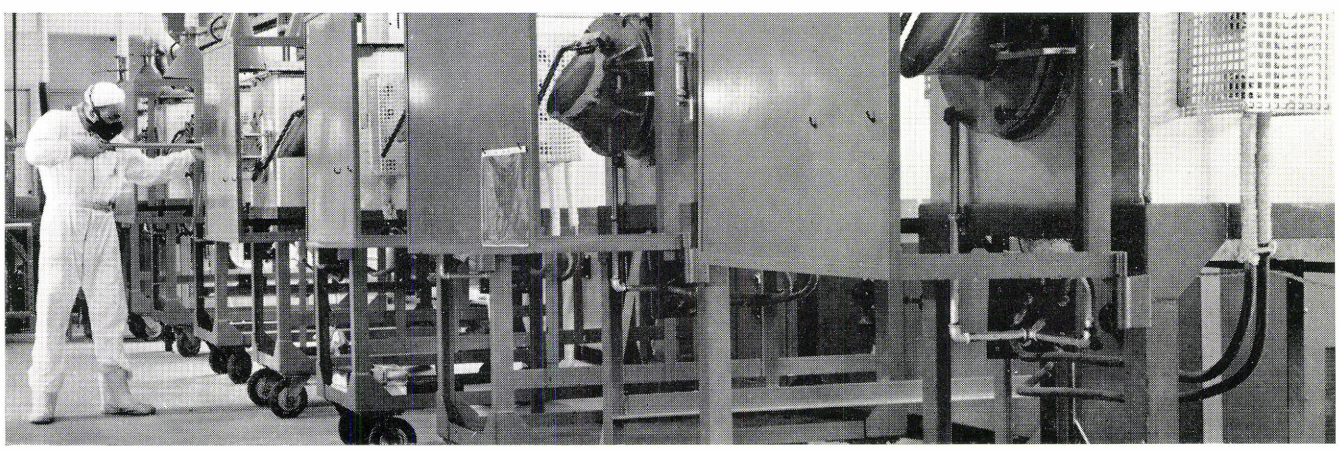
In-pile behaviour

It was stated at the beginning of this article that the irradiation behaviour of uranium oxide was excellent. What does this imply?

Because of its low thermal conductivity, a uranium oxide rod is in normal operating conditions subject to a very pronounced temperature gradient. If the water or mixed water/steam coolant circulating outside the cladding is at the relatively low temperature of 260—300°C, the outer surface of the oxide at once rises to the 500—700° C range as a result of temperature drops at the water/cladding and cladding/fuel interfaces.

The centre of the rod, which is only a few millimetres from the surface, may reach 1,500—2,000°C, which represents a gradient of several hundreds of degrees per millimetre (see Fig. 5). Such a gradient subjects the material to extremely severe thermal stresses, which may (and frequently do) cause the pellet to crack at a number of points. Fortunately for the user, this cracking stops fairly soon and the phenomenon does not reoccur for the rest of the element's life.

The fuel, moreover, undergoes numerous fission processes, which cause the uranium atoms to give place to foreign atoms, either solid or gaseous, which the fission ousts from the position occupied by the uranium atom. If the solid elements thus created are quickly fixed in the new position in which they have landed as a result of the violent nuclear reaction, there is a danger that the gaseous elements



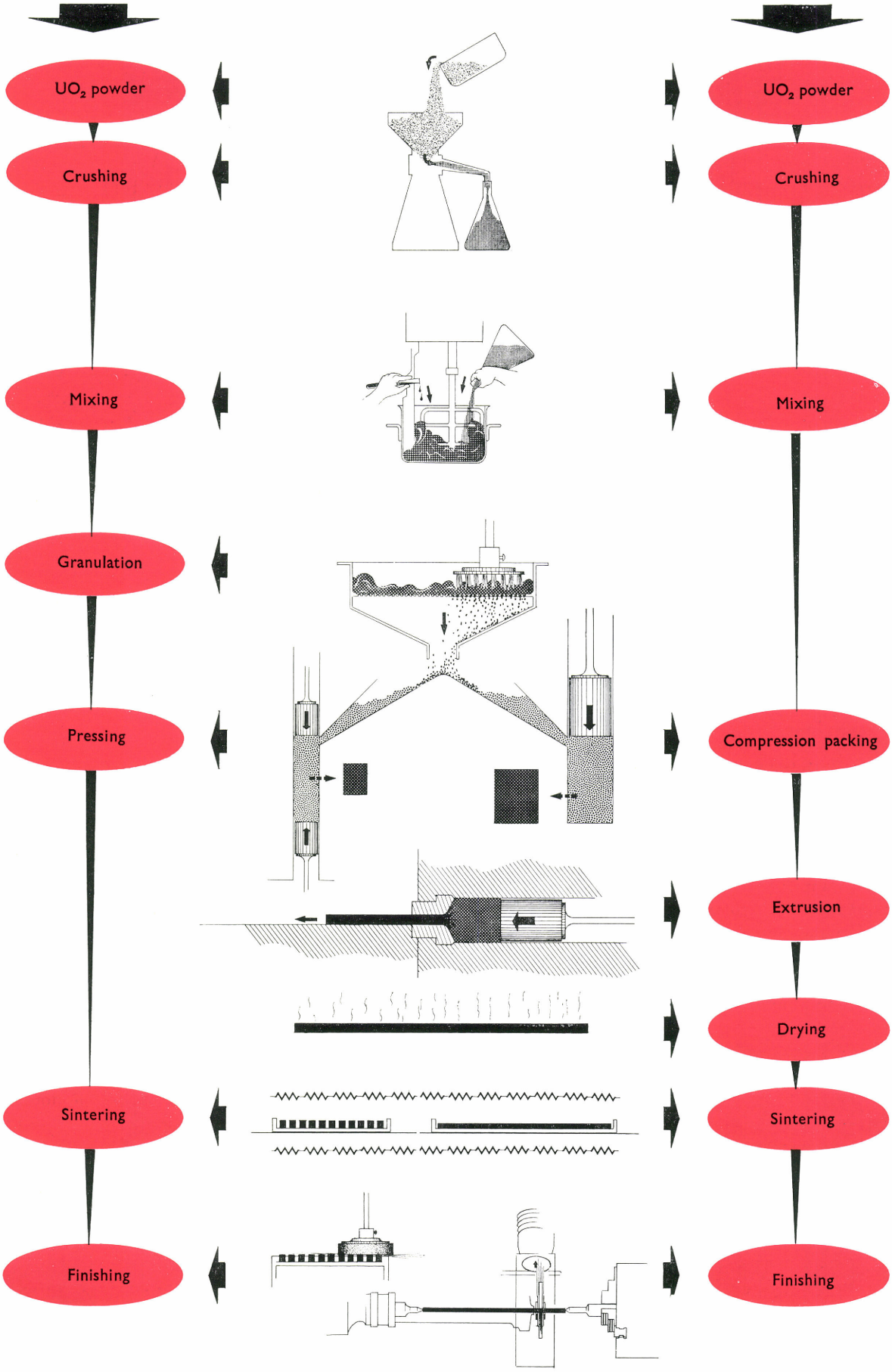

UO_2 powder
Crushing
Mixing
Granulation
Pressing
Sintering
Finishing
UO_2 powder
Crushing
Mixing
Compression packing
Extrusion
Drying
Sintering
Finishing

Figure 4
Diagram showing the process for the production of pellets (left) and rods (right) from uranium oxide powder

(mainly krypton and xenon) will diffuse in the high-temperature zones and rapidly set up pressures liable to burst the cladding.

The diffusion of these gases in uranium metal causes the latter to swell very considerably, so that the fuel has to be removed from the reactor after only a few thousands of MWd/t.[3] (In the CO_2-cooled natural-uranium reactors of British and French design, this maximum burn-up is fixed at 3,000 MWd/t.)

Very fortunately, the extremely advanced experiments show — and the results yielded by the reactors in operation confirm — that almost all the fission gases may stick in the UO_2 lattice, first collecting in small bubbles, which cannot then diffuse, as long as the maximum temperature is kept below 1,600—1,800°C. Moreover, the formation of these bubbles reveals a very high irradiation plasticity on the part of UO_2, at least 40 times that of non-irradiated UO_2 according to recent calculations.

This, one of a number of examples of the changes undergone by the properties under in-pile conditions, shows how difficult it is to extrapolate in-pile the results of the out-of-pile tests, despite the simplicity of these tests from the experimental standpoint.

We have no intention here of giving absolute figures of swelling and fission gas release; it is not difficult to visualize the complexity of the problem and the large number of parameters entailed: density, O/U ratio, dimensions of the sinter grains, distribution and form of the porosities, etc.

However, it may be estimated that the swelling only becomes noteworthy after burn-ups exceeding 40,000 to 50,000 MWd/t and that in a well-designed element the amount of gas released is not more than a few percent of the quantity formed. If the elements are made to work under more stringent conditions, e.g. with the central zone at a temperature close to or higher than the melting point of UO_2, the foregoing remarks no longer apply, as the gases escape in a far higher proportion, which may be as much as 30% or 40%.

In view of the practical importance of an exact solution to this problem, tests are being carried out on special elements, equipped with devices which make it possible to carry out continuous measurements of the pressure within the cans. In addition, a precise estimate of the temperature attained in the fuel is necessary in order to determine the right fission gas release techniques and to predict the long-term behaviour of new elements. Unfortunately, the determination of these techniques is still a very uncertain business, owing partly to the high temperature and partly to the effects of irradiation on the thermo-couple readings.

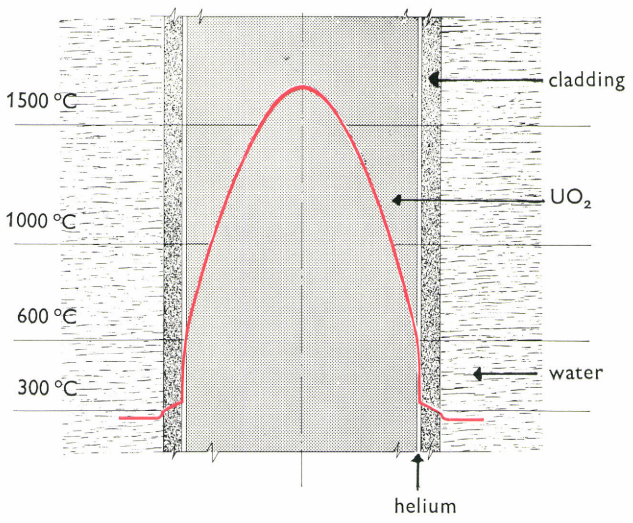


Figure 5
Temperature gradient inside a uranium oxide fuel element

In conclusion, it may be forecast, without claiming that uranium oxide is going to oust the other forms of nuclear fuel—some of which, such as carbide, also have bright prospects—, that in the coming decades UO_2 will be used on a large scale in reactor types which are currently known but which still harbour a considerable potential for further development.

In the longer term, fast-neutron reactors, one of whose salient features consists in their capacity to produce more fissile material than they consume, will no doubt require fuels which harmonize with their special characteristics. It is interesting to note that uranium oxide is in the running and that it has actually been chosen in the case of, for instance, Rapsodie. It remains to be seen whether it will continue to be favoured, but at all events these reactors would not appear to be destined to play a major role in the production of the world's energy for another 15 or 20 years at least. This means that there are still substantial outlets for UO_2 and justifies the scale on which research is being conducted on this material throughout the world.

3. burn-up can be expressed as the amount of energy released per unit mass of uranium. Mwd/t = megawatt day per tonne

The power reactors of the US/Euratom Agreement are water-cooled reactors fuelled by slightly *enriched* uranium. There can be no question of using natural uranium, since U235, the fissile isotope, is only present in the proportion of 0.7%, i.e. in a quantity insufficient for maintaining a chain reaction in reactors of this design. The uranium enrichment process currently used, a highly complicated one, steps up the U235 content by the extra small percentage required. Another method which is, however, now being explored, consists in enriching the fuel with a fissile element that the reactors themselves produce during operation, namely plutonium. The author gives a general outline of the possibilities afforded by this element, at the same time stressing the complexity of the problems involved in its use. Part of the research to which he refers as being directed towards the solution of these problems is being carried out under two Euratom contracts, one with the French Atomic Energy Commission (CEA) and the other with an Association comprising the Belgian Nuclear Study Centre (CEN) and the BelgoNucléaire company.

Plutonium,

a synthetic source of energy

Emile Vanden Bemden

Department Head at BelgoNucléare and Chief of the BelgoNucléare/CEN „Plutonium" Project

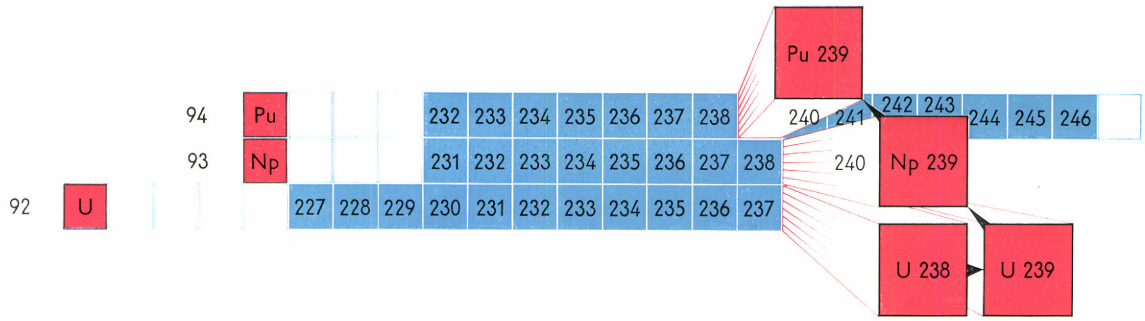
Origin of plutonium

In the winter of 1940-41, G. T. Seaborg, E. M. McMillan, J. W. Kennedy and A. C. Wahl created a landmark in scientific history by being first to create an artificial element, using the cyclotron at Berkeley in the United States. It was heavier than uranium, which has the atomic number 92 and at that time came last in the periodic table. The new element, having the atomic number 93, was observed to disintegrate rapidly, emitting beta rays and giving rise to a much more stable element with an atomic number of 94. Uranium was named after the planet Uranus. Since Neptune is further from Earth than Uranus, element 93 was dubbed neptunium and element 94 christened plutonium after the even more remote planet Pluto.
This exploit was followed up by meticulous research aimed at determining the properties of plutonium. The main result of this work resided in confirmation of the fissile nature of one of its isotopes, plutonium 239. Thus a new source of energy lay revealed.

The production and use of plutonium

Plutonium is generated in nuclear reactors. It is, in fact, a by-product of the chain reaction, the underlying principle of nuclear energy. It should be recalled that this chain reaction is sustained by fissile elements (e.g. U-235), which, when bombarded with neutrons under certain conditions, undergo fission, that is to say, they split up and emit large quantities of energy together with other neutrons. Some of these new neutrons subsequently help to maintain the chain reaction by splitting fissile atoms, but others may be absorbed by U-238, the non-fissile isotope of uranium; the result is nothing more nor less than a transmutation into a new substance, the fissile element plutonium.[1] This element may be produced in this way in all nuclear reactors.

Extract from the nuclides chart, showing the conversion of uranium 238 into plutonium 239



Thus, reactors with slow or "thermal" neutrons running on natural uranium (e.g. the gas-cooled graphite-moderated reactors developed in France and Great Britain) or on slightly enriched uranium (such as the American-designed pressurized-water or boiling-water reactors) generate plutonium. The quantities obtained are comparatively small, but make it possible, in the case of reactors operating on slightly enriched uranium, to "recycle" the plutonium, a technique which consists in processing the fuel which has reached the end of its effective life in order to extract the plutonium generated, which is then used for enrichment purposes instead of uranium 235.

Other reactors, using high-energy neutrons, convert uranium 238 into plutonium in a much more efficient manner. These are known as fast breeder reactors, which can be designed in such a way as to produce enough plutonium to keep themselves supplied with fissile material, at the same time leaving a surplus which can be used to make up a core load for another nuclear power plant. This is the property which has gained for reactors of this type the at first glance paradoxical reputation of being able to produce more fuel than they consume.

The advantages of plutonium may be summed up as follows:

(1) It is a fissile element able, like uranium, to furnish about 24,000 kWh per gram;

(2) It may be used as a substitute for uranium 235 in thermal neutron reactors (slightly enriched fuel) or in fast neutron reactors (highly enriched fuel);

(3) It can be produced in fast reactors in greater quantities than it is consumed;

(4) It makes all the natural uranium potentially fissile by reason of the fact that it is formed from U-238 (which makes up 99.3% of natural uranium).

It is therefore not surprising, in view of the above-mentioned qualities, that plutonium should have attracted a great deal of attention. Its use as a fissile material, i.e. as a reactor fuel, however, raises a number of serious problems, some of which are linked with the element's nuclear properties and give rise to difficulties connected with control. It would, however, appear possible to overcome these drawbacks.

1. *Plutonium production*

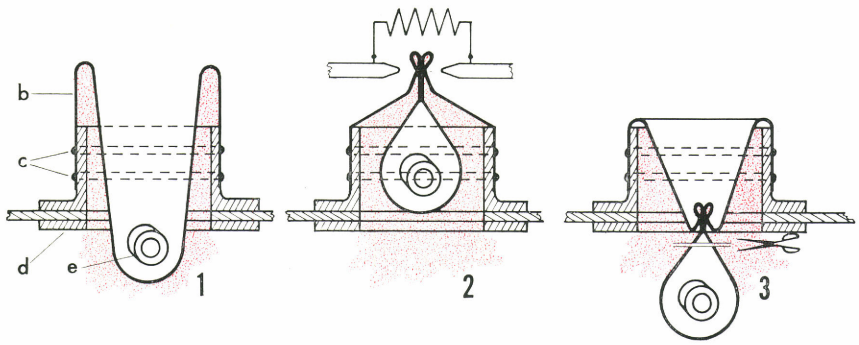
When U-235 undergoes fission it produces fission products, energy (200 MeV) and neutrons (an average of 2.5 neutrons per fission). Some of these neutrons help to maintain the chain reaction, others are absorbed by the various elements in the reactor, and the rest may be absorbed by non-fissile U-238. This U-238 then changes into U-239, which by beta emission gives neptunium 239, which in turn gives plutonium 239, also by beta emission.

This may the expressed in diagrammatic form as follows:

$$U^{235} + N \longrightarrow \text{Fission} \begin{cases} \rightarrow 200 \text{ MeV} \\ \rightarrow 2.5 \text{ N} \\ \rightarrow \text{fission products} \end{cases}$$

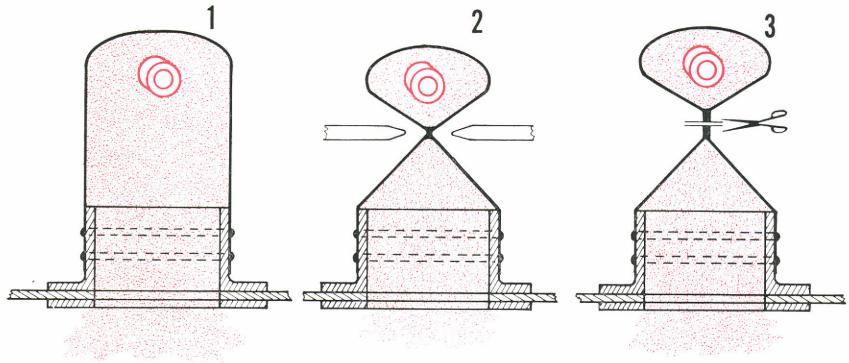
$$U^{238} + N \longrightarrow U^{239} \xrightarrow[23.5 \text{ min.}]{\beta} Np^{239} \xrightarrow[2.33 \text{ days}]{\beta} Pu^{239}$$

Figure 1. Method adopted for introducing objects into and extracting them from glove-boxes
(a) Plexiglass window
(b) Plastic bag
(c) Rubber sealing rings
(d) Plastic bag support
(e) Object



(A) *Introduction of object into glove-box*
—The glove-boxes are fitted with plastic bags (b), which are fixed to supports (d) by means of rubber rings (c) so as to ensure leaktightness.
—In order to introduce an object into a glove-box, the plastic bag is turned inside out in such a way that the object can be placed in the pocket thus formed.
—The tip of the pocket is high-frequency welded.
—The object is withdrawn from the plastic pocket after the latter has been cut open inside the glove-box.

(B) *Withdrawal of object from glove-box*
The process is the reverse of that described above.
—The object is introduced into the plastic bag.
—The bag is welded and cut open along the weld. The glove-box has thus remained leaktight, while the object remains enclosed in the plastic bag.



Problems involved in the processing of plutonium compounds

The preparation of fuel elements containing plutonium also gives rise to difficulties, the greatest of which are due to the fact that it emits very high energy alpha rays.
Plutonium is in fact extremely harmful from a biological standpoint. It lodges in the bone marrow, where it rapidly destroys the hematopoietic cells, where the red blood corpuscles are formed, by emitting α particles. The result is that the air, in order to be normally breathable, must not contain more than the infinitesimally small proportion of a hundred-thousandth part of a millionth of a gram per cubic metre.
This requirement seriously complicates the utilization of plutonium. Because of this toxicity, all work has to be performed in leak-tight boxes, equipped generally with plexiglass windows. Manipulations are performed from outside by means of long neoprene gloves fitted to these boxes, in which the pressure must be maintained below atmospheric pressures, so that in the event of a leak it is always the laboratory air which penetrates into the glove-box and not the glove-box air which escapes into the laboratory.

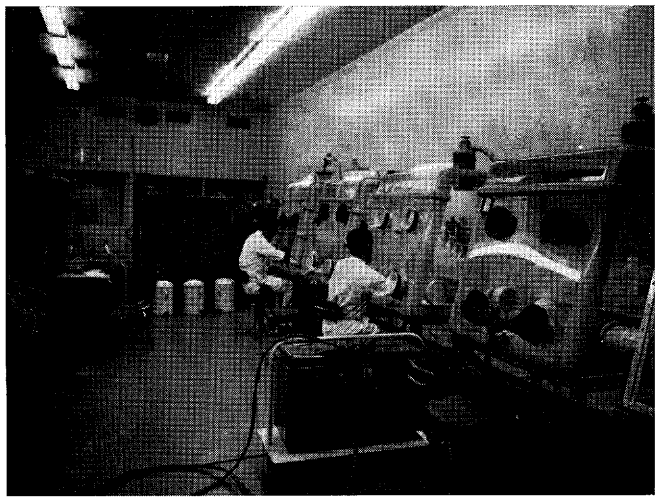
In order to be used as fuel, plutonium has to be converted into a form which will be affected to a minimum by the radiations of extremely high density and power to which the material is subjected in the reactor. The chemical compounds of plutonium employed frequently react with air, so that in most cases the gloveboxes are filled with inert gases (e.g. nitrogen, argon or helium) which must be maintained in a state of high purity. This requires large-capacity gas-purification plants.
As a general rule, an operation, a stage in a fabrication chain, is carried out in a glove-box. These boxes are interconnected by airlocks, in order to facilitate transfers of material between them. When a transfer has to be effected between two unconnected boxes, the operation is performed by means of plastic bags of the type shown in Fig. 1. Meticulous precautions have to be taken to avoid anything in the way of a fire or the slightest violent reaction which might impair the leak-tightness of the glove-boxes. In particular, the plutonium mass processed in one batch must not be allowed to exceed a certain limit value (critical mass), otherwise a chain reaction might be triggered off, jeopardizing the personnel as well as the installations. To preclude the possibility of such an accident, either the volume of the

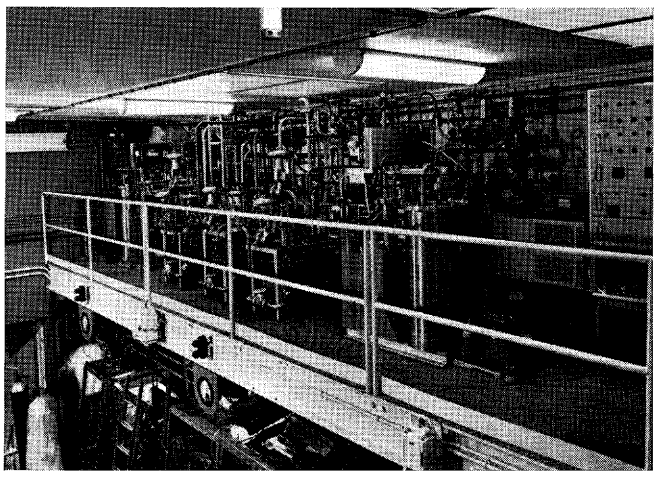
Above:

The glove-boxes in the plutonium analysis laboratory at Fontenay-aux-Roses (photograph by courtesy of P. Jahan - French Atomic Energy Commission)

Below:

Argon purification apparatus in the Plutonium Laboratory at Mol, which is operated jointly by BelgoNucléaire and the CEN





apparatus is designed to accommodate less than half the critical mass, or the plutonium masses handled are restricted and checked at all times to prevent half this mass from being exceeded[2]. It is for this reason that numerous automatic controls are provided to give warning to the personnel of any kind of perturbation, however slight, in the normal condition of the premises or equipment or in the operational processes. Although these controls ensure complete working safety, strict discipline must be observed by the personnel.

Types of plutonium-enriched fuel utilizable in nuclear reactors

We have already said that nuclear fuels must be put into a form which will withstand the special conditions to which they are exposed in reactors. Thus plutonium metal as it stands cannot be used as fuel, for its structure changes markedly with increases in temperature. It is therefore necessary to employ alloys, for instance, or other chemical compounds.

The most common forms which might be applied in reactors are shown in Fig. 2.

These materials all offer particular advantages, and have to be selected according to the reactor type envisaged. Below, in a very condensed form, are set out the characteristics of each of these classes of fuel.

(1) *Alloys:* these materials are worthy of consideration

2. In aqueous solution, plutonium's minimum critical mass is about 500 g. In the solid phase, it is much greater (several kg). The critical mass depends fundamentally on the nature of the plutonium compound and on the medium.

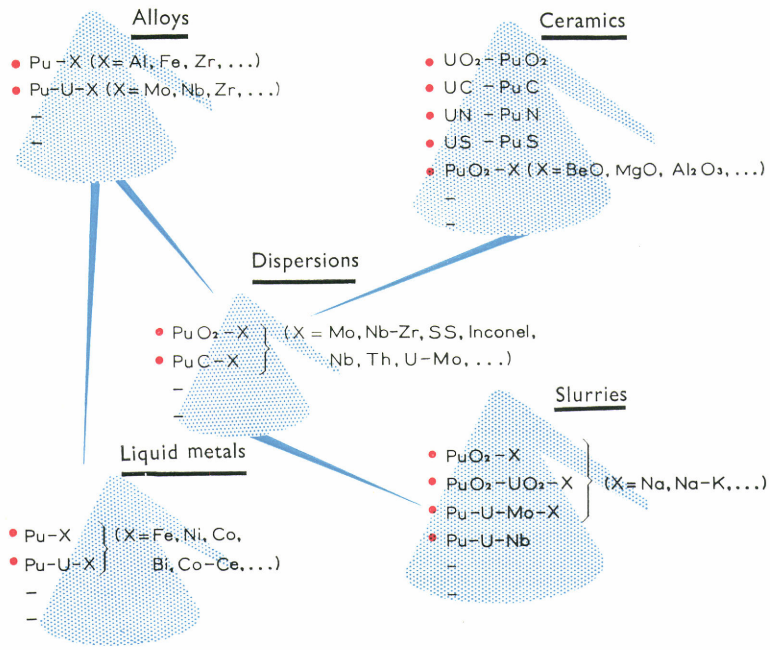


Figure 2
Chart showing plutonium-containing fuels suitable for use in reactors

because of their high density and their satisfactory thermal conductivity. Their indifferent irradiation behaviour, however, may restrict their use; the present trend is to design cladding strong enough to contain fuel swelling induced by irradiation.

(2) *Ceramics:* the chief advantage of these compounds is their irradiation stability; the most thoroughly explored ceramics at present are the oxides. Work on carbides was initiated quite recently and the results appear promising.

(3) *Dispersions:* these fuels have the merit of combining the desirable properties of both alloys and ceramics. Their fabrication cost, however, is still very high, which has the effect of restricting their fields of application.

(4) *Molten alloys and slurries* (high content suspensions of solid particles in a liquid metal): in the liquid or slurry form, fuels may lend themselves to continuous purification, at least with regard to part of the fission products which they contain. They therefore constitute a highly attractive possibility. The technology of these materials is still, however, in its infancy.

Fabrication of plutonium-enriched nuclear fuels

For the safety reasons pointed out above, the processing of plutonium compounds is a very costly business. It is accordingly advisable to reduce the number of stages to a minimum and often to introduce the plutonium into the fabrication system at the latest possible stage. This applies in particular to slightly enriched fuel such as that intended for use in American-type thermal neutron reactors which, for the same power level, require much larger amounts of fuel than fast reactors.

Ceramic fuels

Within the compass of this article, we cannot deal at length with the multifarious methods employed for fabricating the various types of fuel referred to in the previous section. However, let us take a case in point, namely that of ceramic fuels of the uranium and plutonium oxide (UO_2-PuO_2) type. Fig. 3 depicts a typical fabrication system using the vibration-compacting technique. At the first stage, the plutonium salts are converted into the pure oxide. This is then mixed with uranium oxide in order to enrich the latter in fissile material at the time of its insertion into the cladding[3]. This process enables the plutonium to be introduced into the fabrication system "at the last moment", which is definitely advantageous from the economic standpoint. It is, in fact, only at this stage that considerable amounts of plutonium-containing fuel have to be handled in the glove-boxes described briefly above.

Immediately after the oxides are compacted by vibration, the cladding tubes are sealed, decontaminated by ultrasonic treatment and by pickling, and tested for leaktightness. If they betray no detectable leakage, they can be handled in the open air for the purpose of carrying out various non-destructive tests (density, homogeneity, plutonium concentration, cladding-tube integrity, welds, etc.). The flawless tubes are grouped into bundles. For this

3. When thus mixed, these powders are compacted in the cladding tube by vibration. We shall revert to this technique later.

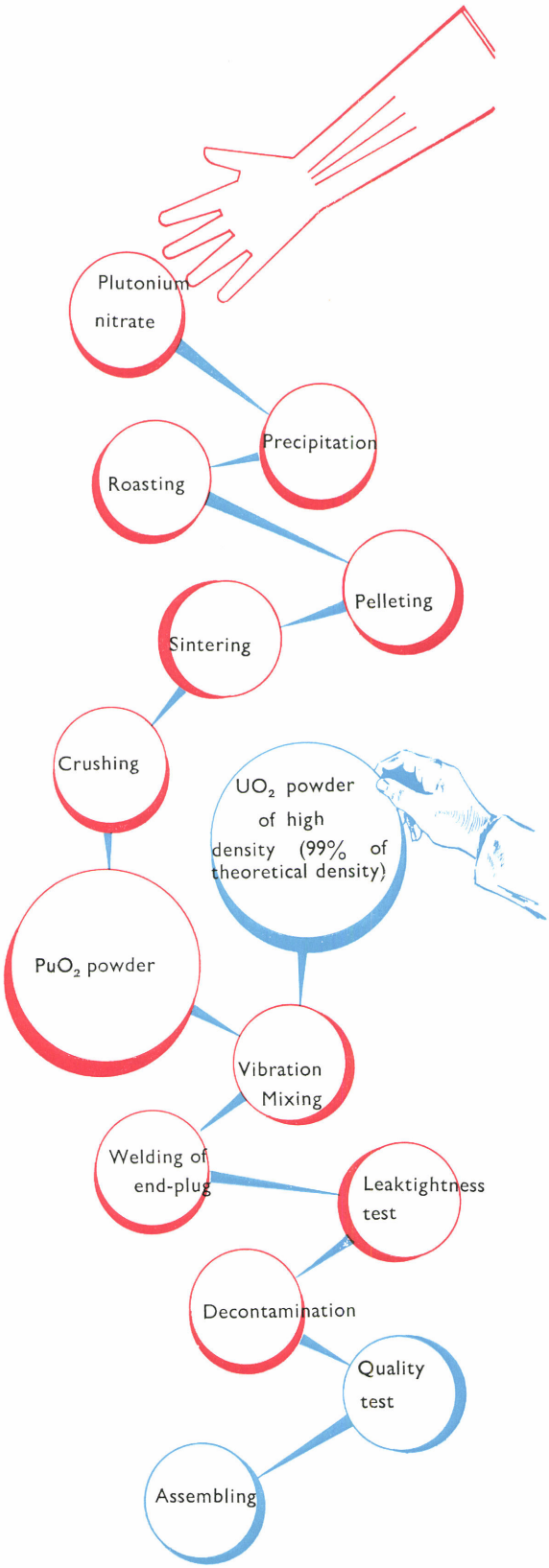


Figure 3.

Fuel-element fabrication system by vibration process
Fuel: UO_2 – PuO_2

Starting material

The plutonium oxalate is prepared by precipitation from the nitric solution.

The plutonium oxalate is roasted in air at 550°C. This yields a plutonium oxide very near to PuO_2.

The plutonium oxide is compressed into pellets of a density equal to about 50% of its theoretical density.

The pellets are heated at 1,500°C in an argon-hydrogen 5% mixture (non-explosive with oxygen). After this heat treatment, the density of the pellets is 97% of the theoretical density of the oxide.

The pellets are crushed into grains of less than 50 microns.

The PuO_2 powder is mixed with the UO_2 powder and compacted by vibration in the cladding tubes. The mixing and compacting processes are simultaneous. The final density after compacting ranges from 88% to 92 % of the theoretical density of the oxides, depending on the type of powder used and the characteristics of the cladding tubes selected.

The welding of the plugs is generally effected by the arc process, the tubes having first been filled with helium.

A helium-leak detection test is carried out with a high-sensitivity mass spectrograph.

The surface of the tubes is decontaminated by ultrasonic treatment and pickling.

The tubes are tested for weld quality, filling homogeneity and plutonium distribution.

The tubes are grouped into bundles, each of which forms a fuel element.

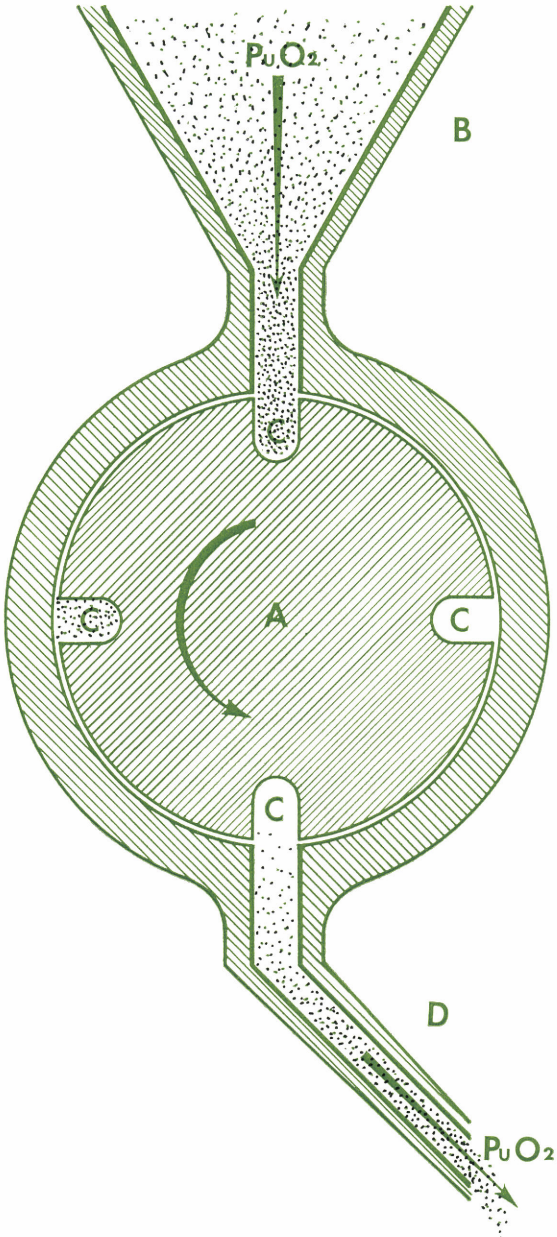


Figure 4
Diagram of a feed control valve

purpose, it is frequently the practice to resort to mechanical assembly, which makes it possible to avoid any delicate weld liable to impair the tubes and result in contamination of the plant by plutonium.

Vibration-compacting technique

The vibration process has to a large extent been developed with a particular view to plutonium-containing ceramic fuels. There are several reasons for this, i.e.:

(1) As we have seen, this method makes possible separate preparation of the plutonium oxide, which is mixed with uranium oxide at the moment when the ceramics are inserted into the tube. For this purpose, use is made of feed control valves of various types, like that shown in Fig. 4.
The plutonium oxide (PuO_2) is placed in the funnel B. The drum A contains calibrated wells C. This drum rotates at a speed depending on the type of enrichment desired. The calibrated wells are filled with powder and discharge their contents into the cladding tubes through the passage D. Thus the filling for PuO_2 and UO_2 is effected simultaneously with valves based on the same principle but differing in capacity.

(2) Vibration enables high final densities to be obtained, i.e. 90-92% of the theoretical density of the oxide. For this purpose, the starting powders must be of very high density in order to reduce the density loss due to the internal porosity of the powder grains. Moreover, the powder is generally graded in several well-defined grain-size fractions, the dimensions of which depend mainly on the diameter of the tubes. The actual vibration is carried out in accordance with a very specific frequency variation, advantage being taken of the resonance frequencies of the material[4].

(3) The vibration process lends itself perfectly to remote-control techniques, which is particularly useful for plutonium. It only needs to be remembered, in fact, that plutonium is formed in nuclear reactors and therefore may be accompanied, in quantities varying with the purification process adopted, by fission products emitting radiations which possess a high power of penetration and are consequently intolerable to the human organism.

4. When the tube is vibrated according to a resonance frequency, the vibration is stepped up to a very high degree. These frequencies are a function of the composite modulus of elasticity of the cladding and the powder they contain and also of the length of the tubes.

Other compacting methods

Vibration does not invariably produce a density which satisfies the requirements of nuclear power plant constructors. Furthermore, the in-pile experiments carried out on fuels fabricated by varying methods have not yet yielded a sufficiently comprehensive body of information for any of these methods to be pronounced the ideal one for the purpose. It has therefore proved necessary to give consideration to other fabrication techniques, including the conventional process of pelleting and sintering, which consists in compressing the oxide powders into small cylinders and subjecting them to thermal treatment (sintering), so as to step their density up to a value quite close to the theoretical density (95%-97%) by altering the crystalline structure. This method is, however, expensive for plutonium compounds, as it involves the simultaneous manipulation in glove-boxes of large amounts of uranium oxide and plutonium oxide in the very first stages of the fabrication process. (For example, a 2%-enriched fuel for a power plant operating on a 100 MWe thermal neutron reactor may require the use of around 40 tons of UO_2 and 0.8 tons of PuO_2.)

Still other methods may be adopted, such as swaging, which may be described as the hammering of a tube filled with ceramic powder. By this means it is possible to achieve very high densities—very nearly as high as those obtained by sintering. Here too, however, the cost is extremely heavy on account of the very strict checks which the swaged tubes have to undergo. These checks are necessitated by the considerable mechanical stresses which are set up during the swaging process and which may cause both an external and an internal deterioration of the tubes.

Conclusions

As we have seen, the use of plutonium entails a variety of problems for the solution of which far-reaching research projects are under way in the United States, the United Kingdom, the Soviet Union and the European Community. In the United States, this research has reached the point at which a special testing reactor using thermal neutrons (the PRTR - Plutonium Recycle Testing Reactor) has for the first time been loaded with plutonium-enriched fuel elements. The endeavours made in this direction are amply vindicated by the fact that plutonium is a fissile element which is produced by reactors from the non-fissile uranium 238. Recourse to plutonium thus opens up the possibility of deriving the maximum advantage from the power potential at the disposal of Mankind.

A few dates in the History of Plutonium

JANUARY 1941 *G. T. Seaborg, E. M. McMillan, J. W. Kennedy and A. C. Wahl produced an artificial element heavier than uranium by bombarding U_3O_8 with deuterons in the cyclotron at Berkeley, USA. This element had the atomic number 93. They observed that its half-life was very short, being hardly over 2 days, and that by β-particle emission it was transformed into a radioactive element emitting α-particles of very high energy with the atomic number 94. The 93 element is known as neptunium, while the 94 element was given the name plutonium.*

AUGUST 1942 *The first few milligrams of a pure plutonium compound, in the form of the oxide, were prepared in the Metallurgical Laboratory of the University of Chicago.*

NOVEMBER 1943 *The metal in the pure state was finally isolated with certainty by H. L. Baumbach and S. Fried in the same laboratory. For military reasons, production of plutonium in kilogram quantities was rapidly built up. During the next few years research work on plutonium was also undertaken in Canada, Britain and the USSR.*

LATE 1948 *100 milligrams of plutonium metal were obtained at Chalk River, Canada.*

1950 *Gram quantities of plutonium were produced at Harwell, England.*

1955 *On the occasion of the first international conference on the peaceful uses of atomic energy, held at Geneva, American, British, Canadian and Russian announcements revealed the paramount importance of plutonium as a fuel for nuclear reactors.*

JANUARY 1956 *France produced its first few grams of plutonium metal. Since then, the French Atomic Energy Commission (CEA) has been engaged on large-scale research projects on plutonium with a view both to its recycling in thermal reactors and its use in fast reactors.*

1957 *In Belgium, a laboratory for the handling of plutonium containing compounds was set up jointly by private industry and the Nuclear Study Centre (CEN).*

1960 *Under the Euratom/United States joint research programme, Euratom concluded two contracts, one of which was with the French Atomic Energy Commission (CEA) and the other with the BelgoNucléaire/CEN association. The purpose of both contracts is to study the recycling of plutonium in thermal reactors.*

1962 *Euratom concluded a contract of association with the French Atomic Energy Commission (CEA) for the construction and operation of the "Rapsodie" fast-neutron reactor and for allied research. Part of this research is aimed at the development of the plutonium based fuel which it is intended to burn in this reactor.*

terminologie terminologia terminology

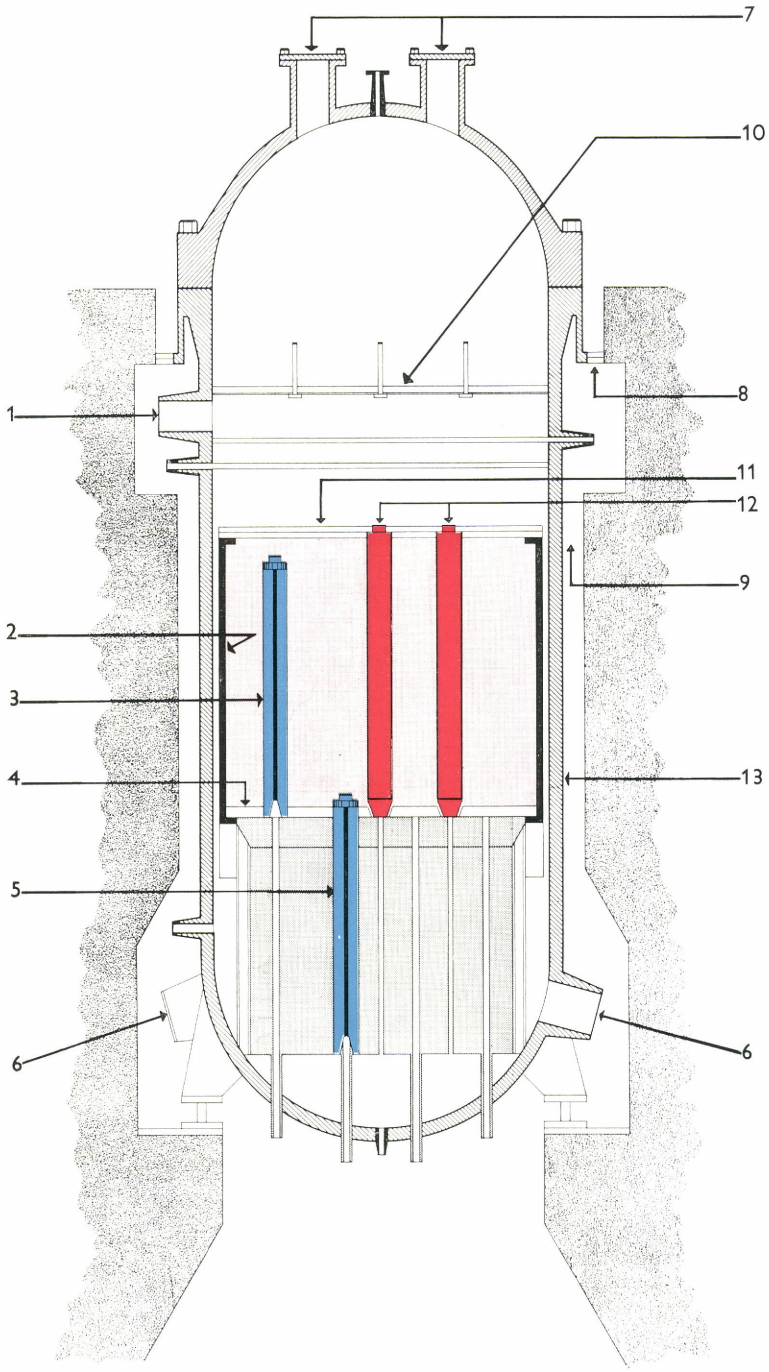
Schnitt durch das Gefäß eines Siedewasserreaktors

Schéma d'une cuve de réacteur à eau bouillante

Schema del contenitore di un reattore ad acqua bollente

Vat van een kokend-waterreactor

Diagram of boiling water reactor vessel



1. Austritt des Dampf-Wasser-Gemisches
   sortie mélange eau/vapeur
   uscita miscela acqua/vapore
   uitlaat water/dampmengsel
   steam/water outlet

2. Wärmeschild
   bouclier thermique
   schermo termico
   warmtescherm
   thermal shield

3. Regelstab (eingefahren)
   barre de réglage (introduite)
   barra di controllo in posizione inserita
   regelstaaf in ingeschoven positie
   control rod in inserted position

4. unteres Stützgitter
   grille support inférieure
   griglia inferiore di sostegno
   onderste ondersteuningsrooster
   lower support grid

5. Regelstab (ausgefahren)
   barre de réglage (retirée)
   barra di controllo in posizione estratta
   regelstaaf in uitgeschoven positie
   control rod in withdrawn position

6. Speisewassereintritt
   admission de l'eau d'alimentation
   entrata acqua di alimento
   inlaat voedingswater
   feed water inlet

7. Beschickungsöffnungen
   portes de chargement
   portelli per la ricarica
   laadluiken
   loading ports

8. Wasserdichtung
   étanchéité d'eau
   tenuta d'acqua
   waterdichting
   water seal

9. Zwischenraum für Ventilationsluft
   espace rempli d'air de ventilation
   intercapedine con aria di ventilazione
   ruimte gevuld met ventilatielucht
   ventilation air space

10. Deflektor
    déflecteur
    deflettore
    deflector
    deflector

11. oberes Führungsgitter
    grille supérieure de guidage
    griglia superiore di guida
    bovenste geleidingsrooster
    upper guide grid

12. Brennstoffelemente
    éléments de combustible
    elementi di combustibile
    splijtstofelementen
    fuel elements

13. Gefäßwandung
    paroi de la cuve
    parete del contenitore
    wand van het vat
    vessel wall

EURATOM NEWS

International Euratom symposium

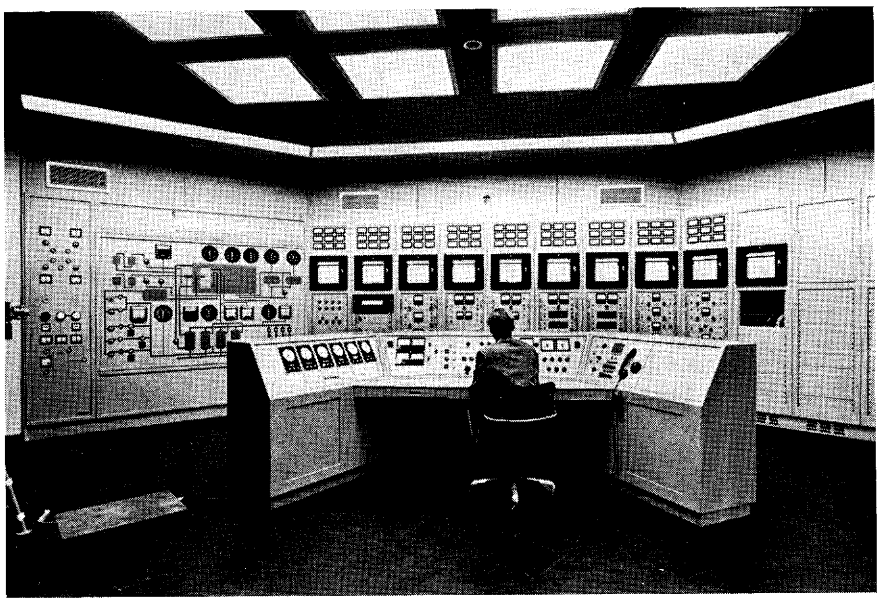
on the radiological protection of workers

A "Symposium on the radioactive contamination of workers", organized by Euratom, was held in Munich from 24 to 26 October 1962. By contamination is meant the pollution of the human body and other objects by radioactive substances; they can either act as external irradiation sources or irradiate the body internally as the result of uptake in food, by inhalation or through the skin.

The symposium was attended by about 250 experts from the countries of the European Community, Great Britain, Canada, Austria, Switzerland, Scandinavia, Spain and the United States. 21 papers were read by participants from Europe and America on the various problems relating to protection against radioactive contamination.

The international symposium in Munich was devoted to measurement problems and contamination detection, physical and medical supervision of nuclear workers, measures to be taken in hospitals, laboratories, industrial firms, mines and particularly in nuclear power plants, and the extent to which contamination presents no health hazard and can therefore be regarded as permissible.

Petten (Netherlands) Nuclear Research Centre. Control room of the High Flux Reactor.



Petten reactor

transferred to Euratom

The Petten (Holland) high flux reactor was transferred to Euratom on October 31, 1962. This marks the setting up of the fourth establishment of the Euratom Joint Research Centre.

The agreement covering this transfer was concluded between the Dutch Government and Euratom on 25 July 1961 and ratified by the Dutch States-General on 24 July 1962. This convention came into force with the signature of the acts of execution which included, among other things, the conditions covering the technical operation of the HFR test reactor, which will be the responsibility of the staff of the RCN (Reactor Centrum Nederland) during a four year period, as well as the conditions on the use of the reactor for the RCN's own programme.

Euratom plans to construct in the near future the necessary buildings and installations. During the next five years $ 10 million will be spent on Petten

EURATOM NEWS

in addition to the investments already undertaken by the RCN, estimated at $ 7 million, in addition to a Euratom contribution of about $ 1 million. Altogether the research staff will amount to 350.

The research programme at Petten will include the optimum use of the HFR for irradiation experiments, the technical co-ordination of research on gas reactors (in particular "Dragon" and the "pebble-bed" reactor at Jülich) as well as experiments on liquid fuels. The Petten establishment will, after Ispra, be the Community's second "general competence" research centre.

USAEC signs two contracts with Euratom Supply Agency

On 27 November, 1962, two contracts within the framework of the US/Euratom Agreement were signed between the US Atomic Energy Commission and Euratom's Supply Agency. A third was signed between Euratom and the Italian SENN (Società Elettronucleare Nazionale) concern. They are in conformity with the Euratom Treaty, which lays down the principle that the Agency shall have the exclusive right to conclude contracts for the supply of special fissile materials produced in or imported into the Community. The Agency also has the right of option on special fissile materials produced in the Community.

The first contract concerns the supply by the Commission to the Agency of up to 4,000 kg. of contained U_{235} uranium for the SENN company's power reactor, now under construction on the Garigliano river, 60 km. from Naples. The contract makes provision, among other things, for a system of deferred payments over a 10-year period for the operating inventory of the reactor.

The second contract covers the return to the USAEC of and the credit for unconsumed U_{235} contained in US-supplied fuel irradiated in the SENN reactor, and the purchase by the USAEC of up to 600 kg. of plutonium produced during a 10-year period.

The third contract (between Euratom and SENN) covers the reciprocal rights and obligations of the two parties concerning the supply by the Agency to the SENN company of the enriched uranium obtained from the USAEC, the return by SENN of unconsumed U_{235} in irradiated fuel elements and the purchase of up to 600 kg. of plutonium produced therein for disposal by the Agency. These provisions will ensure that the SENN company can benefit from the assured market offered by the USAEC for these materials.

Euratom Commission meeting with fuel element manufacturers

On 19 December 1962 at Brussels the Euratom Commission met with around 50 fuel element manufacturers and nuclear power plant operators. The aim was to survey the conditions under which fuel elements for reactors in operation or under construction could be made on an industrial scale in Europe, thus contributing to the development of a European industry specializing in this field.

Euratom's first direct action in favour of the fuel element fabrication industry was the decision to aid power reactor projects. The contracts concluded or to be concluded with the SENN, SIMEA and SENA projects represent a total outlay by the Community on the manufacture of fuel elements of $17 to 20 million. The market thus taking shape could, with the prospect of a rapidly developing power plant construction programme, attain a rate of expansion rising from $ 8 million overall turnover in the initial phase to $ 24 million in 1970 and $ 80 million by 1975.

The Commission's representatives emphasized that, apart from its industrial activities, Euratom is providing substantial aid for research and development, notably under the second five-year research programme, as well as looking after the supply of enriched uranium.

The prospects thus outlined and the measures taken by the Community gave rise to an extremely lively discussion among the industrialists present, especially as regards the possibilities for testing parts of fuel elements in Community high-flux reactors and the testing of prototype fuel elements in power reactors. Such tests would enable industrial concerns to acquire wider and more varied experience in this sector. In addition, a thorough examination was carried out on the technical and economic problems raised by the change-over from fuel-element charges fabricated in the United States or the United Kingdom to charges manufactured in the Community.

The interest shown at the meeting resulted in the adoption of the Commission's proposal that a further meeting should be arranged to discuss further these various problems.