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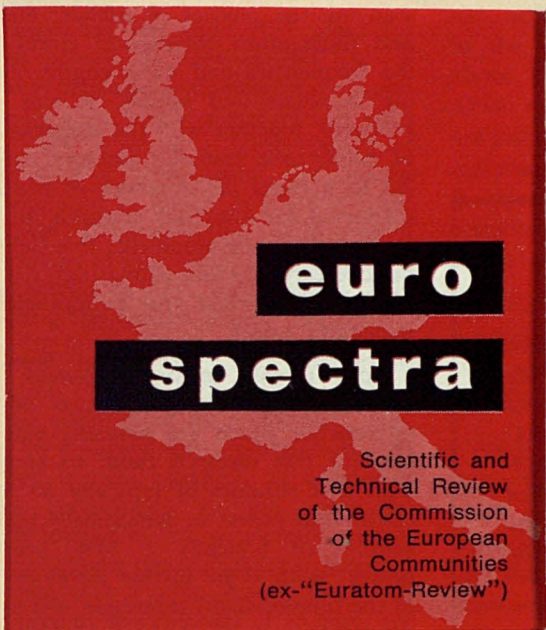
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It would seem that it is neither possible nor desirable to do without petroleum nowadays. The recent crisis has vividly emphasised the extent to which we depend on supplies and how difficult it can be to ensure a continuous flow of oil to meet our requirements. At the same time a number of alternative methods for producing energy are being proposed and several voices have been raised as to their inherent advantages and risks. Assessments of the dangers involved, in particular, are of varying accuracy, and an article on the subject is to appear in our next number. The main point at issue is whether the countries of Europe, who are heavy and committed consumers of oil, could not perhaps indulge in some horsetrading until the supply situation has eased and other sources of energy have been found.

One of our contributors, who will already be familiar to readers on account of two articles he wrote in 1970 and 1971 on hydrogen as a source of energy, maintains that we could alleviate the situation by making better and more immediate use of some of the Community's natural resources. He puts forward his argument in an article on page 107. The topicality and originality of his proposals will undoubtedly provoke a response on the part of our readers, who are therefore invited to let the editor have any comments they may wish to make on the subject.

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# The SORA project at Ispra

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RUDOLF HAAS, ROLF MISENTA

*In February 1973 the Council of Ministers of the European Communities adopted a four-year programme for the Joint Research Centre. As it no longer included the development of a pulsed reactor, this programme consequently put an end to several years' work by the JRC on the SORA pulsed reactor. Before the teams of engineers and physicists are assigned to other work, it was considered essential to finalize the technical file concerning this project. A full EUR report on the experimental potential offered by SORA is now being printed and the file of the reactor design has been updated.*

*It is my opinion that the documents dealing with the experimental part and with the design itself would be of immense value to scientists in the Community who continue to investigate the problem of intense neutron sources. Hence I thought it would be a good idea to ask two physicists, one of whom worked on the reactor design and the other on the design of the experimental devices, to give, in this issue of euro-spectra, a rundown of the work carried out by the various teams over the past years.*



S. FINZI  
Scientific Director  
of the Ispra Establishment

## INTRODUCTION

### The Beginning

Neutrons are an ideal probe for studying the physics of matter; energy and momentum correspond closely to the frequencies and wave-lengths existing in solids and liquids. It was this property, together with the construction of beam tube research reactors, which led to a rapid development of neutron scattering for materials research. In the course of this development, this field of basic research became increasingly oriented towards applied and technologically important topics.

Very intense neutron sources are needed today. There are two design approaches which are basically equivalent for this research field: to employ high flux reactors of typically 50 MW continuous power or periodically pulsed sources of typically 1 MW average power. With some further development of the time-of-flight technique, the latter approach is particularly promising, as has been demonstrated at Dubna (1).

The publication on the successful operation of the first repetitively pulsed reactor, the *IBR* at Dubna (*USSR*), at a mean power of originally 1 kW (2), immediately stimulated wide interest at Ispra in the development of a higher power system.

Since 1962 studies on the design of *SORA*<sup>1</sup> and its research applications have been performed at Ispra. The original feasibility studies with Auxiatom, Belgonucléaire and Siemens-Schuckert-Werke showed a practical power level to be around 0.5 MW, for an extremely compact, liquid metal cooled core.

The reactor was intended for construction within the third five year programme of the Joint Research Centre (*JRC*) (to start in 1968), its main application being to solid state physics research, thus ensuring the continuity of the basic *JRC* research programme. Publications both on design (3) and use (4) were given at various conferences and as a result the *SORA* Project obtained international recognition (5).

RUDOLF HAAS, ROLF MISENTA - *Joint Research Centre* of the Commission of the European Communities, Ispra Establishment

<sup>1</sup> *SORA* : *SO*rgente *RA*pida = Fast Source

### Alternative Pulsed Neutron Sources

A pulsed reactor represents one type of repetitively pulsed neutron source. There are others: linear electron accelerators may produce neutrons at a rate of somewhat less than  $10^{15}$  n/sec for target powers as high as 200 kW. A recently proposed spallation source (operated by a proton accelerator) will yield  $5 \times 10^{15}$  n/sec at a target power of 25 kW. For comparison, the reactor leakage rate is about  $5 \times 10^{16}$  n/sec per MW. It appears that—with present technology at least—very intense neutron sources can be built economically only by using the fission reactor concept.

There is, however, an important variation of the pulsed reactor obtained by the addition of an accelerator, possibly as a subsequent modification. The reactor is then operated as a booster (with periodically varying reactivity) which intensifies the accelerator pulses. In this scheme the high reactor power is matched with a quite short pulse width of 7  $\mu$ sec or less (instead of 75  $\mu$ sec; see Table I). This system is successfully operated at Dubna (6). However, owing to the experimenters' requirements, the plant is operated part time in the "pulsed reactor mode" and part time in the "booster mode".

### The Comparison

It has become evident that the pulsed reactor (of typically 1 MW mean power and 50 pulses per second) represents a new type of research tool, which according to its performance characteristics ranges between the following three types of neutron source:

– *The Single Burst Reactor* produces one very intense neutron burst per hour. Even though the pulses from a repetitively pulsed reactor are considerably smaller, its average neutron output (or, in other words, the information obtained per unit of time) is about 1 000 times larger than that from a single burst reactor.

Therefore the pulsed reactor, especially if operated at low pulse frequencies, can enter the field of technological irradiation experiments and solid state physics of samples under extreme (pulsed) conditions (such as pressure, magnetic fields, etc.). This research field—apparently due to the lack of proper neutron sources—is not developed in Europe.

– *The High Intensity Pulsed Accelerator* produces a mean neutron output a few hundred times inferior to the Pulsed Reactor, the pulse width of the accelerator being a few microseconds or less, depending on the application. Owing to the relatively large pulse width of about 75  $\mu$ sec of the pulsed reactor which results in a poor resolution, there are only few applications in the epithermal energy range where the pulsed reactor is superior. However, in this range the accelerator pulsed reactor presents considerable advantages.

– *The Continuous High Flux Reactor* produces a thermal flux of about  $10^{15}$  n/cm<sup>2</sup>/sec at a power of about 50 MW.

Since the pulsed reactor *SORA* produces the same peak flux (see Table I), both instruments are compatible for typical solid state physics experiments in the domain of elastic and inelastic neutron scattering. This is true even without consideration of the specific advantages offered by the low background of pulsed reactors.

### The SORA Programme

The Dubna *IBR* was brought step by step to about 30 kW mean power, with pulse frequencies of 100–0.2 sec<sup>-1</sup>; it competed well with the available research reactors. Finally Dubna proceeded to the construction of a 4 MW pulsed reactor (7).

The *JRC Ispra* made also a big effort to update the *SORA* Project. A design contract was passed in 1971 to an international industrial consortium (Interatom, Belgonucléaire, Neratom, FIAT, PMN) in order to obtain a "fixed price offer" in mid-1972. Theoretical and experimental work in support of the design work, above all physics and safety studies, were conducted at the *JRC*.

The design contract was supplemented by an intense effort to define an experimental programme for *SORA*, in the fields of elastic and inelastic neutron scattering, nuclear physics and neutron data measurements and radiation damage. Many other research applications could be added to this programme, its importance in today's physics' and technology's applications being well established.

Table I

	Mark 1	Mark 2	Mark 3
<i>Reactor Core</i>			
Operation mode	reactor	reactor	booster
Mean power [MW]	1	3	1 ÷ 3
Pulse frequency [sec <sup>-1</sup> ]	50	5 ÷ 50 <sup>+</sup>	5 ÷ 50
Peak power [MW]	200	6 000	2 000
Power pulse width [ $\mu$ sec]	75	75	7
Peak/Background	1 200	35 000*	35 000*
<i>Water Moderator</i> (5 cm thick) leakage fluxes:			
<i>Thermal energies:</i>			
Average flux: [n/cm <sup>2</sup> /sec]	$4.5 \times 10^{12}$	$1.3 \times 10^{13}$	$4.5 \times 10^{12}$
Peak flux: [n/cm <sup>2</sup> /sec]	$0.5 \times 10^{15}$	$2.7 \times 10^{16}$	$2.5 \times 10^{15}$
Pulse width ( <i>FWHM</i> ) [ $\mu$ sec]	90	90	30
<i>Epithermal energies:</i>			
Peak flux: [n/cm <sup>2</sup> /sec]	$1.4 \times 10^{14} \frac{\Delta E}{E}$	$4.2 \times 10^{15} \frac{\Delta E}{E}$	$1.3 \times 10^{15} \frac{\Delta E}{E}$
Pulse width ( <i>FWHM</i> ) [ $\mu$ sec]	75	75	7

<sup>+</sup> the underlined value is consistent with the set of data.

\* for Pu core, for U-core the value would be 12 000.

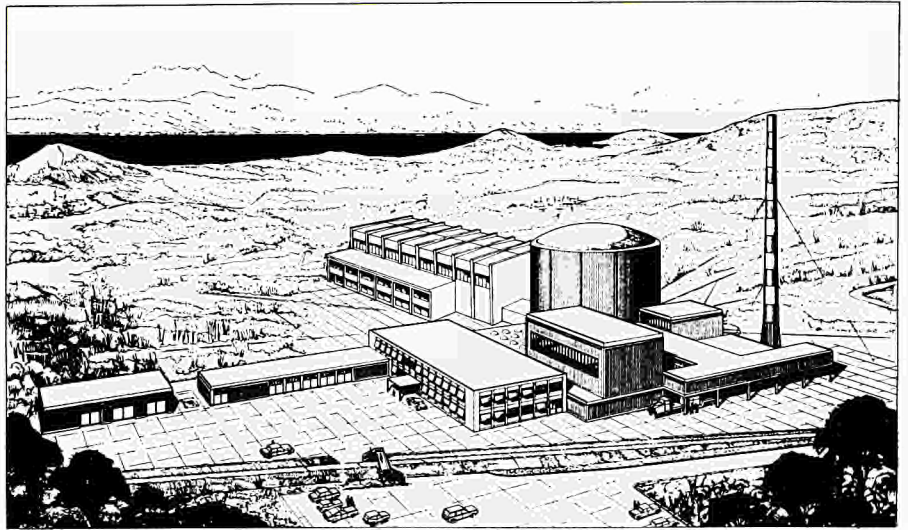


Fig. 1: Artists' view of the SORA plant.

An artist's view of the siting of *SORA* at the north-east corner of the Centre is shown in Fig. 1.

The *SORA* plant is conceived as part of an international institution, which assures access to the reactor to all interested countries and institutions. In this model the *JRC Ispra* operates the whole plant, including the experiments, and contributes its general infrastructure, which is particularly well adapted to support multinational teams.

#### The Flexibility

Since 1966 the construction decision has been repeatedly put off, and the performance characteristics required of *SORA* have been steadily increased. The project has coped with this situation by making provision as far as possible for extrapolation of the *SORA* plant to later versions. In this way construction could proceed without delay and the whole project would always remain based on available experience and technology. *SORA* Mark 1 is the immediate construction goal; the Mark 2 and Mark 3 versions are already included into the overall planning. The three versions are described in Table I.

Depending on the experimental requirements, either Mark 2 or Mark 3 or a combined operating schedule would represent the final stage: Mark 2 with 3 MW and 5 pulses/sec would be a very

powerful irradiation instrument, while Mark 3 with a probable 1 MW, 50 pulses/sec and an accelerator for shorter pulses would best satisfy the demands for thermal-epithermal time-of-flight measurements.

## THE SORA DESIGN

### General

*SORA* is a very compact sodium cooled fast reactor with specific pulsed reactor features. Therefore the following description will pay particular attention to these features. A collection of characteristic *SORA* data is presented in Tables I and II.

The physics design of *SORA* is based on computer codes which were checked against the results of the critical experiment (9). This experiment was performed at Oak Ridge Nat. Lab. (USA) in 1965-66 under a Memorandum of Understanding between the Euratom Commission and the United States Atomic Energy Commission (USAEC). Fig. 2 shows the physics mock-up in the Oak Ridge critical facility hall.

The design work was directed towards the Mark 1 goal. However, built-in power limitations were avoided and the pulsation device was designed to be capable of the variable frequency option and to reach the Mark 2 stage. The new core would allow a power density of 300 kW/liter. The fuel performance under pulsed conditions could be tested already in Mark 1.



A sufficiently redundant design of the cooling loop system ensures that the radioactive decay heat is taken care of in any operating condition. In general the procedures to ensure safe design are the same as for any compact fast reactor.

As far as possible provision was also made for later incorporation of an accelerator, which would be located outside the reactor building between beam channels C1 and C2 (Fig. 3), the length being 20-200 m depending on the type of machine. The ducts inside the reactor building required for the transfer of the beam to the reactor core are provided in the building design. No means of studying the target design at this stage of the project were provided.

### Plant Layout

The plant consists of the reactor containment and the ancillary complex, which is concentrated in a small sector. The remaining space is at the disposal of the reactor users.

The plant is oriented and positioned inside the *JRC* (see Fig. 3) to allow the use of beam channels up to about 900 m in length. The beam channels cover an angular range of  $230^\circ$ . The neutron beams from all horizontal beam channels can be led to the area outside the reactor containment, where several measuring stations and experimental halls are planned.

The reactor building is a gas-tight cylindrical steel shell. It houses the reactor block with all primary circuits. There are also the fuel handling machines and other reactor support equipment stationed. The space on the main floor is largely reserved for experimental equipment.

The reactor ancillary complex houses essentially:

- the technological block for secondary and tertiary core circuits, ventilation and workshops;
- the handling block for installations pertaining to handling, washing, storing and shipping of fuel and other active materials;
- the operation block for control room and associated equipment, health physics, decontamination laboratories and offices;
- the electrical block for the various power supplies and emergency units.

### Principle of Pulsed Reactor Operation

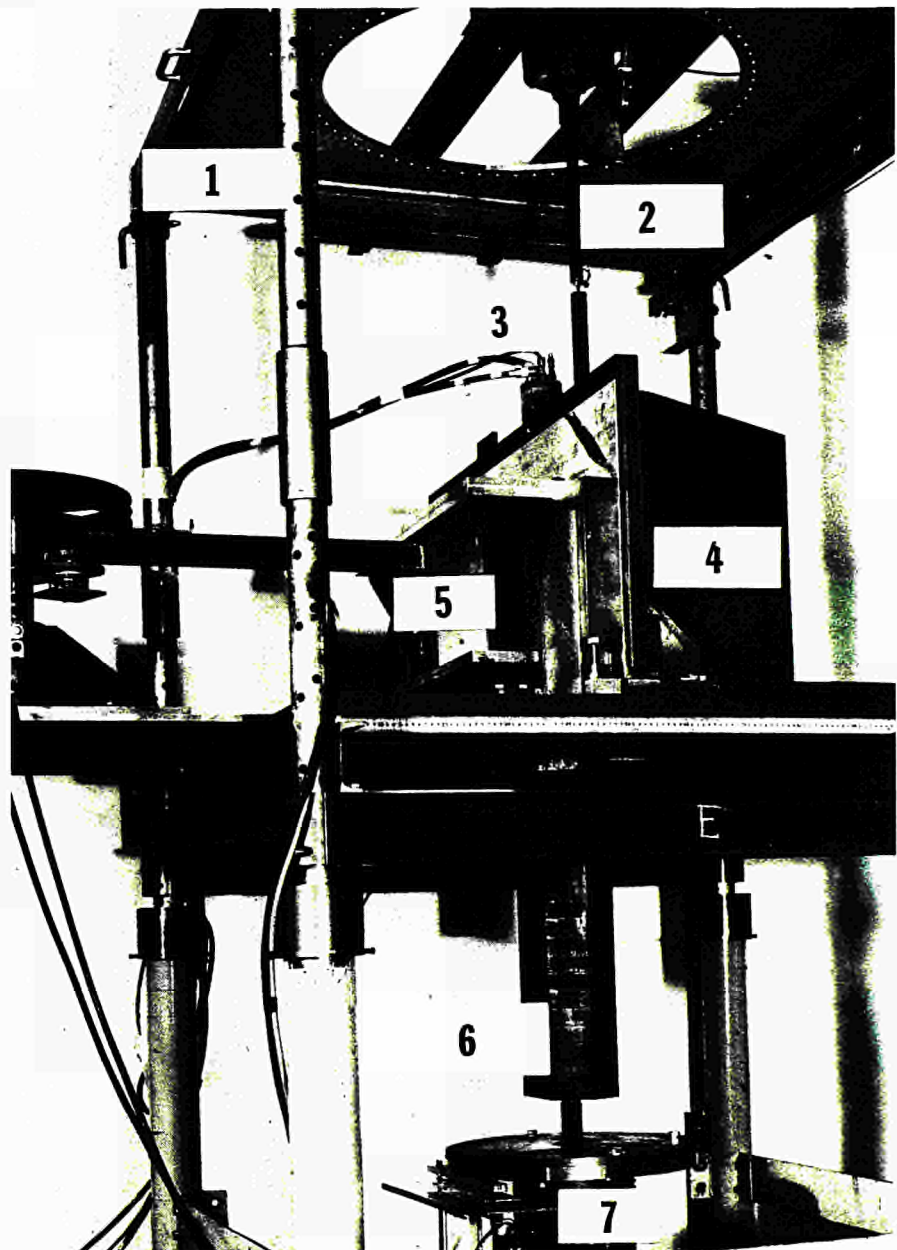
In a periodically pulsed reactor the reactivity is periodically varied in time. During each period there is a reactivity pulse which for a very short time raises the reactivity slightly ( $\epsilon_m \approx 130$  pcm) above prompt criticality. The neutrons coming from the delayed emitter decay are rapidly multiplied during this time. In this way a power pulse and thus a fast neutron pulse is produced. The relation between reactivity, power and delayed emitters is shown in Fig. 4.

The steady state condition for the pulsed reactor at pulsed criticality can

be formulated in very simple terms: the delayed emitters effectively decayed between two pulses are to be replaced (see Fig. 4) during the pulse.

This condition determines the background power  $P_b$  in terms of the prompt subcriticality between pulses,  $\epsilon_n$ , and

Fig. 2: SORA Critical experiment.  
1: support for rod-drive motor; 2: rod drive and connection; 3: detector; 4: fixed reflector; 5: movable reflector; 6: core withdrawn from reflector; 7: lift.



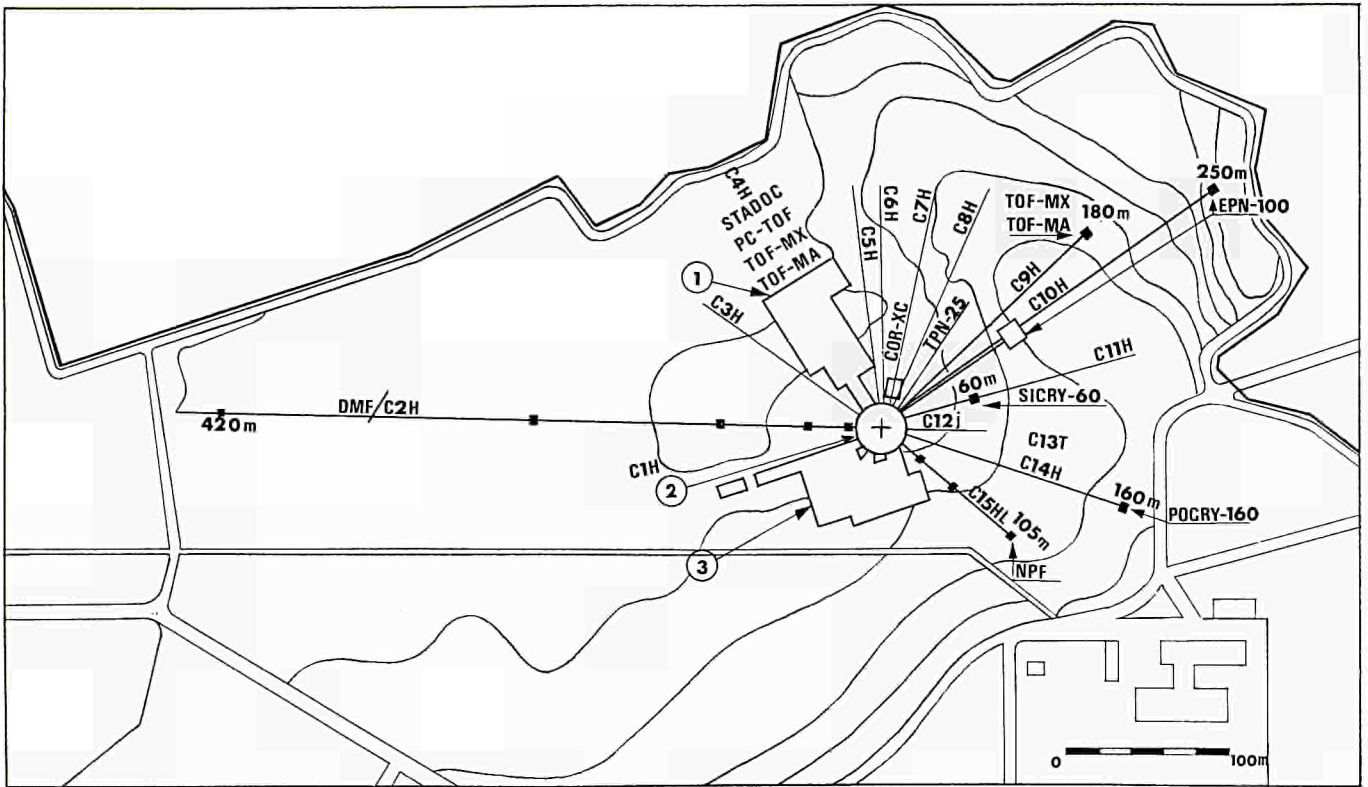


Fig. 3: SORA plant area with arrangement of experimental facilities.  
 1: experimental hall; 2: reactor building;  
 3: reactor ancillary complex.

the effective delayed neutron fraction,  $\beta$ , of the core

$$P_b = \bar{P} \cdot \beta / \epsilon_0 \cdot P \cdot 0.18$$

The remainder of the power is produced in the  $\nu$  pulses per second

$$\nu \cdot \int P_p(t) dt = \bar{P} (1 - \beta / \epsilon_0) = \bar{P} \cdot 0.82 \text{ pulse}$$

The shape of this pulse—an important characteristic for the use of the pulsed reactor—depends on the sharpness of the reactivity pulse and on the neutron lifetime in the core. In order to keep the lifetime short ( $\approx 30$  nsec) a fast neutron energy spectrum is required together with a very compact core. This leads to the application of liquid-metal-cooled fast reactor technology, highly enriched fuel and highest possible fuel volume fraction. The sharpness of the reactivity pulse depends both on the core compactness and on the mechanical performance of a unique component, called a pulsation

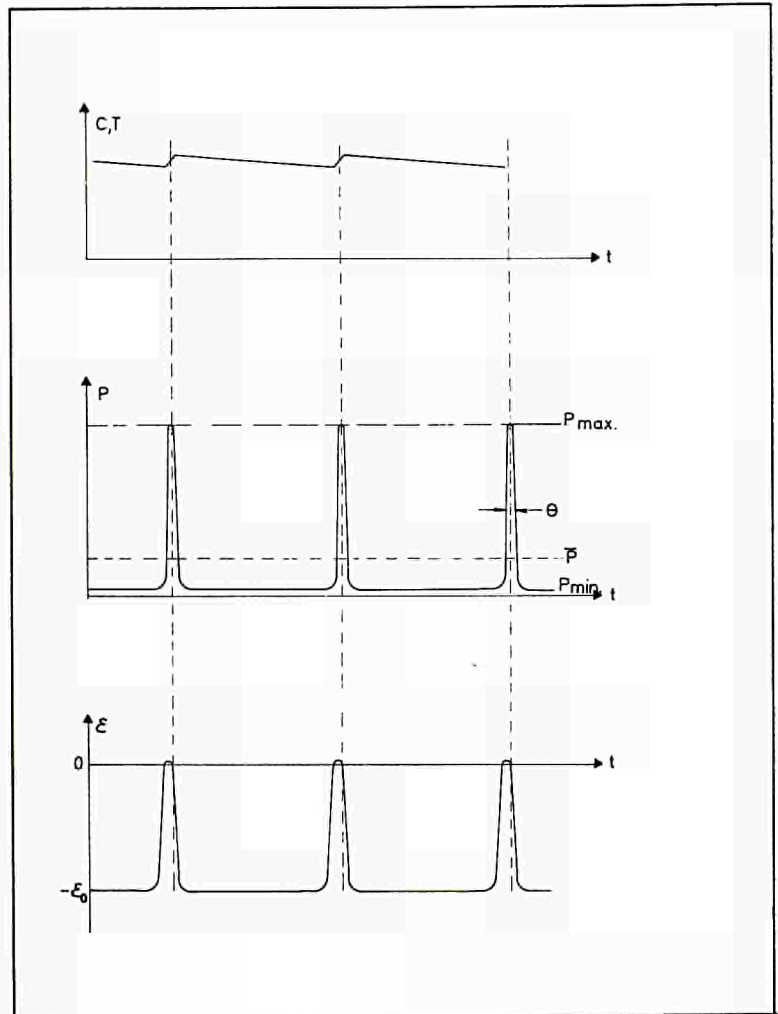


Fig. 4: Pulsed Reactor steady state conditions: variation of delayed emitters (C), core temperature (T), core power (P) and reactivity ( $\epsilon$ ) with time.

device, which carries a suitable piece of reflector at high speed past an unreflected side of the core. In this way power pulse widths down to about 75  $\mu$ sec can be obtained for *SORA*.

### Reactor Core and Reflector

The core design is a problem of primary importance. The most compact arrangement was chosen which was compatible with industrial experience in fast reactor development and with the experimenters' demands concerning the power pulse shape and the mean core power. An optimization study has been performed at Dubna (9) for a plutonium oxide core, correlating pulse characteristics with power and compactness. This study substantiates that the 10 l *SORA* core with a power extendable up to 3 MW (Mark 2) represents a good choice.

The reactor core (Fig. 5) is composed of 228 fuel pins arranged in hexagonal symmetry. The nominal spacing of 0.7 mm is ensured by a spiral wire. The fuel choice for the first core loading is rather conservative. Within the fast reactor development much experience has been accumulated on uranium-molybdenum alloys. The fuel slugs are liquid metal bonded to the 0.2 mm thick stainless steel cladding. An upper molybdenum reflector slug is incorporated into each pin. The core is 30 cm high. The fuel pins are collected into three concentric subassemblies, which are separated by wrapper cans. This results in a fuel fraction as high as 61% in a core volume of 10 liters.

The core is surrounded by a tightly fitting double-walled vessel. The coolant inlet is arranged above the reactor core. Through channels in the vessel wall the coolant reaches the lower plenum. In this way maximum space is provided below the core for accelerator drift tube and target (Mark 3).

Around the vessel there is a stainless steel structure, which guides the reactivity control rods which occupy five sides of the core. The safety rods next to the core window are made of beryllium, and can be disengaged with an initial acceleration of about 10 g. The other rods are made of a tungsten alloy and can be extracted by gravity. One core face is reserved for the reactivity pulsation.

### Moderators

Core and reflector are surrounded by a boron absorber layer, which prevents the return of slow neutrons to the core, and therefore keeps the neutron lifetime to a minimum. Outside this layer there are three hydrogen-containing moder-

ators, which tailor the fast neutron spectrum of the core to the distribution required by the experiments. The moderators are introduced into the reactor through special loading channels. The design of these facilities, in particular the positioning of the cylindrical moderators, provides largest possible freedom

Table II

<i>General Characteristics</i>	
Planned location	Joint Research Centre, Ispra
Reactor type	Repetitively Pulsed Fast Reactor
Use	Neutron beam research
Fuel	Uranium-Molybdenum (10%) alloy
Enrichment	93% U-235
Fuel loading	82 kg Uranium
Mean power:	
pulsed operation	1 MW
stationary operation	100 kW
Average core flux	$1.1 \times 10^{14}$ n/cm <sup>2</sup> /sec
<i>Core</i>	
Geometry	hexagonal
Subassemblies	3 concentric
Volume	10.2 liters
Height	30 cm
Fuel volume fraction	61.8%
Pin diameter	9.8, 11.5, 12.3 mm
Number of pins	228 including 10% dummies
Average power density	100 kW/l
Coolant velocity in core	2.0 m/sec
Coolant temperature rise	50 °C
Maximum fuel temperature without hot spot factor	400 °C
Temperature step per pulse	1.0 °C
Isothermal reactivity feedback	1.3 pcm/°C
<i>Control Rods</i>	
Type and material	reflector pieces, Be and W-alloy
Number of rods	9 (including 2 fast safety rods)
Total reactivity	6.2%
<i>Pulsation device</i>	
Pulse frequency	50 sec <sup>-1</sup>
Tip velocity	282 m/sec
Reactivity swing	4.0%
<i>Core cooling system</i>	
Prim. and second. coolant	sodium
Inlet temperature	220 °C
Design temperature	400 °C
Heat removal in nat. convection	50 kW
<i>Experimental facilities</i>	
Number of moderators	3 + movable reflector
Beam channels	
horizontal	13
inclined	9
average section at source	400 cm <sup>2</sup>

in the choice of moderator geometry, beam size, moderator material and temperature.

Fig. 7 shows the time-dependent neutron group fluxes in a 5 cm thick water moderator. Zero time corresponds to the centre of the power pulse. The moderator data given in Table I correspond to this figure. A large number of calculations has been done on very refined moderator designs with the result that the spectra can be tailored to any need, with energy distributions

Fig. 5: Core and reflector, horizontal section. 1: core; 2: vessel; 3: safety rods; 4: control rods; 5: start-up source; 6: boron absorber layer; 7: moderators; 8: movable reflector block; 9: coolant gas channels; →: beam directions.

peaking in the cold thermal or epithermal range (8).

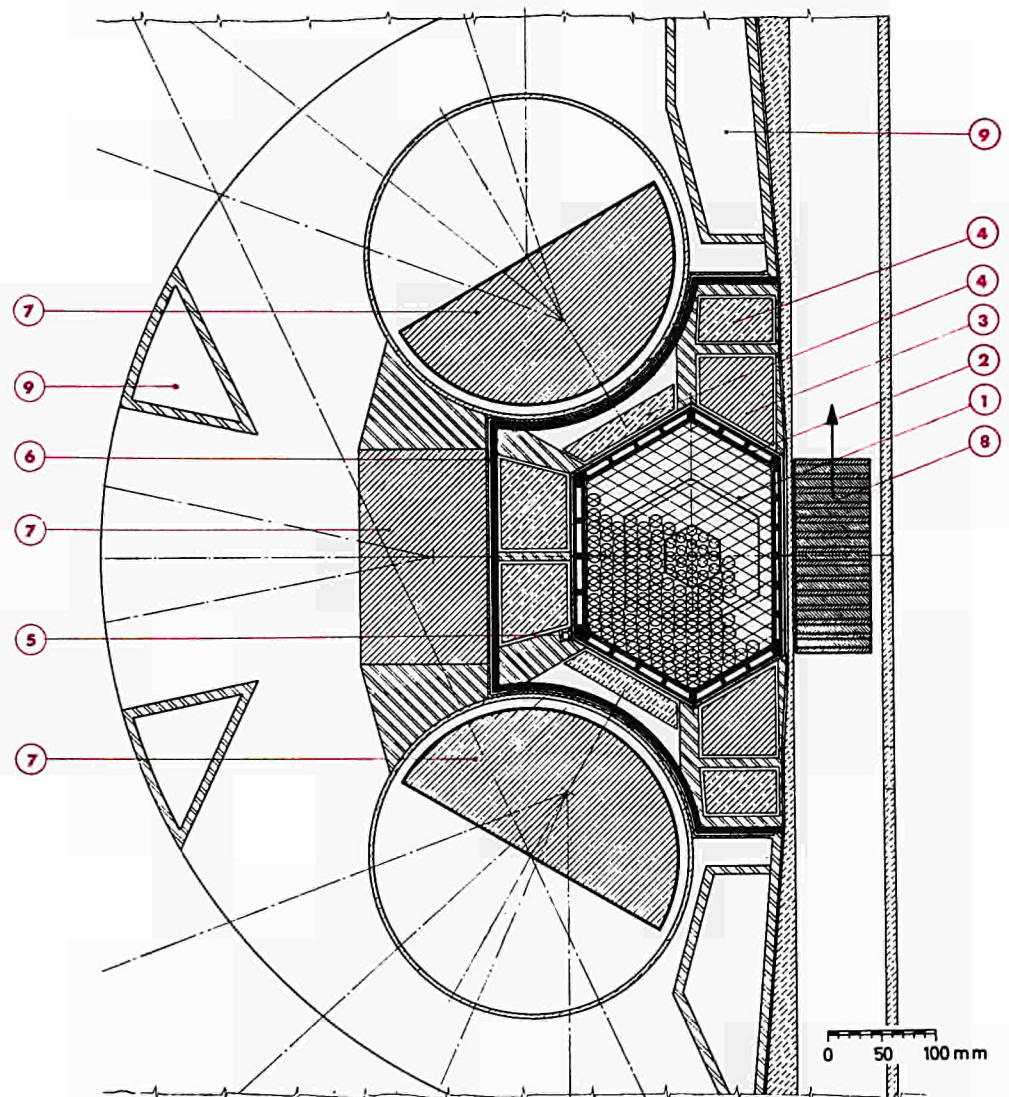
If pulse widths of 120  $\mu$ sec are tolerated, the peak leakage flux can be raised to  $1,4 \times 10^{15}$  n/cm<sup>2</sup>/sec. It is of particular interest that the necessary cooling power for the low temperature part of a cold moderator is only about 100 Watts at a 1 MW pulsed reactor. This gives great advantages in design and operation. It should in addition be pointed out that most of the flux values obtainable by *SORA* and its later versions are far beyond the reach of other types of neutron sources, including High Flux Reactors.

#### Pulsation Device

The pulsation device produces rapid reactivity variations and is therefore a

unique component of a pulsed reactor. The method used by the *IBR*, of moving a piece of fissionable material through the core, is not applicable at elevated core power because the heat production in the pulsation device itself would create big problems. Instead the reactivity is pulsed by the movement of a reflector piece past an unreflected side of the core (see also Fig. 5).

A vertical section of the pulsation device is shown in Fig. 7. The rotor consists of an Avional (aluminium alloy) disk of 150 cm diameter with one 29 cm high reflector piece protruding radially. The movable reflector is designed as a high strength sandwich structure of Avional and beryllium, 19 cm wide and 7 cm thick. Travelling at 282 m/sec it covers the reactor window 50 times per



second for a very short time ( $\sim 100$   $\mu\text{sec}$ ) and thereby raises the reactivity each time by 4 000 pcm.

The rotor moves in a helium-filled aluminium housing dissipating a friction power well below 60 kW, the projected power of the drive motor. The heated helium is circulated through an external loop for cooling and purification. The ball bearings are lubricated by a central oil circuit. The speed of the rotor is kept constant to  $\pm 0,1\%$  or better. Signals on speed and rotor angle are available for the experiments to synchronize the "time-of-flight" equipment.

In the housing, space is provided for a second rotor to be mounted concentrically to the first. The reactivity (i.e., the thickness) of the moving reflector blocks is then adjusted in such a way that a power pulse occurs only if both reflector blocks are simultaneously present in front of the core. In this way the pulse width is still determined by the first, fast-moving rotor and the frequency is determined by the speed of the second rotor. In this way pulse frequencies down to  $5 \text{ sec}^{-1}$  are attainable.

### The Reactor Block Design

The reactor block structure is shown in Fig. 8. Core and pulsation device are rigidly fixed in relation to each other. There are three downward-inclined handling channels for the moderators. They are viewed by thirteen horizontal beam channels (see Fig. 9)—one of which is a tangential tube—and nine upward-inclined channels. The larger part of the beam channels are intercepted by the vertical shutter tubes which play an important role in the experimental layout in pulse shaping, background attenuation and rough neutron energy band definition.

The reactor vessel continues from the core in an upward direction through a thermal column, which houses the neutron detectors, into a space serving as loop room for those components which need to be near the vessel, such as valves, vapour traps etc.

At the reactor top the vessel ends in a gas-tight and radiation-absorbing closure which also serves as interface to the various handling machines for the vessel internals. Specific handling provision had to be made for the pulsation device at one lateral face of the reactor block.

The access provided from below the reactor core serves for the control rods, and the accelerator beam tube and target for the Mark 3 version.

## UTILIZATION AND INSTRUMENTATION

### The Research Fields

For a research programme and the planning of the experimental facilities, research fields have been selected which are still developing and where the JRC neutron physics group has acquired experience during the last ten years:

*Condensed matter:* elastic or inelastic scattering of unpolarized or polarized neutrons in the thermal energy range.

*Nuclear physics and Neutron Data Measurements:*

- 1) the elastic scattering of epithermal and fast neutrons;
- 2) the elastic or inelastic scattering of polarized neutrons.

*Radiation Damage of solids.*

The fields "Condensed matter" and "Nuclear physics" use the property that the flux of SORA is pulsed, whereas for the field "Radiation Damage" only the time-averaged flux is of interest. The

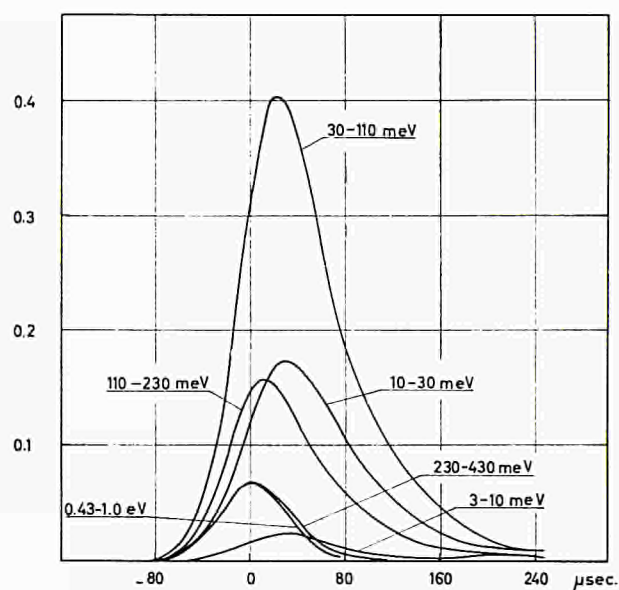
latter field will not be further treated in this article.

In the field "Condensed matter" which consists of neutron diffraction and inelastic scattering, the interaction of the neutrons with the atoms is used to obtain information on the structure and the dynamics of liquids and solids and on the properties of molecules.

Diffraction of neutrons by crystals is possible, since neutrons with thermal energies have wave-lengths that correspond to the spacing of the lattice planes in crystals. The main application of neutron diffraction is the structural analysis of hydrogen-containing crystals, which is difficult to effect with X-rays, and the analysis of the magnetic structure of crystals, which cannot be performed at all by X-rays.

In the real crystal the atoms in a lattice oscillate continuously around their equilibrium position. The frequency or the energy of the oscillation depends on the mass of the atoms, and the distances and forces between neighbours. The energy of the atomic motions, or the dynamics of the crystal, can be studied by inelastic scattering of thermal neutrons, since their energy is of the same order as the energy of the atomic motions. This information cannot practically be obtained by X-rays. Neutrons are a unique tool for this kind of measurement. The principle of the method is the measurement of the energy spectrum of an initially mono-

Fig. 6: Neutron group fluxes from room temperature water moderators. Zero time corresponds to power pulse centre.



chromatic beam after it has been scattered by a sample.

The use of polarized neutrons in neutron diffraction and inelastic scattering is a new experimental technique, which became feasible after the construction of high flux research reactors such as that at the Max-von-Laue/Paul Langevin Institut in Grenoble. This

new technique will allow a number of new measurements, especially on magnetic samples.

In the field of "Nuclear physics" certain aspects of the interaction of neutrons with nuclei are studied. The field of "Neutron Data Measurements" aims at the measurement of a large range of neutron data which are important for reactor physics. In addition to the usual time-of-flight technique special experimental techniques are used.

moderated neutrons pass at different velocities through the various beam channels to the experimental equipment, and interact with a sample the properties of which are to be investigated. In doing so the neutrons will be scattered in all directions. Some of the scattered neutrons reach a detector, and the time which has elapsed since the start of the reactor pulse is measured. This flight time is a measure of the neutron velocity, and hence of its energy and for each reactor pulse. The number of scattered neutrons that arrive at the detector with a certain velocity is measured.

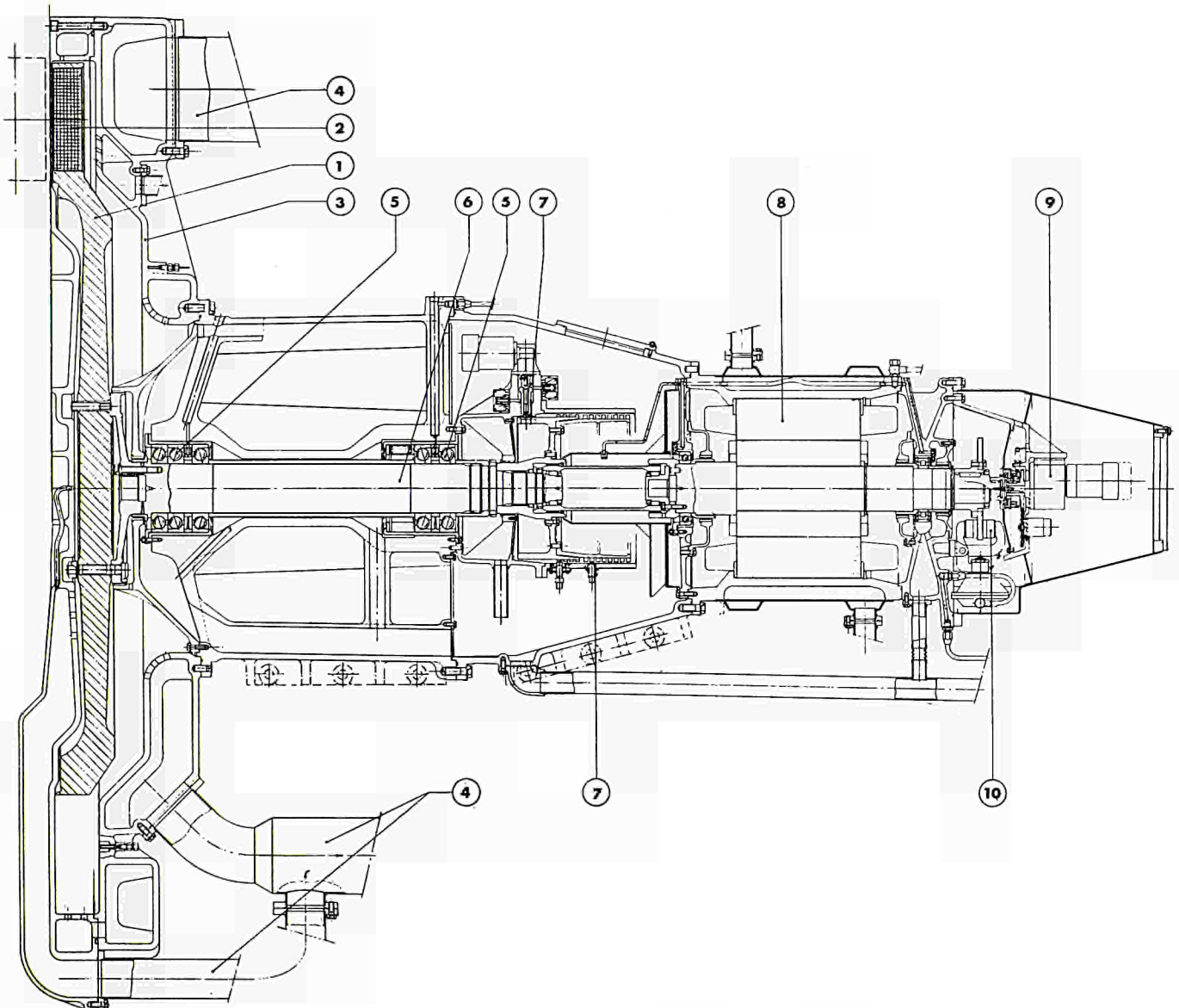
The time-of-flight technique is as simple as that for elastic scattering proc-

Fig. 7: Vertical section of SORA pulsation device.

- 1: Avional rotor;
- 2: movable reflector;
- 3: rotor housing;
- 4:helium ducts;
- 5: bearing sets;
- 6: rotor shaft;
- 7: instrumentation, drum and blocking mechanisms;
- 8: drive motor;
- 9: auxiliary drive;
- 10: brake.

### The "Time-of-Flight" method

The fast neutrons which are created during a reactor pulse are slowed down in the moderators. From there the



esses where the neutron velocity remains unchanged. For inelastic scattering, when the neutron energy changes at the sample, an additional and independent velocity determination must be done either before or after the sample.

From the measured flight time  $t_F$  and the known flight path length  $L$ , the velocity of the neutrons, which is the main parameter in time-of-flight experiments, is calculated by the simple formula

$$v = \frac{L}{t_F}$$

From the velocity  $v$ , the energy  $E$  and the wave-length  $\lambda$  of the neutrons can be calculated by the relations:

$$E = \frac{m}{2} v^2 \text{ and } \lambda = \frac{h}{m \cdot v}$$

where  $m$  is the mass of the neutron and  $h$  Planck's constant.

The fact that the neutrons of a reactor pulse start within a certain time interval  $\Delta t$  introduces an uncertainty into the velocity measurement

$$v \pm \Delta v = \frac{L}{t_F \pm \Delta t} = \frac{L}{t_F} \left( 1 \pm \frac{\Delta t}{t_F} \right)$$

The ratio

$$R = \frac{\Delta t}{t_F} = \Delta t \cdot \frac{v}{L}$$

is called the resolution and is an important characteristic of a time-of-flight facility. The resolution of a time-of-flight facility improves with a longer flight path. A limit to the length of the flight path is the loss of neutron intensity, which is in theory inversely proportional to the square of the flight path length. The loss of intensity can be appreciably reduced by using neutron guide tubes as flight path. These guide tubes have the property of reflecting neutrons, which impinge under certain angles on the walls, in the flight direction.

Another important consideration is the overlap of the most rapid neutrons of a reactor pulse with the slowest neutrons of the preceding pulse. In order to avoid this overlap, the difference of the flight time between the slowest and most rapid neutrons in a pulse must not be longer than the time interval between two successive pulses, the period  $T$ :

$$\frac{L}{V_{\max} - V_{\min}} = T$$

Eliminating the flight path length from this relation with the help of the formula for the resolution and using the relation between the velocity and the wave-length one obtains:

$$\frac{\lambda_{\min}}{\Delta \lambda} R_0 = \frac{t}{T}$$

where  $R_0$  is the resolution for the neutrons with the minimum wave-length or maximum velocity in the beam. This formula relates the design parameters of the time-of-flight facilities, the minimum wave-length  $\lambda_{\min}$ , the wave-length band  $\Delta \lambda$  of the beam and the resolution  $R_0$  at the minimum wave-length to the operating parameters of the reactor or the moderators, namely to the period  $T$  and the widths  $\Delta t$  of the neutron pulse.

Fig. 10 illustrate the above explanations for elastic scattering. Fig. 10 gives the distance of the neutrons with a velocity  $v$  within the velocity band  $V_{\min}, V_{\max}$  from the source as function of the time elapsed since the start of a period.  $L$  indicates the distance source-sample. If the minimum velocity is chosen too small there is an overlap between neutrons of two consecutive pulses. By increasing the minimum velocity, i.e. reducing the width of the velocity band, the overlap disappears.

Fig. 11 represents the inelastic scattering. Neutrons with the energy  $V_0$  are scattered by the sample and gain energy from the sample, i.e. becoming faster, or lose energy to the sample, i.e. becoming slower. At this velocity the neutrons fly over the distance sample-detector unit. Again, in planning an experiment the overlap of neutrons from two successive pulses has to be avoided.

### General Description of the Experimental Facilities

The common features of the time-of-flight facilities are:

- an incident beam system;
- the primary flight path;
- the sample system;
- the secondary flight path;
- the detector unit, and
- the data system.

The incident beam system consists mainly of two or more choppers and defines the velocity band of the neutrons in the incident beam. The primary

flight path is a distance between the reactor and the sample which consists of an evacuated tube and (especially for long flight path distances) a neutron guide tube, which guides the neutrons to the sample. The length of the flight path is inversely proportional to the resolution.

The sample system depends almost entirely on the intended experiment. Generally it allows the orientation of the sample with respect to the incident beam. In addition it may, by special means, make it possible to keep the sample at low or high temperature, maintain it under pressure or in a magnetic field.

The secondary flight path is the distance between the sample and the neutron detector.

The detector unit combined with the data system counts the number of neutrons that are scattered by the sample in a certain angular range with reference to the direction of the incident beam, measures the arrival time and perhaps analyses the energy of the scattered neutrons and their state of polarization. The data are stored in the data system until transfer for further evaluation.

In addition to the above-mentioned systems the facilities for work with polarized neutrons include the following:

- a polarizing system in the incident beam, and
- an analysing system in the secondary flight path.

A polarizer and analyzer exhibit a high transmission for neutrons with one spin orientation and a low transmission for neutrons with the opposite spin orientation.

### The Neutron Diffraction Facilities

The generally used method for neutron diffraction is similar to that of X-ray diffraction. A monochromatic neutron beam impinges continuously on a sample and the diffracted intensity is measured as function of the scattering angle. The spectral distribution of wave-lengths in a neutron beam with thermal energies has led to the development of time-of-flight neutron diffraction. In this method polychromatic neutron pulses impinge periodically on the sample and the distribution of the intensity neutron flight time is measured by one or several detectors at fixed scattering angles as function of the flight time.

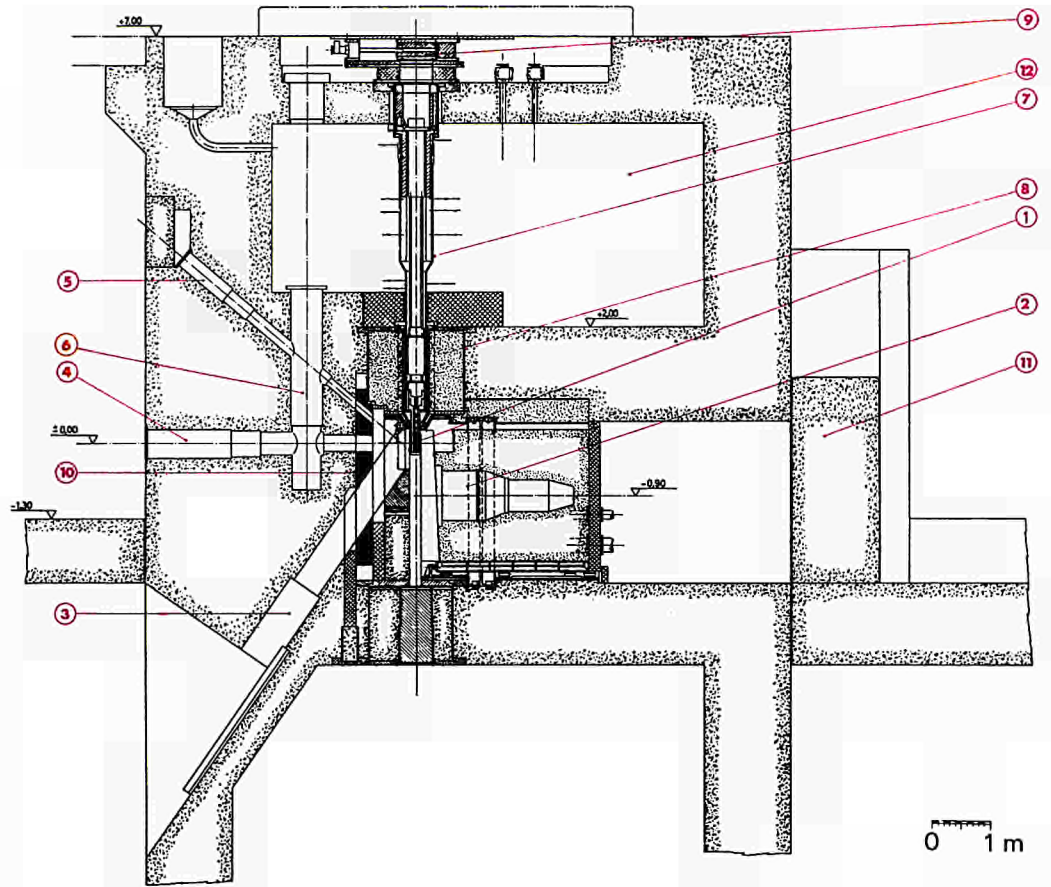


Fig. 8: Vertical section of reactor block.  
 1: core; 2: pulsation device;  
 3: moderator loading tube;  
 4: horizontal beam tube; 5:  
 inclined beam tube; 6: shutter  
 hole; 7: reactor vessel; 8:  
 thermal column; 9: vessel  
 closure; 10: thermal shield;  
 11: pulsation cell door; 12:  
 upper loop room.

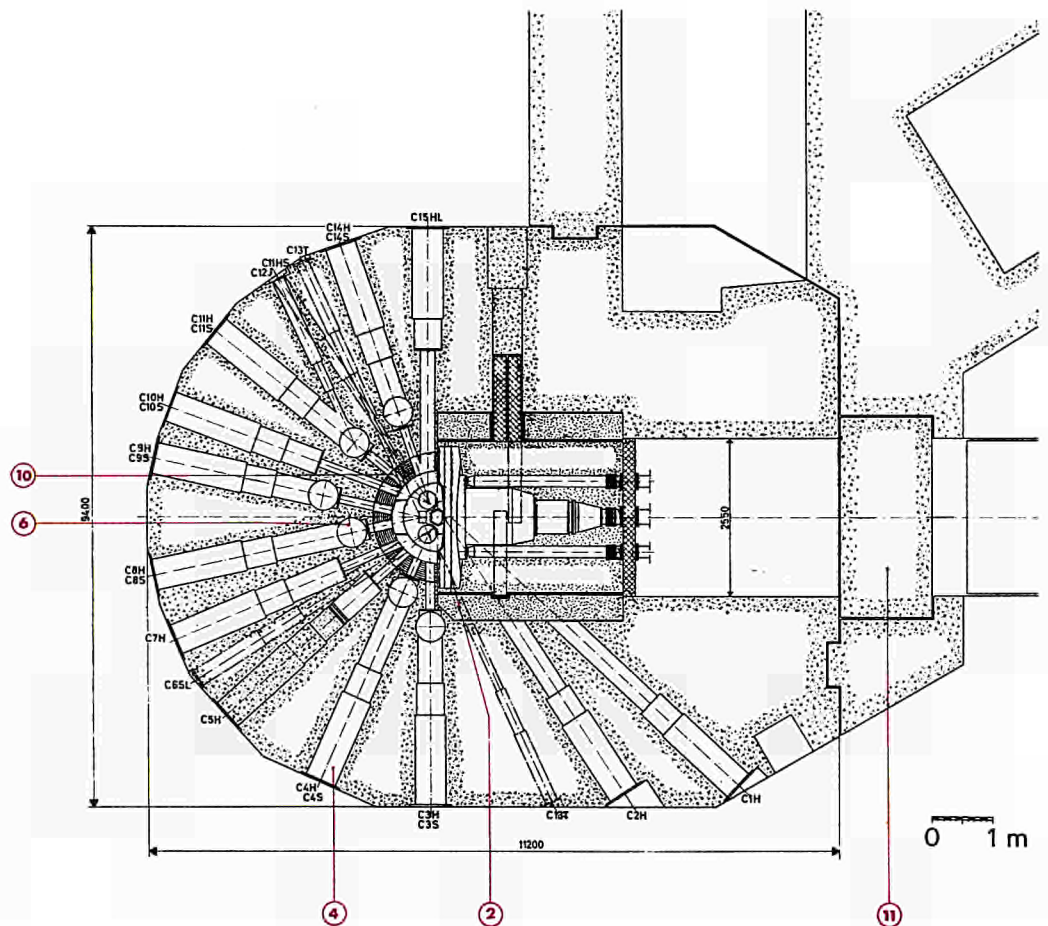


Fig. 9: Horizontal section of reactor block with the horizontal beam tubes.  
 1: core; 2: pulsation device;  
 3: moderator loading tube;  
 4: horizontal beam tube; 5:  
 inclined beam tube; 6: shutter  
 hole; 7: reactor vessel; 8:  
 thermal column; 9: vessel  
 closure; 10: thermal shield;  
 11: pulsation cell door; 12:  
 upper loop room.



Fig. 10: Distance vs. flight time diagram for pulsed beams.  $\Delta t$  is the pulse length,  $T$  the pulse repetition period.

Development of the conventional neutron diffraction technique started in the late Forties, while the principle of the time-of-flight technique was demonstrated in 1963 by Buras and coworkers (12).

The time-of-flight method offers certain advantages as compared with the conventional technique, especially for samples under extreme conditions, e.g. high pressure and low temperature. Fig. 12 shows a time-of-flight neutron diffraction pattern of bismuth at a pressure of 26 000 bar, measured at a scattering angle of  $60^\circ$  (13).

Fig. 13 shows a qualitative comparison of the conventional double axis/crystal technique using a continuous neutron source and the time-of-flight technique using a pulsed neutron source (14).

In a double axis spectrometer a white spectrum of high intensity hits the monochromator crystal, but only a small fraction of the incident neutrons is reflected in the monochromatic beam, and with a monochromatic beam at one scanning position, only one reflection is measured. With the time-of-flight technique the pulse with a lower number of neutrons per pulse and a certain wavelength range is incident on the sample and all the Laue reflections possible in this wavelength range are observed simultaneously in the detector. With the large number of reflections to be observed in structure analysis work with single crystals, the time-of-flight technique gives a much higher data collection efficiency.

Fig. 14 shows the lay-out of a time-of-flight powder diffractometer. Detector rings which consist of several detectors lie along a portion of Debye-Scherrer cones of semi-angles  $2\theta$ . The collimated incident beam is pulsed and contains a range of wave-length. Each family of crystal planes with an interplanar spacing of  $d_h$  will reflect the beam into the detector ring at times when neutrons of the wave-length  $\lambda_h$  arrive at the sample. The wave-length  $\lambda_h$  is related to the interplanar spacing  $d_h$  and the semi-angle of the detector ring by the Bragg equation.

The intensity arriving at the detector ring is measured as a function of time. The measured flight time is used to identify the reflection. By this technique all possible reflections with lattice spacings  $d$  in a certain range can be observed

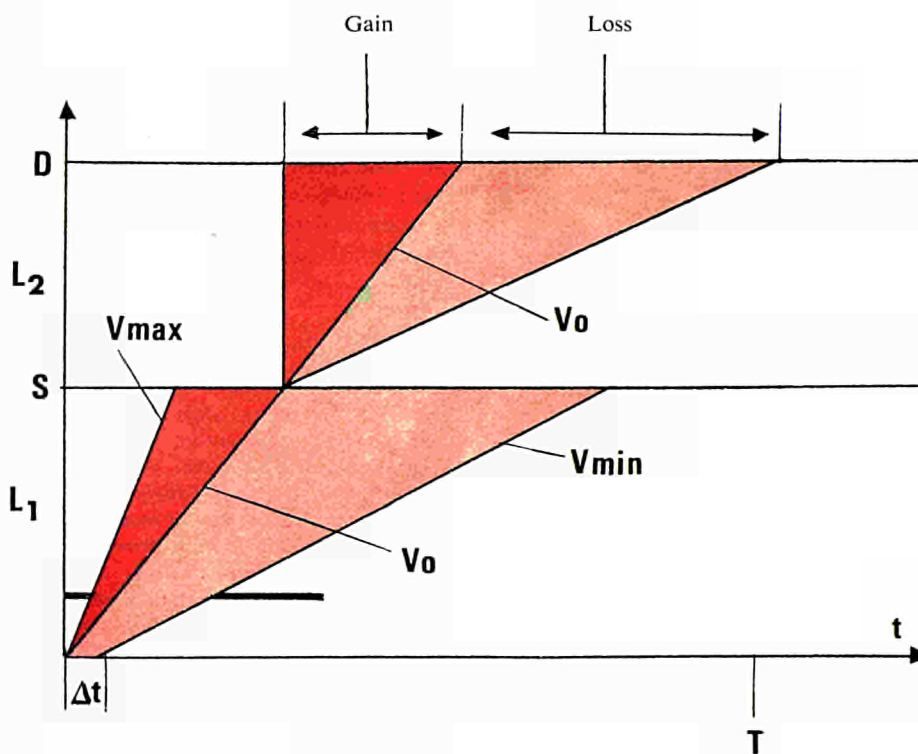
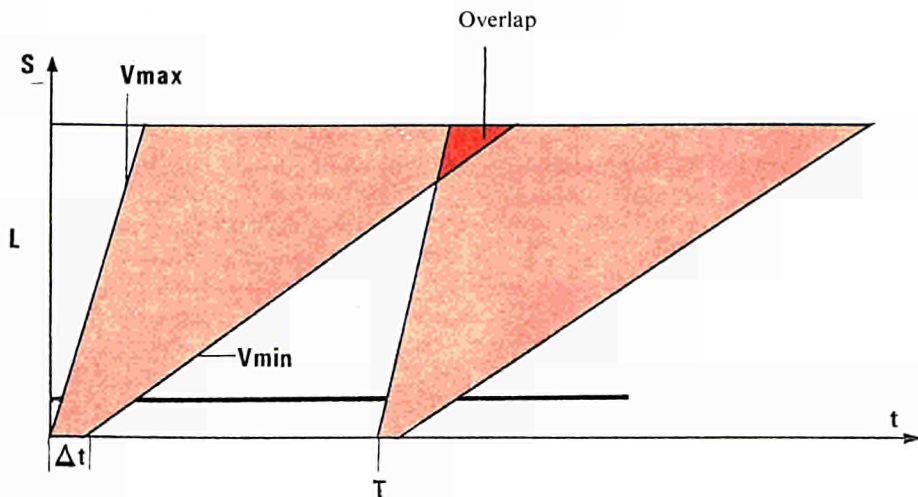
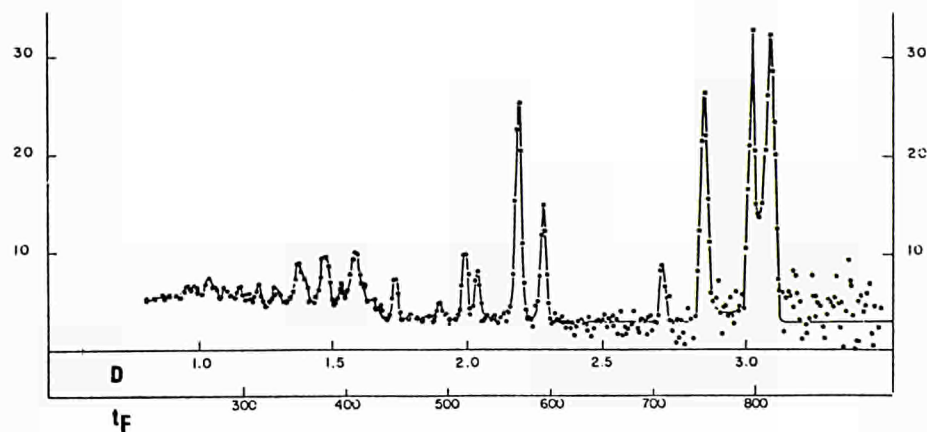


Fig. 11: Distance vs. flight time diagram for inelastic scattering experiments. Gain and loss correspond to processes where the energy of the incident neutrons is increased or decreased by the scattering.

Fig. 12: Time-of-flight diffraction pattern of bismuth at 26 k Bar pressure for a scattering angle of  $60^\circ$ .  $D$ : interplanar spacings for  $60^\circ$  scattering, in Ångströms;  $t_F$ : neutron time-of-flight, in  $10^{-6}$  sec/m.



simultaneously and no mechanical scanning procedure is required.

By placing detector rings at different scattering angles it is possible, with a limited wave-length range in the incident beam, to measure a large range of lattice spacing.

Fig. 15 shows the lay-out of a simple time-of-flight crystal diffractometer with an extended detector which should be considered as a section of a sphere with the sample as centre. The incident pulsed beam again contains a range of wave-lengths. Each time neutrons of the wave-lengths  $\lambda_{hkl}$  arrive, the sample reflects these neutrons in an angular direction which is related to the wave-length and the lattice constants of the crystal by the Laue equations. By using a time-of-flight single crystal diffractometer with an extended detector many reflections can be measured simultaneously without the need for a mechanical scanning procedure.

Two time-of-flight diffractometers for powder measurements (*POCRY-160* and *POCRY-16*) and one for single crystal measurements (*SICRY-60*) have been studied. The main characteristics of these diffractometers are given in Table III. The distribution of the various experimental facilities with their flight path and measuring stations in the *SORA-Plant* area is shown in Fig. 3. *POCRY-160* is a high resolution diffractometer with an allowed wave-length range of 0.4 Å or a quasi-monochromatic incident beam, whereas *POCRY-16* is a moderate resolution diffractometer with an allowed wave-length range of 3.2 Å, which corresponds to a quasi-white incident beam.

The detector unit of *POCRY-160* consists of a set of nine counter rings, placed at scattering angles between 15° and 170°, in such a way that the measurable range of interplanar spacings completely converts the minimum value,  $d_{\min} = 0.5 \text{ \AA}$ , to the maximum value  $d_{\max} = 5.4 \text{ \AA}$ .

For *POCRY-16* a similar detector unit consisting of three counter rings is used. Fig. 16 shows the conceptual design of *POCRY-16*.

The detector unit of *SICRY-60* consists of a set of two-dimensional detectors. Ten of these detectors are mounted on a movable segment above and at a distance of 1 m from the sample position.

Fig. 13: Schematic diagram showing a comparison of the intensities obtained from a continuous neutron source using the conventional and the time-of-flight techniques. 1: spectrum of a continuous incident beam; 2: Laue reflections from a pulsed incident beam; 3: pulsed incident beam spectrum; 4: a continuously monochromated component; 5: reflected intensity available from a monochromatic component.

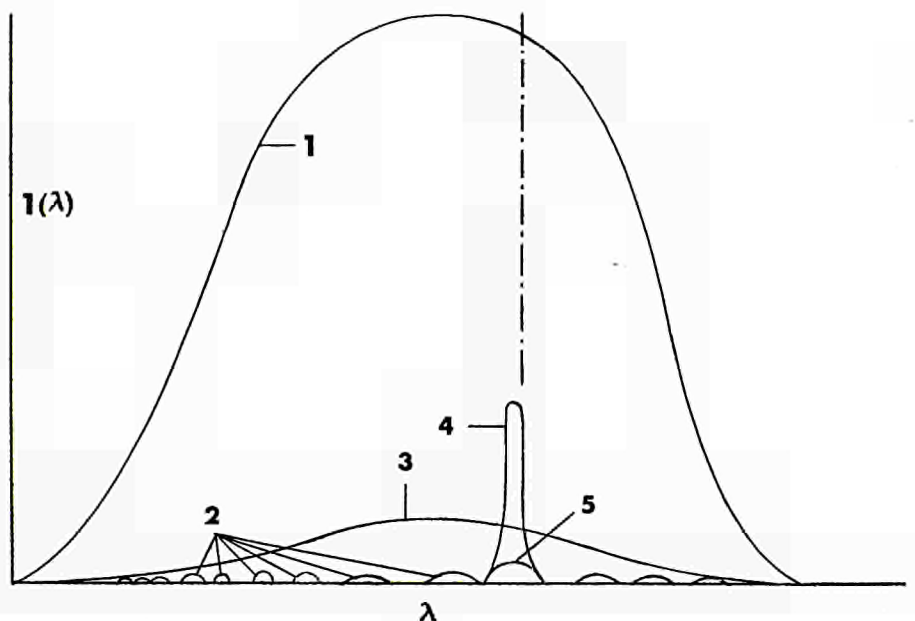


Table III: Characteristics of the facilities for time-of-flight neutron diffraction. They include the facility name, the beam hole, the method for the energy determination for the incident neutrons, the width of the wave-length band  $\Delta\lambda$ , and the minimum wave-length  $\lambda_{\min}$ , the flight path length  $L$  with the indication as to whether a guide tube is used, and the wave-length resolution over the complete wave-length band.

Spectrometer	Beam hole	Energy determ. of	Characteristics the incident beam		Flight path		$\Delta\lambda/\lambda$ , wave-length resolution, %
			$\Delta\lambda$ , Å	$\lambda_{\min}$ Å	length $L$ , m	guide tube	
1 <i>POCRY</i> — 160	C 14 H	ToF	0.4	1.0	160	yes	0.2 — 1.5
2 <i>POCRY</i> — 16	C 9 S		3.2	2.0	16	—	1.2 — 1.9
3 <i>SICRY</i> — 60	C 11 H		1.6	1.0	60	—	0.7

Table IV: Characteristics of the facilities for inelastic scattering experiments. This table differs from Table III in that the width of the energy band and the maximum value of the energy of the neutrons in the incident beam and the energy resolution are indicated. In addition, the method for the energy analysis of the scattered neutrons is given.

Spectrometer	Beam hole	Energy Det./Properties of the incident beam			Flight path		Energy analysis of scatt. neutrons by	Energy resolution $\frac{\Delta E}{E}$ , %
			$\Delta E$ meV	$E_{\max}$ meV	length $L$ , m	guide tube		
1 ToF/MX — MR	C 4 H	ToF	110	130	70	yes	crystal	0.6 — 1
2 — HR	C 9 H		130	200	180	yes		0.3 — 0.5
3 ToF/MA — MR	C 4 H	ToF	110	130	75	yes	crystal systems	0.6 — 1
4 — HR	C 9 H		80	130	180	yes		0.2 — 0.4
5 COR/XC	C 5 H	Crystal	95	100		—	ToF	0.8 — 1.2
6 /XM	C 6 SL		145	150		—		0.8 — 1.3
7 PC—ToF	C 4 H	ToF	42	50	60	yes	ToF	0.7 — 1
8 STADOC			120	130	40	yes		0.7 — 2
9 TPN (*)	C 8 H	ToF	52	60	25	—	crystal	0.5 — 1.5

(\*) TPN: The Thermal Polarized Neutron Facility has a polarizer system in the incident beam and a system for analysing the polarization state of the scattered beam.

Table V: Characteristics of the facilities for nuclear physics experiments with epithermal and fast neutrons. The minimum and maximum energy  $E_{\min}$  and  $E_{\max}$ , of the neutrons in the incident beam and the energy resolution are given.

Spectrometer	Beam hole	$E_{\min}$ eV	$E_{\max}$ eV	Energy Det. of incident beam	Flight path length m	Energy analysis of scatt. neutr.	Energy resolution %
1 NPF	C 15 HL	0.01	10	ToF	100	—	0.2 — 5
2 EPN (*)	C 10 H	0.1	100		100/250	crystal	0.3 — 10
3 DMF	C 2 H	10eV	1MeV		450	—	0.1 — 6

(\*) EPN: The Epithermal Polarized Neutron Facility has a polarizer system in the incident beam and a system for analysing the polarization state of the scattered beam.

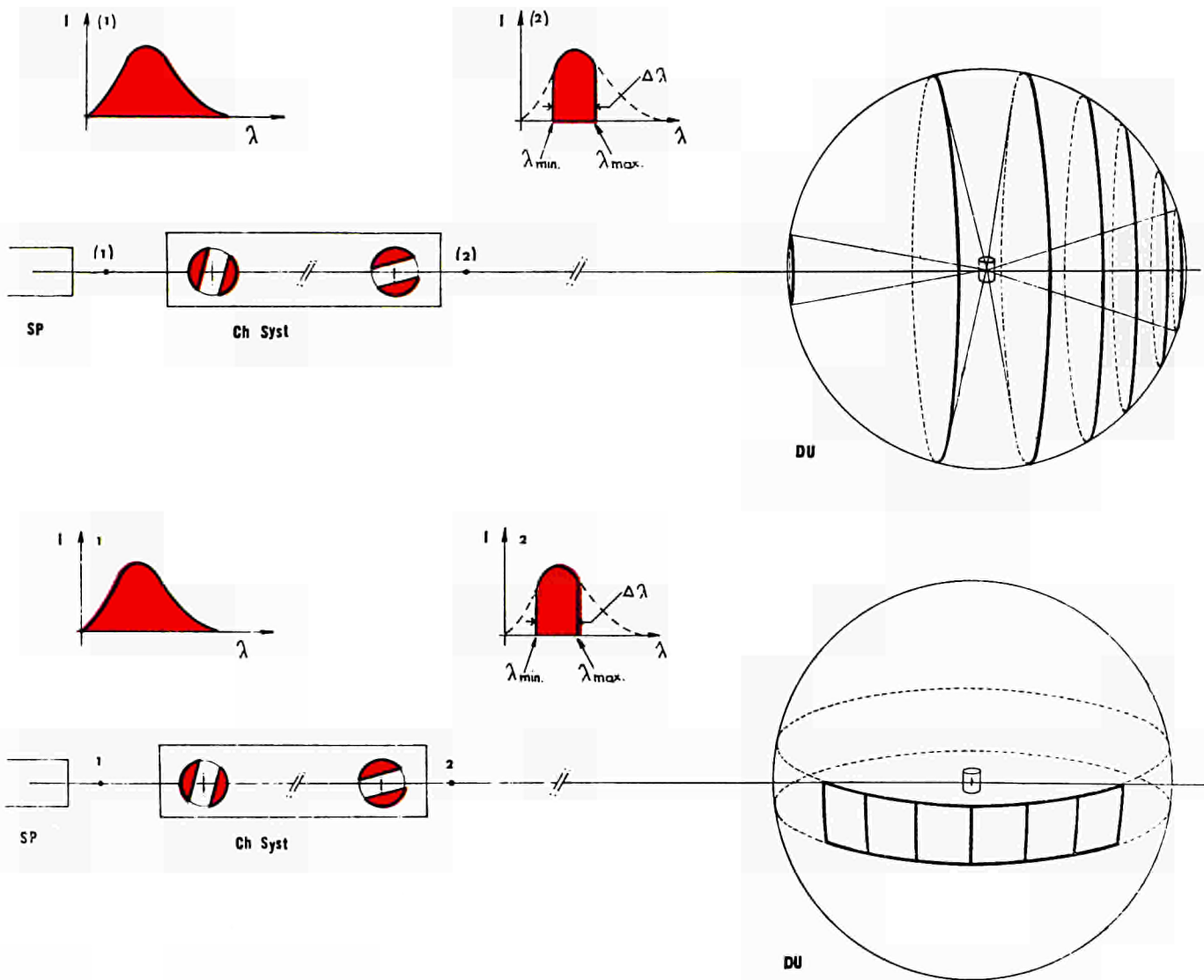


Fig. 15: Diagram showing the lay-out of a time-of-flight single crystal diffractometer with an extended, two-dimensional position sensitive detector unit.

SP: source of pulsed neutrons; Ch Syst: system of several choppers, defining the wave-length band; DU: detector unit. The inserts (1) and (2) show schematically the neutron spectrum in front of and behind the chopper system.

Fig. 16: Design of POCRY-16.

1: core; 2: moderator; 3: background rotor; 4: conical beam channel; 5: overlap rotor; 6: beam monitor and diaphragm; 7: detector conus; 8: ring detectors with diaphragms; 9: sample position; 10: sample loading tube; 11: beam monitor; 12: containment.

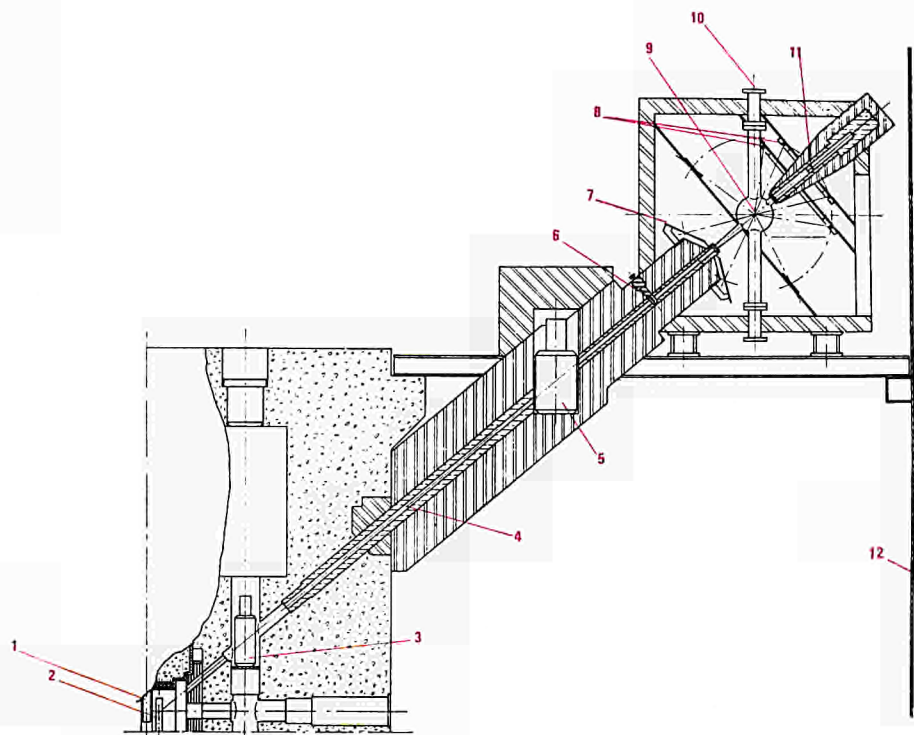




Fig. 14: Diagram showing the lay-out of a time-of-flight powder diffractometer with a detector unit consisting of seven detector rings (POCRY-160).

SP: source of pulsed neutrons; Ch Syst: system of several choppers, defining the wave-length band; DU: detector unit. The inserts (1) and (2) show schematically the neutron spectrum in front of and behind the chopper system.

The geometrical dimensions of the detector unit of ten detectors cover at 1 m distance an angular region of  $43^\circ \times 11.7^\circ$ .

### The Inelastic Scattering Facilities

The methods of inelastic scattering of neutrons which became possible after the development of research reactors opened new research fields in the physics of solids and liquids. In reactors with continuous flux the time-of-flight method is used as well as the more common method which produces the incident monochromatic beam and analyses the energy distribution of the scattered beam by crystals. Fig. 17 shows a time-of-flight spectrum of an anthracene sample, which has been obtained by using one of the first time-of-flight methods developed, the Be-filter method, in the reactor *ISPRA-I*. In the region (1) the time-of-flight distribution of the practically monochromatic beam is seen and in region (2) the spectrum of the scattered neutrons. The scattered neutrons have a shorter flight time, i.e. they have gained energy during the scattering process.

Parallel to the technical work on the *SORA* project the existing time-of-flight methods have been critically reviewed with the aim of proposing new and better time-of-flight methods for *SORA* which has a peak flux that corresponds to the neutron fluxes of high flux reactors and is an order of magnitude higher than the fluxes of the generally available research reactors.

Nine spectrometers:

- 4 time-of-flight/crystal analyser spectrometers: ToF/MX and ToF/MA for each one a medium and high resolution spectrometer,
- 2 monoenergetic correlation spectrometers: COR/XC and COR/XM,
- 2 double time-of-flight spectrometers: PC-ToF and STADOC,

- 1 time-of-flight spectrometer for polarized neutrons: TPN-facility,

have been studied for inelastic scattering. The main characteristics of these spectrometers are summarized in Table IV.

The *time-of-flight/crystal analyzer spectrometers* use as incident spectrum all neutrons within a wave-length band which is defined by a special rotor system. The energy determination of the incident neutrons is made by measuring the time-of-flight over a guided path. Scattered neutrons are analyzed by three movable single crystals or by a system of four multi-angle reflecting crystals. In this system the neutrons are counted by four parallel position-sensitive detectors, which allow the simultaneous measurement of neutrons within an extended range of scattering angles. Two of these spectrometers are medium resolution spectrometers and two high resolution spectrometers with a neutron guide tube with a flight path of 180 m.

The *correlation spectrometers* use the correlation technique in order to improve the resolution which is otherwise limited by the inherent long reactor pulse. The polychromatic neutron pulse coming from the reactor is monochromatized by a system of monochromators and chopped statistically. The chopped pulsed beams are then scattered by samples and velocity-analysed by time-of-flight measurements. The advantages of the "pulsed" correlation method enable these spectrometers to be used for high-resolution experiments to investigate small details of density of states curves or of molecular spectra.

The *PC-ToF and the STADOC-spectrometers* are double time-of-flight spectrometers. Both the incident and the scattered neutrons are selected and/or analyzed by measuring the flight times over certain distances using periodic or statistical disk choppers to define the time-of-arrival of the incident neutrons.

The TPN-Facility polarizes the incident neutrons by a polarizing system which consists of a polarizer and a device termed a "spin rotator". With this device the neutron polarization vector can be reversed and oriented in various directions, according to the requirements of each experiment. The polarization of scattered neutrons is analysed by an analysing system consisting of a second spin rotator followed by an analyser.

This spin rotator can be used for analysis of the polarization vector components of scattered neutrons, or simply as a polarization inverter. The analysing system can be positioned at different scattering angles by rotating it around an axis coinciding with the sample position. A magnetic mirror system is used as polarizer and as analyser. The magnetic mirrors are highly polished plates of a Co—Fe alloy and are maintained in a permanent magnetic field. These plates have the property of reflecting neutrons of one spin state almost completely and of being transparent for neutrons in the other spin state. Only the reflected neutrons form the neutron beam which impinges on the sample.

### The Nuclear Physics and Neutron Data Measurement Facilities

The time-of-flight method makes it possible to study a number of phenomena (e.g., the fission process, capture gamma rays) with the energy of neutrons as parameter. The range of possible experiments is extended to spin-dependent effects polarizing the white neutron beam by the use of the recently developed highly polarized neutron target. In addition to these more fundamental investigations in the field of low energy nuclear physics it is possible to measure neutron data that are useful for reactor physics calculations.

Three different facilities:

- the Data Measurement Facility (DMF),
- the Nuclear Physics Facility (NPF) and
- the Epithermal Polarized Neutron Facility (EPN)

have been studied.

The main parameters of the facilities are summarized in Table V. The general lay-out of each of the facilities corresponds closely to the description given in a former chapter. Each of the facilities has along its flight path two or more measuring stations with detector units at different distances from the reactor.

The Epithermal Polarized Neutron Facility has, in the same way as described in the preceding chapter for the TPN-Facility, a polarizing system in the incident beam and an analysing system in the scattered beam. As polarizer and

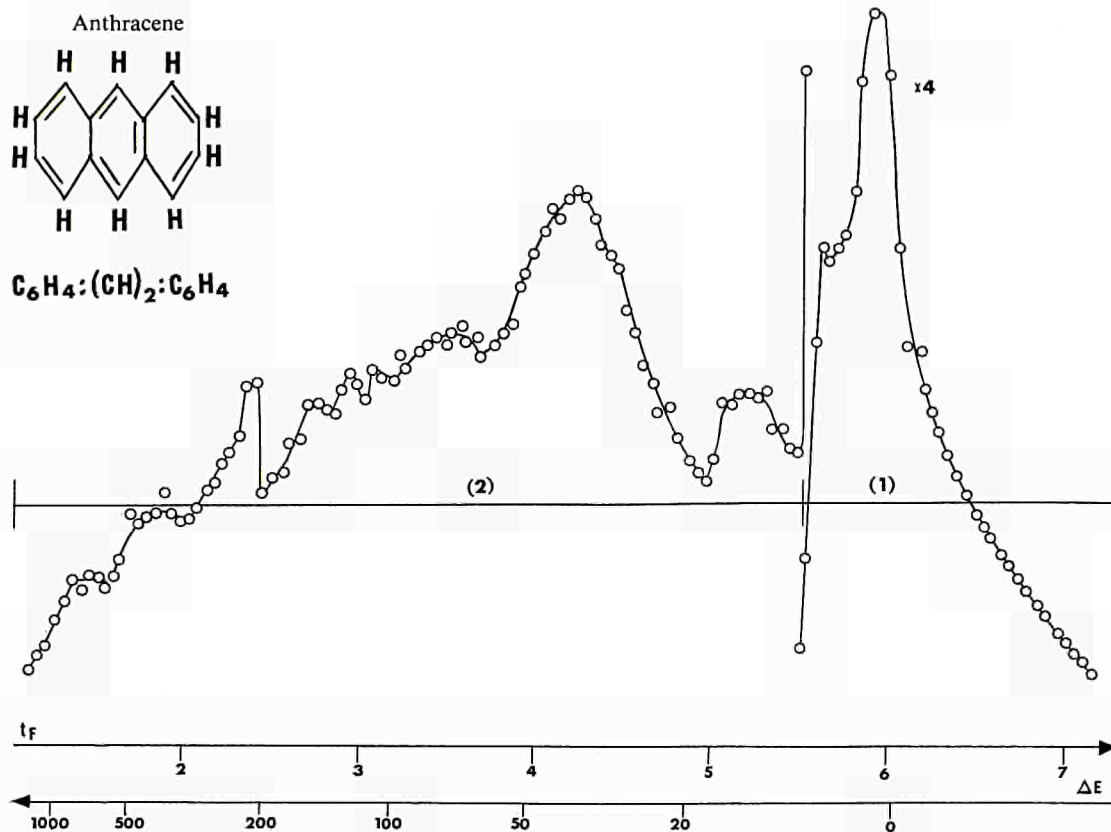


Fig. 17: Energy spectrum of a monochromatic beam scattered by an anthracene sample.  $t_F$ : neutron time-of-flight, in  $10^{-3}$  sec/m;  $E$ : energy transfer from the sample to the neutrons in arbitrary units.

as analyser, a polarized proton target is used because of the higher energies of the neutrons. By the use of these recently developed targets, a beam of highly polarized neutrons is obtained by transmission. Neutrons in one spin state pass the polarized proton target with practically no scattering, whereas a large fraction of the neutrons in the other spin state is scattered out of the beam.

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# Gas : the great energy asset\*

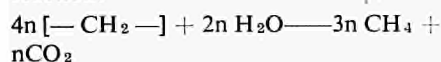
*Model of a strategy to snatch  
a bargaining power in the oil deal*

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CESARE MARCHETTI, CARLO RINALDINI, ALBERT SCHNEIDERS

THIS PAPER PRESENTS an analysis of the secondary consequences of the introduction of Substitute Natural Gas (SNG) processes (1) using oil as the feedstock, whose development started in Britain about 20 years ago and is now ready for industrial application.

SNG plants are a kind of reforming refinery in which oil fractions, and even crude oil itself, are in a sense "burned" in steam according to the overall reaction:



The symbol  $[-CH_2-]$  is used to represent the building block of an aliphatic molecule, although there is no implication that only aliphatic molecules can be used. The reaction is slightly exothermic.

Alternative processes (2) are based on straight hydrogenation, hydrogen being produced in the usual way by the steam-reforming part of the feedstock.

A very important feature of these SNG plants is that in capital and operating costs and in terms of efficiency they are comparable to a present-day refinery producing the usual mix of oil products if desulphurization is included.

## The energy picture in Europe

A mere twenty years ago the question of energy supply problems in Europe simply did not arise. Recoverable coal reserves were sufficient for at least 200 years and only a small fraction of energy requirements was covered by oil imported from the very abundant reserves in the Middle East. But the decline of the coalmining industry, the intrinsic advantages of fluid fuels, and the "cheap

oil" policy of the oil companies have since brought about a profound change.

In the present pattern of Europe's energy supply, and in accordance with the most authoritative forecasts, petroleum is the main source of energy (Table 1) (3). More than 95% of this petroleum is imported. The European Community's dependence on a highly unstable and politically volatile market is great even now and will increase in the future.

For structural and economic reasons coal consumption is not expected to increase and nuclear energy, if confined to the production of electricity, will be unable to cover more than a small fraction of the energy market in the short term (by 1985 about 4% of the energy delivered to the consumer is expected to come from nuclear plants).

Natural gas entered the picture fairly recently. Owing to its intrinsic merits it tends to replace oil in stationary applications, the degree of substitution depending chiefly on its availability. The elasticity of a system in accepting natural gas is shown by Fig. 1, which shows the increase in gas consumption in The Netherlands following the discovery of the Groningen gas fields. It shows that, where gas is available, it soon comes to supply virtually all stationary installations.

Europe's oil and gas reserves are given in Table II (4).

Although Europe clearly cannot rely for its energy supply on these sources alone, *proven gas reserves are equivalent to about five years' total oil consumption at the present rate.*

## Natural gas as a fuel

Natural gas is a premium fuel for stationary installations, as is shown by its rapid and wide acceptance by the consumer whenever it is available.

Availability actually seems to be the only limit to its widespread use.

The advantages of methane as a fuel are the following:

1. It is the least polluting of hydrocarbons, because it contains no sulphur and burns cleanly (Table III) (5).
2. It needs simple burners, and furnaces can operate unattended for long periods.
3. Distribution is via underground pipes which are fixed and unobtrusive, while oil is distributed chiefly by road tankers, greatly contributing to traffic congestion.
4. Distribution costs for large quantities tend to militate in favour of methane.
5. No stocks are needed at the consumer site.

Methane can also be used to fuel vehicles. In Italy about 50 000 cars run on this gas (chiefly for tax reasons), which is carried in heavy steel cylinders. The use of fibre-wound plastic containers would be a great improvement.

Buses and trucks using LNG (liquefied natural gas) are in experimental service in Italy, Switzerland and Germany (6). Advances in low-temperature insulation have made losses due to boil-off quite negligible (< 1% per day).

The point is, however, that a perfectly usable, if not perfect, technology exists for running cars on natural gas. It may become compulsory to use natural gas in towns as an anti-pollution measure, as has been proposed for the Los Angeles area.

## SNG Plants

Plants to produce SNG from an oil feedstock (mainly naphtha fractions) were originally developed by the British Gas Council (1) and are now licensed around the world. As the market evolved, the British Gas Council extended the scope of its SNG processes to use all petroleum fractions as feedstock, and even straight crude.

Table IV gives an economic comparison of these plants in relation to the

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The opinions expressed in this paper are those of the authors and are not meant to reflect those of the Commission of the European Communities.

CESARE MARCHETTI, CARLO RINALDINI, ALBERT SCHNEIDERS - *Joint Research Centre* of the Commission of the European Communities, Ispra Establishment

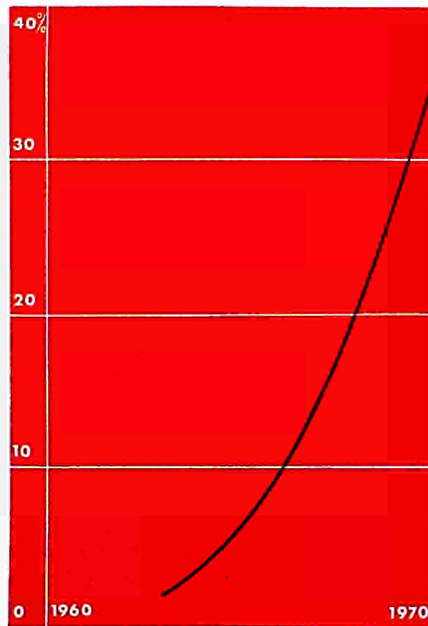


Fig. 1: Gas consumption as a percentage of total energy consumption in the Netherlands.

- 2) – efficiency, defined as the ratio between the calorific values of products and feedstock of an oil/SNG plant and a refinery, is very similar;
- 3) – SNG comes out sulphur-free; this is facilitated by the complete breakdown of the feedstock molecule.

Efficiency and capital cost at present look less good for the heavy fractions (8), with a break-point at a boiling point of 300 °C (see Fig. 4), above which capital costs increase by a factor of almost three, and energy losses almost double. This is chiefly due to the primitive state of SNG technology for this grade of feedstock, which explains why the data used herein relate more particularly to naphtha reforming.

feedstock used (8). It will be observed that the saving due to the use of cheaper feedstocks tends to be balanced by higher capital costs and lower conversion efficiency.

Two comments must be made on this table: (a) it is too schematic in that if, say, 10% of the heaviest fractions of the crude are used for other purposes (e.g. firing a power station), efficiency and economics would be greatly improved, and (b) processes handling crude are in an early stage of technological development.

It is important to observe that:

- 1) – the capital cost of an oil/SNG plant and of a refinery of similar capacity is roughly the same;

#### Distribution costs of natural gas vs oil products

It is not easy to assess the distribution costs of gas and oil on a really comparable basis owing to the inevitable variations involved, the variable “mix” of oil products, and geographical factors. The ideal situation for our purposes is to compare the cost of distribution from a refinery serving a given area, assuming that it either:

- (a) produces the usual mix of oil products, or
- (b) produces only SNG.

Distribution costs for a typical European situation estimated with the collab-

Table I: Primary Energy Consumption in the Communities (million tce)

	Solid fuel	Liquid fuel	Natural gas	Primary electricity*	Total
1960	476 (64%)	214 (29%)	13 ( 2%)	39 ( 5%)	742
1965	453 (48%)	423 (45%)	22 ( 2%)	40 ( 4%)	938
1970	368 (31%)	653 (56%)	88 ( 7.5%)	55 ( 4.7%)	1 164
1975	292 (19.6%)	905 (61%)	203 (13.6%)	88 ( 6%)	1 488
1980	267 (14%)	1 165 (62%)	296 (16%)	163 ( 8.6%)	1 891
1985	246 (10%)	1 459 (61%)	381 (16%)	292 (12.5%)	2 378

\* Primary electricity is expressed in the tons of coal equivalent (tce) which would be necessary to generate it in conventional power plants.



Table II: Natural Gas and Oil Reserves of the European Communities (1972)

	Gas (10 <sup>9</sup> m <sup>3</sup> )			Oil (10 <sup>6</sup> t)
	Confirmed	Estimated	Total	Confirmed
Netherlands	1 990	342	2 332	37.3
United Kingdom	700	300	1 000	686.9
West Germany	202	72	274	81.6
Italy	180		180	32.9
France	215	85	300	12.8
Denmark		50	50	33.3
	3 287	849	4 136 (= 4 550 million tce)	885 (= 1 248 million tce)

Table III: Pollutants from oil and gas-fired equipment (kg per ton of fuel)

	Fuel oil	Natural gas
Sulphur oxides (as SO <sub>2</sub> )	30	—
Nitrogen oxides (as NO <sub>2</sub> )	13.5	6.9
Organic acids (as CH <sub>3</sub> COOH)	13.5	1.3
Aldehydes (as HCHO)	1.3	1.0
Other organics	4.6	1.4

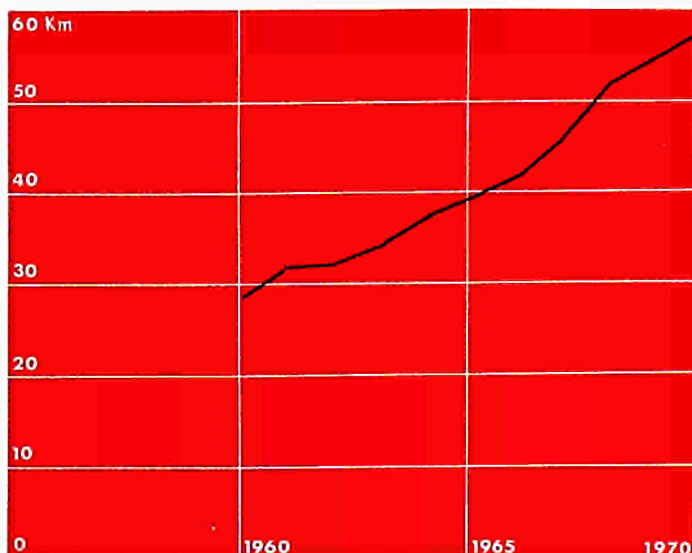


Fig. 2: Length of gas transport pipelines in the six original member countries of the European Communities.

oration of oil and gas companies (9), are given in Table V.

They are clearly of the same order, with natural gas having the advantage when consumption is increasing because the distribution networks are designed with substantial overcapacity.

### SNG as the main fuel

The foregoing data led us to consider the possibility of using SNG in the place of oil products, with SNG plants gradually replacing refineries and the gas transport and distribution network progressively extended. Our analysis is a first-order approximation aiming at evaluating the capital costs and the relaxation time of the system.

On the basis of the data in Table I, refinery capacity in Europe is expected to increase by 5% per year over the next ten years. The lifetime of equipment, excluding the infrastructure and general facilities, has been conservatively set at about 10 years.

On these assumptions, and if SNG plants are built as part of the natural replacement and extension of refinery capacity, a major fraction of the energy supply can be provided in the form of SNG within a few years (Table VI).

Assuming that SNG plants are built on the same sites as existing refineries (certainly not an optimum assumption), the large-bore pipeline network will increase during those years by about 2 000 km/year. In view of the rapid extension of the gas distribution network in Europe in recent years (Fig. 2), pipeline production and laying capacity is not expected to be overstretched.

Thus, within the limits of our approximation, the gradual replacement of an oil/oil products distribution system by an oil/SNG distribution system can be carried out at substantially nil differential cost.

We have not included here the cost of conversion, e.g., of burners, at the consumer's end because from previous experience with natural gas it is assumed that the consumer reaps a number of marginal benefits which compensate for the expense of the substitution, e.g., simpler equipment, no need to preheat the fuel, no storage tanks and stocks, and automatic delivery.

### An energy supply strategy based on natural gas and SNG

The fact that 95% of our oil is at present imported shows how great is the European Communities dependence upon the Middle East, and it will increase in the future with the evergrowing demand for oil. The threat of a sudden interruption of supplies is a real possibility and, if it materialized, would be very dangerous to the Communities. Moreover, the threat can be successfully used to push up the price of oil. The recent increases in crude oil prices showed how this works in practice.

True, Europe now has mandatory oil stockpiles of two to three months' requirements. These stocks are tactically important as they provide cover against a strike by the oil carriers, for example, or similar short term possibilities. It can easily be demonstrated, however, that they have no strategic importance, because they have been paid for and therefore no harm can be inflicted on the exporting countries by using them. These countries will actually benefit from the holding of large oil stockpiles by the Communities, since they can invest the proceeds and because no effective measures, with the possible exception of war, could be taken within such a short time.

In our opinion a really important means of placing Europe in a strong bargaining position vis-à-vis the exporting countries would be to enable it to withdraw from the oil market for a substantial period, e.g., five or ten years. This could be done by a strategy based on the following measures:

- (a) Compulsory construction of SNG plants instead of oil refineries to provide new and replacement capacity.
- (b) Use of European gas fields as strategic reserves. The rate of depletion should be rapidly cut to zero, with proper compensation for owners.
- (c) Construction of an emergency trunkline system linking these gas fields to the gas transport network, with sufficient capacity to offset the shut-down of all SNG plants.

### The cost of the strategy

With reference to (a) above:

As emerged from the foregoing discussion, the differential cost of producing

Fig. 3: Trunkline (dotted) and emergency networks

Du = Dublin	Rt = Rotterdam	Lu = Luxembourg	Bo = Bordeaux
Gw = Glasgow	Gr = Groningen		Ag = Angers
Li = Liverpool		To = Turin	
Lo = London	An = Antwerpen	Mi = Milan	
Ko = Copenhagen	Ch = Charleroi	Ve = Venice	
Hm = Hamburg	Li = Lille	Ts = Trieste	
Hn = Hannover	LH = Le Havre	Cor = Cortemaggiore	
Fr = Frankfurt	Pa = Paris	Ge = Genoa	
Ka = Karlsruhe	Me = Metz	Fi = Florence	
St = Stuttgart	Ly = Lyon	Na = Naples	
Mü = Munich	Ma = Marseille	Ta = Taranto	
Kö = Cologne	To = Toulouse	Pa = Palermo	
	Lacq	Au = Augusta	

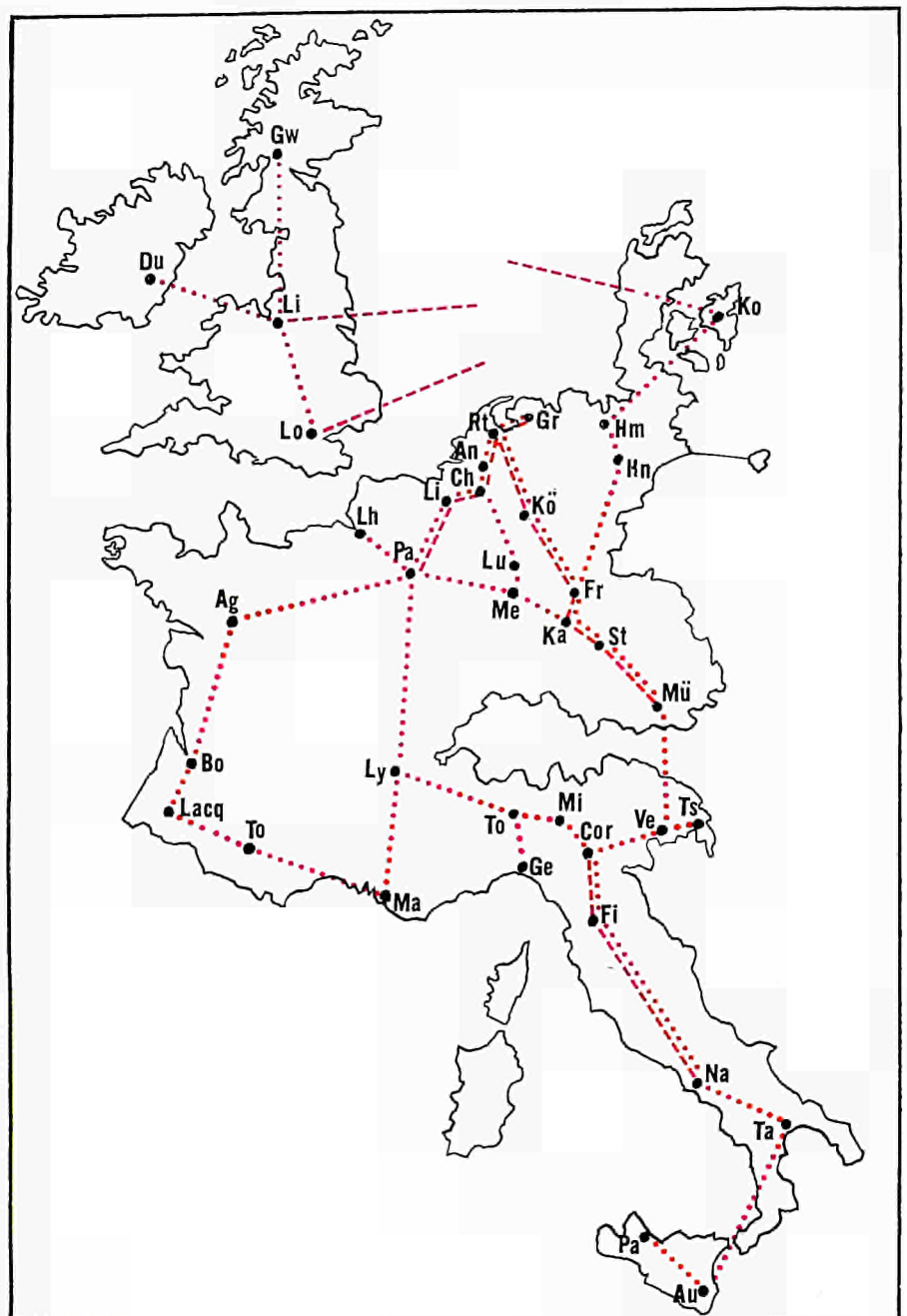


Table IV: SNG production cost factors for various feedstocks, mills/Nm<sup>3</sup> - Gas capacity 7 · 10<sup>6</sup> Nm<sup>3</sup>/day

Feedstock	Naphtha	Kerosine/ Light gas oil	Med/Heavy gas oil	Crude
Utilities, catalyst, etc.	1	1.1	1.3	1.5
Labour and related overheads	0.4	0.4	0.6	1.0
Capital charges	4.4	7.4	10.8	13.3
Total non-feedstock cost	5.8	8.9	12.7	15.8
Thermal efficiency, %	91	90	84	79
Investment \$ 10 <sup>6</sup>	70	70	105	130

Table V: Natural gas transport and distribution cost in a typical situation  
Transport cost (from port to city gates)

	Natural gas (by pipeline)	Oil	
		(by pipeline)	(by tank truck)
For an average distance of 300 km (\$/Tcal)	380	200	600

Distribution cost (from the city gates)

Use	Natural gas (\$/Tcal)	Heating oil	
		(\$/Tcal)	\$/t
Residential	2 100	1 500	15
Commercial	1 350	1 500	15
Industrial	200	120—500	1.2—5

Table VI: Energy consumption in Europe in 1982 under three different circumstances (10<sup>6</sup> tce)

CASE I		CASE II		CASE III	
Solid fuel	258	Solid fuel	258	Solid fuel	258
Oil	1 275	Oil	271	Oil (North Sea)	200
		SNG from oil	1 004		
Nat. gas	327	Nat. gas	327	Nat. gas	1 040
Prim. el.	206	Prim. el.	206	Prim. el.	206
	2 066		2 066		1 704

Case I — Normal conditions. Derived from Table I.

Case II — Oil-refineries begin to be replaced by SNG plants in 1975 under the assumptions set out in the text.

Case III — As in Case II, but oil imports are cut off starting from 1 January 1982 and energy consumption is reduced by 20%. Oil from the Community controlled zones of the North Sea will be used entirely for transport.

and distributing the SNG substituted for oil products can be considered zero, so that no cost should be charged to the strategy on that score.

With reference to (b) above:

If the gas reserves are "frozen" as strategic reserves, a scheme has to be devised to compensate their owners and keep up the level of exploration. A very straightforward, unsophisticated form of compensation would be for the Community to buy the gas at the rate at which it would have been extracted, e.g., 3% per year of the proven recoverable reserves.

We estimate that compensation on this basis would amount to some \$2 × 10<sup>9</sup>/year. The money is not really lost but in a sense frozen, since it will be recovered when the gas is finally used.

With reference to (c) above:

A possible emergency system is illustrated in Fig. 3. The estimated cost is around \$8 × 10<sup>9</sup> (Table VII). *The system is not optimized.*

Table VIII shows the total net cost of the strategy, assuming it to be operative for 20 years under the conditions set out in the table. As a rule of thumb this cost related to oil consumption is equivalent to an increase of about \$ 0.3 a barrel, which is certainly not negligible but should be compared with the recent increases of more than \$ 1<sup>(1)</sup> a barrel. In other words, if the only result of the strategy were to prevent a single increase of \$ 0.3 a barrel in the price of oil, it would pay for itself.

#### Possible Refinements of Basic Strategy

Since the most conservative boundary conditions were used, improved versions of the strategy can be devised. For example, Norwegian and Soviet gas, LNG imports, the conversion of coal to liquid and gaseous fuels, not to mention new gas discoveries in Europe or real optimization of the layout and operation of the gas grid, are beneficial factors in that they reduce the cost of implementing the strategy and extend the period during which the Communities can withdraw from the market.

— The importance of Norwegian and Soviet gas is obvious, and some details are given in Fig. 4.

<sup>1</sup> This article was written beginning 1973.

Table VII: Additional trunkline network needed in emergency conditions (end 1980)

	Pipeline length (100 km)	Additional capacity (Gcal/sec)	Cost of additional line (10 <sup>9</sup> \$)
Groningen-Amsterdam	1.8	158	2.1
Amsterdam-Rotterdam	0.68	120	0.58
Rotterdam-Köln	2.40	36	0.62
Köln-Frankfurt	1.83	25	0.32
Frankfurt-Karlsruhe	1.52	18	0.21
Karlsruhe-Stuttgart	0.76	19	0.10
Stuttgart-München	2.20	18	0.30
Rotterdam-Antwerpen	0.87	54	0.34
Antwerpen-Charleroi	1.08	24	0.18
Charleroi-Lille	1	32	0.23
Lille-Paris	3	18	0.37
North Sea-London	2.9	25	0.51
North Sea-Liverpool	3.6	37	0.96
North Sea-Danemark	3	7	0.15
Cortemaggiore-Florence	1.5	6	0.56
Florence-Naples	4.8	6	0.19
		Total	7.71

The capital cost of the pipelines and the cost of laying them and building pumping stations was estimated by the optimization procedure described in (10). However, no scale effect has been taken into account for flows over 10 Gcal/sec (corresponding to a maximum pipe diameter of about 1.7 m).

- LNG imports make SNG plants unnecessary for the relevant market share, although there is not much difference between the specific capital cost. An LNG tanker fleet would be a valuable asset, however, if the strategy were to cover all oil-importing countries (OPIC).
- If even part of the Community's 100 000 million tons of coal reserves could be converted to liquid fuel at competitive prices, it would also be of obvious importance.
- The relative location of gas fields and SNG plants is plainly an important factor in the total cost of the gas grid. A less obvious point is that, via the emergency trunklines, the gas fields can be used as a back-up or for daily and seasonal peak-opping. The SNG plants could then operate continuously at full capacity and without reserves. The resulting savings on capital and operating costs should be deducted from the cost of the emergency grid.
- Dividing Europe up into areas of

consumption and natural gas reserves, we find a fairly good match between them except for the south, which therefore requires very long large-bore emergency lines coming down from the north. These could be greatly reduced in size if they were used to boost local emergency reserves by slowly replenishing totally or partially depleted local gas fields, or even to feed man-made reservoirs, e.g., in aquifers. This would also facilitate high rates of gas supply to the emergency network.

### Conclusions

This analysis, although based on a first-order approximation, shows clearly that SNG technology can offer the same advantages as natural gas—low pollution, unobtrusive distribution and simplicity in use—and can do so with marginal cost differentials over the existing system. It also shows that SNG can rapidly attain substantial penetration of the energy market by taking advantage

Table VIII: Cost of strategy over 20 years

Cost of strategy (\$ 10 <sup>9</sup> )			Cost of an equivalent oil stockpile (\$ 10 <sup>9</sup> ) <sup>3</sup>				
Total discounted cost <sup>1</sup>	Annual cost <sup>2</sup>		Stockpile equivalent to the gas reserves <sup>4</sup>		Stockpile equivalent to year's consumption (1980) <sup>5</sup>		
Freezing reserves 11.4 Emergency trunklines 14.3	Constant rate	Rate proportional to consumption in 1980	Discounted	Annual	Capital	Interest (8%)	
Total	25.7	2.75	1.93	53.5	5.7	16.3	1.3

<sup>1</sup> The total cost of the strategy is the sum of:

- the economic penalty due to the deferred use of 3%/year of European gas reserves;
- the cost of an emergency trunkline network designed to meet European needs in 1980 and expanded at the rate of 5%/year to cater for the annual increase in consumption. These costs are discounted at 10%/year to the first year in which the strategy is in operation (1980). The value of the gas was worked out from a cost of \$ 20/ton for oil, as suggested in (11).

<sup>2</sup> The total costs are converted into a constant annual rate or an annual rate proportional to oil consumption (assumed to increase by 5%/year).

<sup>3</sup> As a comparison, the cost of an oil stockpile was calculated in two cases. Only the cost of the stockpiled oil (\$ 20/ton) was included and not the cost of the storage tanks; the latter will be about the same as the former, doubling the total cost.

<sup>4</sup> Total discounted cost and constant annual cost of an oil stockpile with the same energy content as Europe's gas reserves.

<sup>5</sup> The capital cost, and the interest on it, for an oil stockpile equivalent to annual consumption in 1980.

of the rapid obsolescence of refinery plant and of the expansion of the market itself.

An incidental bonus of an SNG-based strategy is that it provides a technically feasible way of acquiring a much-needed bargaining strength vis-à-vis the oil exporting countries.

The implementation of such a strategy should do much to calm the hysteria stemming from a sense of impotence that now characterizes the attitudes of the oil-importing countries, and the oil-exporting countries would certainly benefit, in the long term, if the former—so strong in so many ways—felt reasonably secure. The authors moreover realize that, for the actual implementation of an SNG-based strategy, a high degree of political coordination is necessary.

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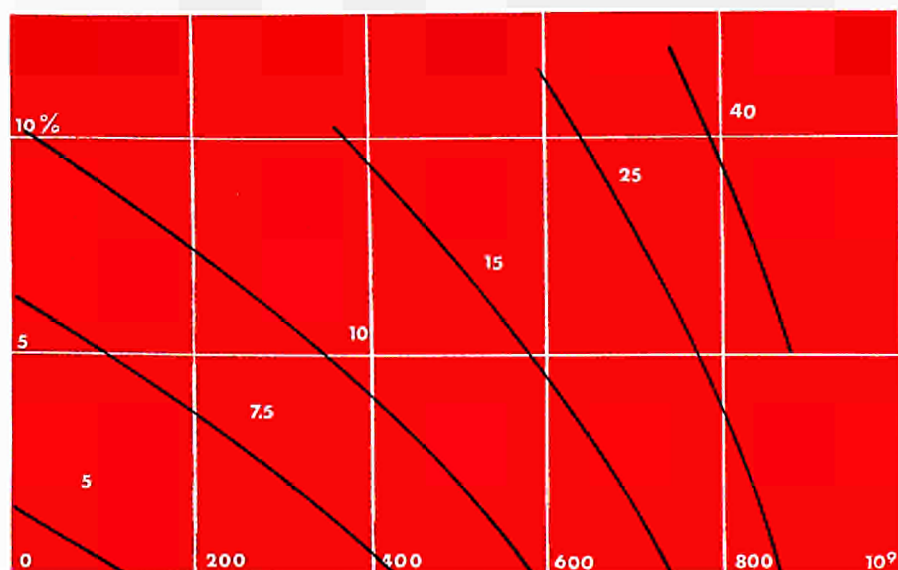


Fig. 4: Static index of European gas reserves (years). Number of years the reserves will last when used to replace all imports from 1982. On the absciss the natural gas import (m<sup>3</sup>/year); on the ordinate the annual increase in natural gas discoveries (%).

# Crystallogenesi

PIERRE BEUCHERIE, MARCEL PORTAL

## History of the crystal

SINCE TIME BEGAN, man has attempted to imitate the processes of nature; right up to the beginning of the twentieth century, the alchemist's dream was to transform lead into gold and to manufacture precious stones. In 1904, in a simple sentence from a report to the Academie des Sciences, Professor A. Verneuil, continuing the work of E. Fremy, announced "... from the chemical, physical and crystallographical point of view, the molecular structures and properties of a synthetic ruby and a natural ruby are identical". This fusion process achieves, from the scientific standpoint, true synthesis of the ruby. From this technique grew the industrial production of synthetic gems, precious stones created by man.

The art of the crystal therefore stemmed originally from the search for an artificial gem and the reproduction of these special stones provided so sparingly by Nature.

The gem was a symbol of power stretching from Egypt to Rome. In the Middle Ages it abounded in church treasures, reaching its apogee in the fifteenth century when precious stones (diamonds, emeralds, rubies, sapphires etc.) were enhanced by appropriate cutting.

Although the synthetic crystal, purer than the natural stone, is still used in the jewel form for which it was created, in modern physics it is valued as the perfect form of solid matter. The many studies carried out by solid state physicists and chemists led to extraordinary technical applications, particularly in the field of electronics and optics.

PIERRE BEUCHERIE, MARCEL PORTAL - Chemical Division of the *Joint Research Centre* of the Commission of the European Communities, Ispra Establishment, Italy.

Crystallogenesi, both the science and the art of "growing" crystals, was about to excel itself and produce crystals surpassing even those provided by Nature.

But these techniques, in order to develop, had to await either industrial outlets or a better understanding of crystal physics and, more particularly, from the thirties onwards, greater knowledge of the atomic concepts of crystallogenesi.

The increasing value of crystals in research led, in recent decades, to new growing processes which brought a number of improvements to the purity and structural perfection of the crystal.

The development of electronics, semiconductor devices, optical crystals transmitting *IR* and *UV*, crystal lasers, piezoelectric quartzes, ferroelectric memories, optoelectronics and holography are all problems solved during the evolution of crystal-growing techniques towards both known and new materials.

## Crystalline perfection (1)

Materials are solids, and solids are classed under two main headings: the crystalline solids which comprise the metals and most of the minerals in which the atoms are stacked in more or less regular designs, and the amorphous solids (e.g., glass) in which they are distributed at random. The properties and structure of the crystal depend on the way in which the atoms will assemble, not chaotically but lying in regular networks following a strict pattern, each atom being surrounded by a certain number of others at specific distances and in definite directions. The geometrical form may be simple or complex, but is astonishingly regular.

A common example is ordinary household salt, also known as sodium chloride or halite, where each sodium

atom is surrounded by six chlorine atoms and conversely, each chlorine atom is surrounded by six sodium atoms. Fig. 1 shows the basic structure of such a crystal; the structure contains all the physical, chemical and geometrical characteristics of the mineral.

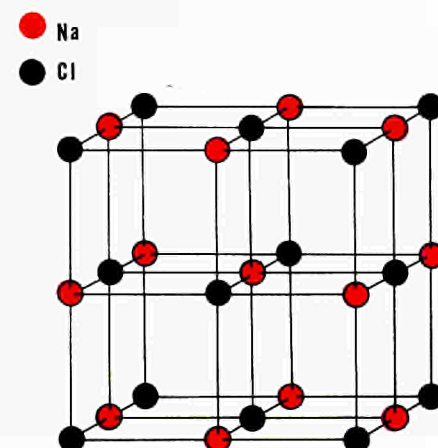
Not all crystals have this very simple form. Fig. 2, for instance, shows the more complex rhombohedral structure of the basic lattice of calcium carbonate (calcite), of which Iceland spar is a curious and rare form.

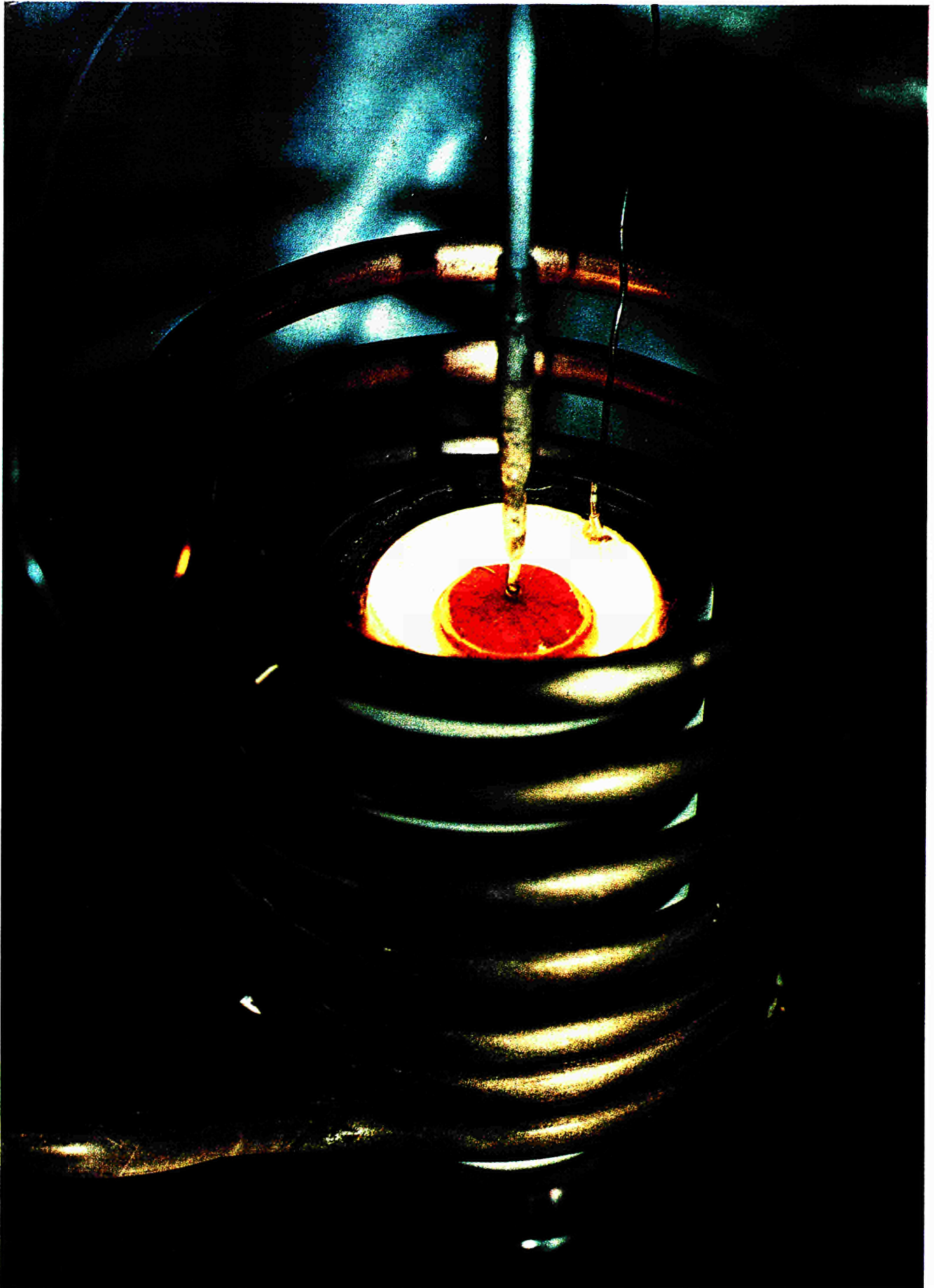
This natural order and symmetry of form attracted the attention of scientists who sought to understand and classify the phenomenon.

With the advent of X-rays, crystallography studied the laws governing the formation of crystals and attempted to explain their morphology. In 1874, Haüy laid down the basic principles; in the nineteenth century Bravais perfected the concept of the crystalline structure, while the fundamental experiment carried out by von Laue in 1912 on X-ray diffraction by a zinc sulphide crystal confirmed that the crystalline state consists of spatial networks whose nodes are represented by atoms or ions.

Using the symmetry factors of the crystal as a basis, an attempt was made to sketch out a classification of the various minerals. They were reduced to seven crystalline systems for which crystallographical axes were defined in order to identify and locate the position of the crystal faces in relation to their centre. Axis *x* faces the observer, axis *y*

Fig. 1: Basic lattice of a sodium chloride crystal (rock salt).





is to the right and axis  $z$  is upwards (Fig. 3).

To determine the position in space, occupied by a crystal face, the three parameters defined by the intersection of the face with the three crystallographic axes are measured; the resulting values are proportional to the distances between the nodes.

In 1839 Miller proposed a useful method, now universally adopted, for defining a family of reticular planes. Fig. 4 shows the positioning of several principal planes with their notation according to the Miller indices.

The presence of an index 0 in the notation of a face indicates that the latter is parallel with the corresponding axis: (1.1.0) is a face parallel with the  $z$  axis; (0.0.1) is a face parallel with both the  $x$  axis and the  $y$  axis, parallel to plane  $xy$ .

#### How crystals grow

Natural crystals are slowly formed by the fractionated crystallization of cooling lava, lava rich in all types of minerals.

Must we therefore wait centuries, perhaps thousands of years, to develop a crystal? Fortunately, even when trying to imitate nature, we need only a few hours or, at the most, several weeks to grow a crystal to a tangible size.

To obtain a crystal from a disordered (amorphous, liquid, gaseous) atomic state, the atoms must first be enabled to move and hence to rearrange themselves correctly. This reorganization into solid crystal can be done from the vapour state, the liquid state or the solid state.

R.A. Laudise (2) has proposed a classification of growth methods which summarizes most of the crystallogenic processes used today to synthesize single-component (Table I) or multi-component crystals (Table II).

We only have room here for a very brief outline of the principal techniques in use. We will briefly describe the commonest of these, omitting special or refined techniques.

There are two types of growth methods:

- a) rapid growth;
- b) slow growth.

Rapid growth methods generally operate at high temperatures which produce large crystals, frequently of inferior

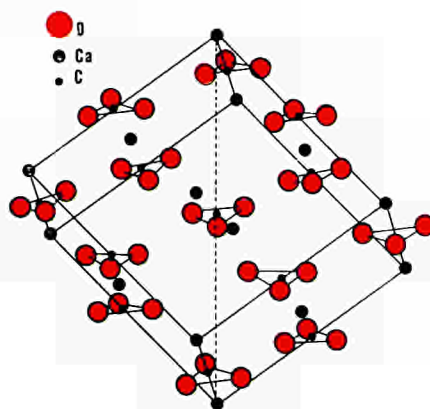


Fig. 2: Basic lattice of calcite (calcium carbonate) with a more complex structure (rhombohedral system).

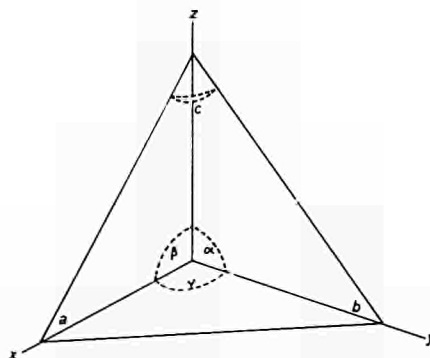
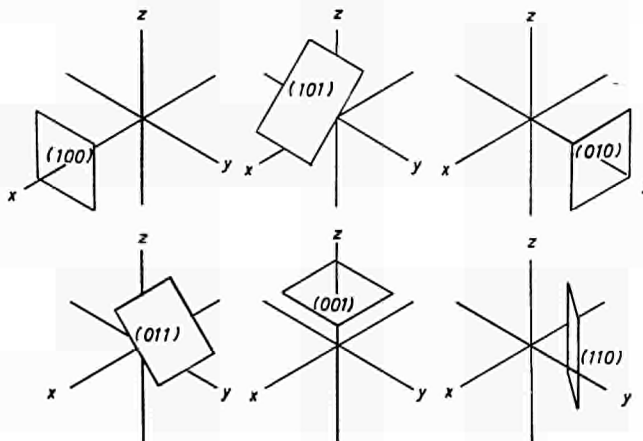


Fig. 3: The crystallographic axes serve to identify and locate the position of the crystal faces in relation to their centre. Axis  $x$  faces the observer, axis  $y$  faces to the right and axis  $z$  faces upwards.

Fig. 4: Miller indices of the principal crystallographic planes: position of several main planes and their notation.



quality (high density of dislocations, irregular lattice, non-homogeneous composition) requiring annealing. In general these methods consist in directed growth from a molten bath (Bridgman, Czochralski, Kyropoulos, Verneuil, zone fusion, etc.).

The slow-growth methods operate at low temperatures. They are required when the crystal to be obtained would dissociate at high temperature owing to non-congruent fusion. The crystal produced is usually of good quality and stress-free, but it can be contaminated by the solvent or the material of the reaction vessel. In general these are methods of growth in solution, using aqueous, organic and inorganic solvents, fused salts and hydrothermal growth.

#### Bridgman-Stockbarger method (3)

This method consists in principle in lowering a melt bath slowly in a temperature gradient, bringing the bath from the liquid to the solid state (Fig. 5).

The upper part of the furnace is kept at approximately 50-80 °C above the melting point of the compound, while the lower part is at 50-80 °C below the melting point of the compound. A platinum screen between the two parts of the furnace stops radiations along the tube and increases the temperature gradient. A gradient of 25 °C per centimetre is usually suitable.

The base of the crucible is conical, the point entering first into the cold zone; in this way only a small number of crystal



Table I: Method and technique of growing single-component crystals.

- 
- a) **SOLID-SOLID:**
1. recrystallization by annealing;
  2. devitrification;
  3. polymorphic transformation by changing the sintering phase.
- b) **LIQUID-SOLID:**
1. *conservative techniques* (the bath is not modified):
    - a) directional solidification or controlled crystallization (Bridgman and Stockbarger method);
    - b) use of cooled seed (Kyropoulos method);
    - c) extraction of crystal during growth (Czochralski method).
  2. *Non-conservative techniques* (matter may be added or removed from bath):
    - a) zone fusion method (melting of horizontal, vertical and floating zones);
    - b) Verneuil process (fusion by flame, plasma torch, arc image furnace and solar furnace).
- c) **VAPOUR-SOLID:**
1. Sublimation, condensation;
  2. Cathodic pulverization.
- 

Table II: Method and technique of growing crystals with several components.

- 
- a) **SOLID-SOLID:**
1. Precipitation of a solid solution.
- b) **LIQUID-SOLID:**
1. *Growth in solution through evaporation, slow cooling* (effect of thermal gradient):
    - a) aqueous solvents;
    - b) organic solvents;
    - c) fused salts;
    - d) solvents in hydrothermal condition;
    - e) other inorganic solvents such as molten metals.
  2. *Growth by reaction* (thermal gradient, concentration gradient):
    - a) chemical reactions;
    - b) electrochemical reactions.
- c) **VAPOUR-SOLID:**
1. *Growth by reversible reaction* (thermal gradient, concentration gradient):
    - a) Van Arkel (hot wire) process.
  2. *Growth by irreversible reaction:*
    - a) epitaxial growth.
- 

seeds are formed, and this favours the initiation and growth of the crystal in single-crystal form.

The apparatus must be thermally and mechanically stable to ensure a flat, horizontal surface at the bath-crystal interface. If the solid/liquid interface becomes concave, e.g., when the lowering

speed is too rapid, the ingot obtained is usually polycrystalline.

Growth speeds of 1-10 mm an hour are commonly obtained.

This method has found many uses in the growing of crystals of alkaline halides, calcium fluoride ( $\text{CaF}_2$ ) and lithium fluoride ( $\text{LiF}$ ); as the work can be done

in a pressure-sealed crucible, the method is particularly suited to the forming of numerous semi-conductor compounds which cannot be melted without dissociating, e.g., sulphurs and selenides of lead, tin, cadmium, zinc ( $\text{PbS}$ ,  $\text{PbSe}$ ,  $\text{SnS}$ ,  $\text{ZnSe}$ ,  $\text{SeCd}$ ) and the lead and cadmium tellurides ( $\text{PbTe}$ ,  $\text{CdTe}$ ) (4).

A slightly different method was used at Ispra to grow crystals of a gold/silver alloy (Fig. 7) and calcium fluoride.

As Fig. 6 shows, the graphite crucible, no longer supported by the base, hangs from a wire; it is slowly lowered through the coils of a high-frequency induction heater.

The thick-walled graphite crucible is thus brought to a high temperature in the induction zone. It cools gradually as it leaves this zone and is low enough to be unaffected by the induction heating.

#### Zone fusion

Zone fusion, developed by Pfan towards 1952 and mainly considered as a method of purifying materials, lends itself to crystal growing and has produced high-quality crystals (Fig. 8).

The molten zone travels along the ingot starting from an oriented and cooled seed. This method makes it possible to control the stoichiometry and reduces the risk of contamination by the crucible. For maximum purification, the molten zone should be as narrow as possible. The heating conditions required for this purpose can be met by using nichrome or "KANTAL" for low-melting-point materials and, at higher temperatures, high-frequency induction heating or an electron beam. This method is used to grow a number of metal crystals with a melting point of not more than 1500 °C: antimony, tellurium, tin, arsenic, cobalt, nickel, germanium, etc.

The floating zone method (Fig. 9), proposed by Keck and Golay in 1953, developed the growth of single-crystals of high-melting-point elements, e.g., refractory metals.

The molten zone created in the vertical ingot is supported by the liquid's surface tension forces, which effect a kind of natural levitation of the molten zone.

In this case, there is no crucible to be a source of contamination; the material is purified by zone refining and by the evaporation of certain volatile impurities.

Although the method is difficult to operate, it provides very pure crystals, particularly metal crystals such as vanadium, niobium, molybdenum, tungsten, rhenium and tantalum, or metal alloys of these metals (Mo/Re, Ta/Nb, Mo/W).

A particular use of zone fusion, developed at Ispra, is in the preparation of uranium carbide single crystals (5). Owing to the high reactivity of this material in the molten state, we used the technique of core fusion by induction heating in a self-crucible (Fig. 10).

The outside of the rod remains sufficiently cool to be solid, while in the centre a molten zone develops which travels from the top to the base of the rod. This self-crucible fusion is made possible by the temperature distribution inside the material. It is the result of thermal balance between the induced heat which develops in the "skin layer" and the radiated heat which cools the external surface. By selecting a suitable frequency, the depth of action of the induced currents can equal the radius of the treated rod and operate radially to the centre. Here, then, the induced currents do not act on the skin layer but have a deeper action capable of melting the core.

Fig. 11 shows a single crystal of a few cubic centimetres of uranium monocarbide extracted from the treated rod.

#### Extraction method:

#### Czochralski's method (6)

This is one of the methods most commonly used to produce a number of single crystals. An oriented crystal seed is plunged into the molten bath. The bath solidifies on the slightly colder seed; the crystal formed is extracted by lifting it slowly upwards. The growth of the crystal's diameter can be controlled by varying the temperature of the bath or the speed of extraction. Although a crucible is used, the solid/liquid interface never comes into contact with it. Proper designing of the crucible shape enables the radial temperature gradient to be reduced.

The interface must be flat, in order to avoid stresses which would greatly increase the density of the dislocations. Heat losses due to radiation through the crystal surface are balanced by additional heating in this area.

We will describe this method, applicable to a number of crystals, by reference to the development work we did at Ispra in connection with the production of single crystals of lithium niobate ( $\text{LiNbO}_3$ ), which because of its ferroelectric properties is of great value as a component in optical data-processing and electronics.

A diagram of the extraction apparatus designed in our laboratory is given in Fig. 12.

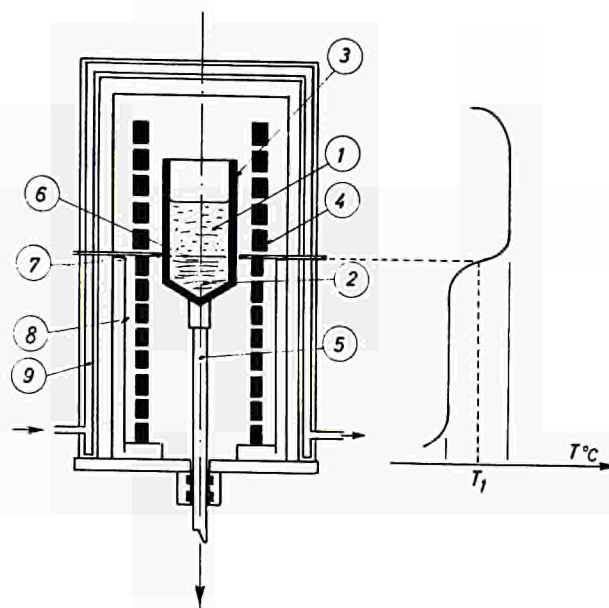


Fig. 5: The Bridgman and Stockbarger growth method.  
 1: hot zone, liquid bath;  
 2: cold zone, solid single crystal;  
 3: crucible;  
 4: heating elements;  
 5: crucible lifting and support rod;  
 6: solid/liquid interface;  
 7: thermal screen;  
 8: refractory lining;  
 9: cooling water;  
 $T_1$ : melting point of compound.

The upper part comprises the extraction head which, attached to the seed-holding shaft that enters the furnace, enables the crystal to be removed from the molten bath.

The lower part consists of the heat-insulated crucible, heated by high-frequency induction. Above it is an auxiliary furnace for annealing the crystal. The device can be operated equally well in the ambient atmosphere, under inert gas and under vacuum.

The crystal's perfection often depends on the mechanical perfection of the extraction device. The slow displacements produced by screw-and-nut systems set up intense friction stresses.

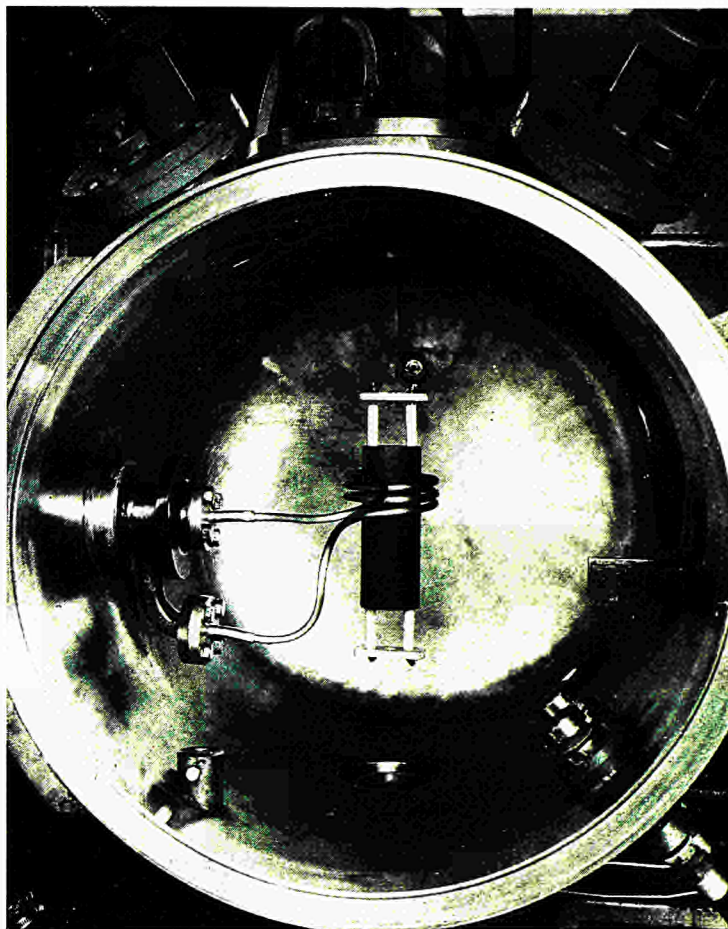
These stresses, which begin on the extraction rod, are released at sudden intervals; the resulting vibrations pass along the seed-holding shaft to the solidification interface and cause dislocations. In order to prevent this, we have designed a balanced extraction head, where the traction stress is small and constant and is limited to the crystal-lifting effect. The seed-holder rises slowly and regularly about 5 mm/h and rotates at 30-50 rpm. Excessive extraction speed results in the formation of bubbles in the crystal.

Fig. 13 shows a  $\text{LiNbO}_3$  crystal growing, while Fig. 14 shows a single crystal boule which is completely transparent, colourless and flawless.

At the present time, thanks to its ease of operation, its profitability and the satisfactory results it has provided, the Czochralski extraction method is in widespread use for the production of crystals of silicon and germanium semiconductors and the principal materials for solid lasers:

- Corundum ( $\text{Al}_2\text{O}_3$  doped with Cr);
- Doped garnets ( $\text{Y}_3\text{Al}_5\text{O}_{12}$ ,  $\text{Y}_3\text{Ga}_5\text{O}_{12}$ ,  $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ );
- Doped tungstate, calcium molybdates, strontium, lead ( $\text{CaWO}_4$ ,  $\text{SrWO}_4$ ,  $\text{CaMoO}_4$ ,  $\text{SrMoO}_4$ ,  $\text{PbMoO}_4$ );
- Alkaline-earth fluorides ( $\text{CaF}_2$ ,  $\text{SrF}_2$ ,  $\text{BaF}_2$ ).

Certain semiconductor single crystals and optoelectronic compounds ( $\text{GaAs}$ ,  $\text{InP}$ ,  $\text{ZnTe}$ ,  $\text{ZnSiP}_2$ ) are difficult to obtain owing to dissociation of the bath during growth. This can be prevented by using the liquid encapsulation technique, which consists in placing the bath and the seed-holder in a sealed capsule.



Extraction is effected by magnetic coupling between the seed-holding shaft in the capsule and the extraction head outside the capsule.

Similarly, a special method developed by J.B. Mullin (7) consists in preventing thermal dissociation by pressure balancing with inert gas.

Among the most recent achievements in this field, mention must be made of the curious performances and the high degree of technicality obtained by Chalmers and Labelle (8) in the high-speed extraction (up to 2 cm/min) of sapphire crystals in such complex and various forms as films and round or square section tubes.

#### Verneuil or Flame-Fusion Process (9)

This process born, as we have said, at the beginning of this century, has

Fig. 6: Bridgman method used to grow an Au/Ag crystal (original photograph).

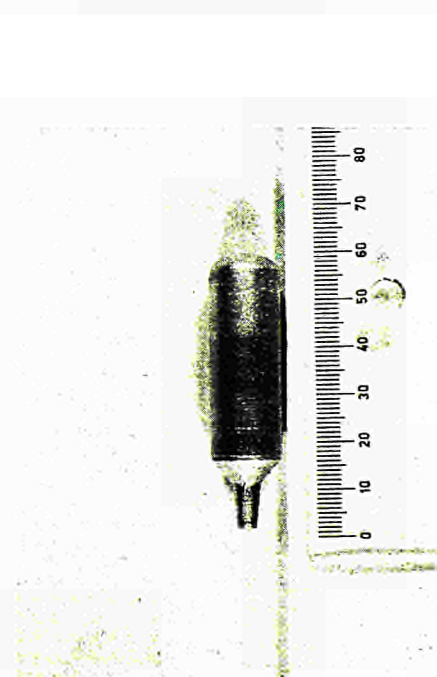


Fig. 7: Au/Ag crystal (original photograph).

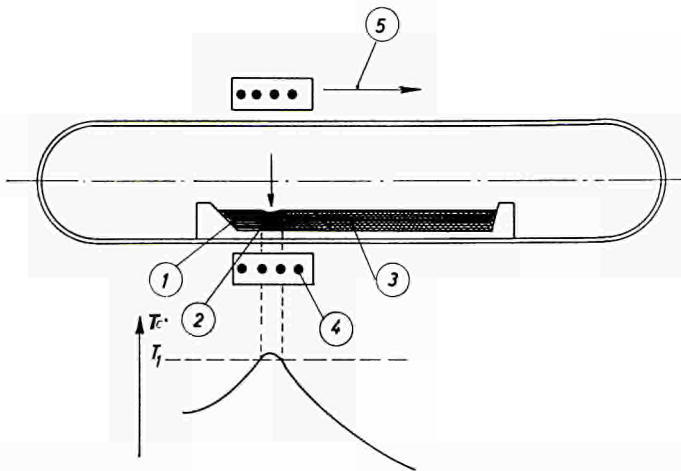


Fig. 8: Growing a crystal by zone fusion. 1: single crystal; 2: melted zone; 3: polycrystalline rod; 4: heating device; 5: zone movement;  $T_1$ : melting point of compound.

remained virtually the same to this day. Its use is important in the production of corundum ( $Al_2O_3$ ) for clock-making, synthetic gems (rubies, sapphires), spinels ( $MgO$ ,  $Al_2O_3$ ) used as supports for thin-layer epitaxy, and rutile ( $TiO_2$ ). The DJEVA company, established in Switzerland and equipped with 1 440 blowtorches, manufactures 300 000 000 carats of synthetic gems annually, which represents approximately 60 tonnes of material.

Alumina powder, with a perfectly defined grain size and of very high purity (obtained by calcination of ammonium alum, tinted by adding metal oxides), is melted towards  $2\ 050\ ^\circ C$  in the flame of an oxyhydrogen torch. The admission of  $Al_2O_3$  powder entrained by the oxygen current is not continuous; it is controlled by tapping with a mechanically-actuated hammer, the tapping frequency of which is adjustable. The powder fuses in the central part of the flame at the tip of the torch and accumulates on the seed. The lower part of the seed is supported by a ceramic shaft which is slowly lowered as the crystal grows and lengthens. A muffle composed of a refractory casing surrounds the growing crystal and protects it from thermal shocks (Fig. 15).

The growth time for a ruby boule 20 mm in diameter and 5 cm long is of the order of four hours.

In order to eliminate the drawbacks inherent in the composition of the flame atmosphere, several variations of this

process were tried, namely, the radio-frequency plasma torch, the arc image furnace and the solar furnace for the growth of highly refractory  $MgO$ ,  $ZrO_2$  oxide crystals.

#### Growth method in vapour phase

Two principal methods are used to grow single crystals in a vapour phase:

- growth by condensation of the vapour of a sublimated or evaporated material;
- single-crystallization by reaction to gaseous phase transport.

The first method, used quite commonly, has found a number of applications in the preparation of sulphide and selenide crystals ( $CdS$ ,  $ZnS$ ,  $SePb$ ). The second, although frequently used to obtain binary or ternary oxide single crystals or to prepare thin epitaxial layers and to purify certain metals, has only been developed to a certain degree owing to the small size of the crystals obtained.

It is based on the heterogeneous balance of the reaction between the

Fig. 10: Plan of self-crucible core fusion of a uranium carbide rod.

1: melted UC rod, UC single crystals; 2: auto-heater 10 000 Hz 50 kva.; 3: melted zone; 4: UC rod before melting; 5: solid external crust of the self-crucible; 6: refractory rod support; 7: device for slow translation of rod (1-5mm/h).

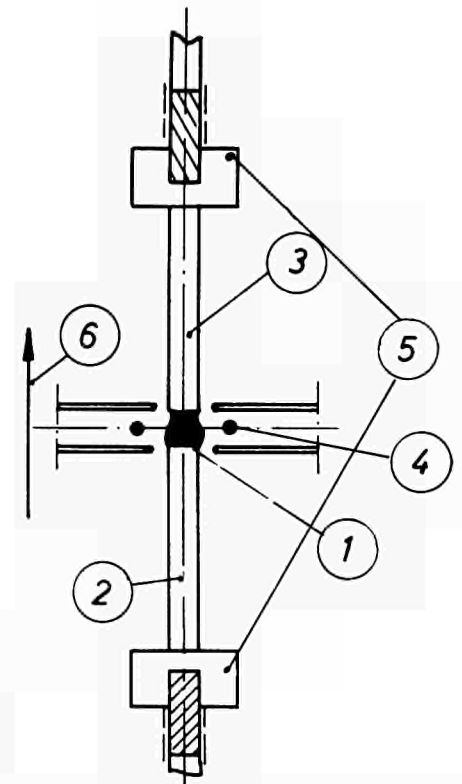


Fig. 9: Floating zone technique using electron beam.

1: melted zone; 2: single crystal; 3: polycrystalline rod; 4: heating filament for electron beam; 5: rod support clamp; 6: displacement of melted zone.

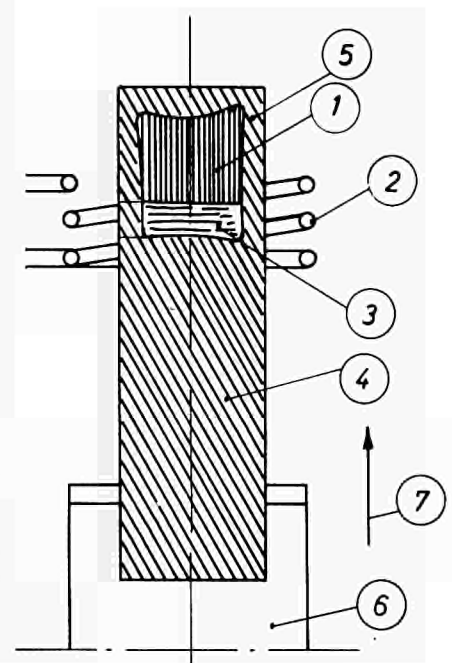


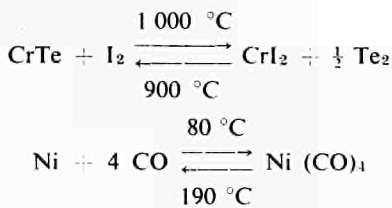


Fig. 11: Uranium monocarbide (UC) single crystal (original photograph).

solid and a chemical transport agent, usually a halogen.

The method is continuous, the gas flowing over the solid which is maintained at temperature  $T_2$  ...; at  $T_1$ , the reverse reaction occurs and the solid separates out in a sealed capsule in a furnace with two heating zones (Fig. 16).

Some practical examples:



This method has been used to prepare a number of crystals of binary chalcogenides (cobalt telluride:  $\text{CoTe}$ ; chromium telluride:  $\text{Cr}_3\text{Te}_4$ ; iron telluride:  $\text{Fe}_2\text{Te}_3$ ; iron disulphide:  $\text{FeS}_2$ ), and ternary chalcogenides ( $\text{CoCr}_2\text{Te}_4$ ,  $\text{FeLa}_2\text{S}_4$ ).

### Methods of growth in solution

These are based on crystallization by precipitation from a supersaturated solution. The solution can be supersaturated by various methods:

- gradual lowering of the solution temperature (very slow cooling);
- gradual evaporation of the solvent (growth at constant temperature, isothermal evaporation);
- constant addition of solute to compensate for its precipitation, or gradual addition of a reagent.

The first method is used for substances with sufficient variation of solubility in relation to the temperature. The speed

of crystallization is controlled and can be kept constant with a high degree of precision to within  $\pm 0.002\text{ }^\circ\text{C}$ . The programme of temperature reduction by  $0.01$  to  $0.1\text{ }^\circ\text{C}$  a day must be maintained without interruption and without deviation for several months. The crystals formed may develop stresses due to temperature changes.

The second method is used for any soluble material. Since the crystal grows at a constant temperature, it is free from

the stresses caused by temperature changes. On the other hand, it is difficult to maintain the rate of crystallization sufficiently accurately; the homogeneity of the solution depends on the diffusion and convection of the solute from the surface of the bath inwards.

One of the major drawbacks of this technique is the risk that the crystal, while forming, may capture traces of the solvent.

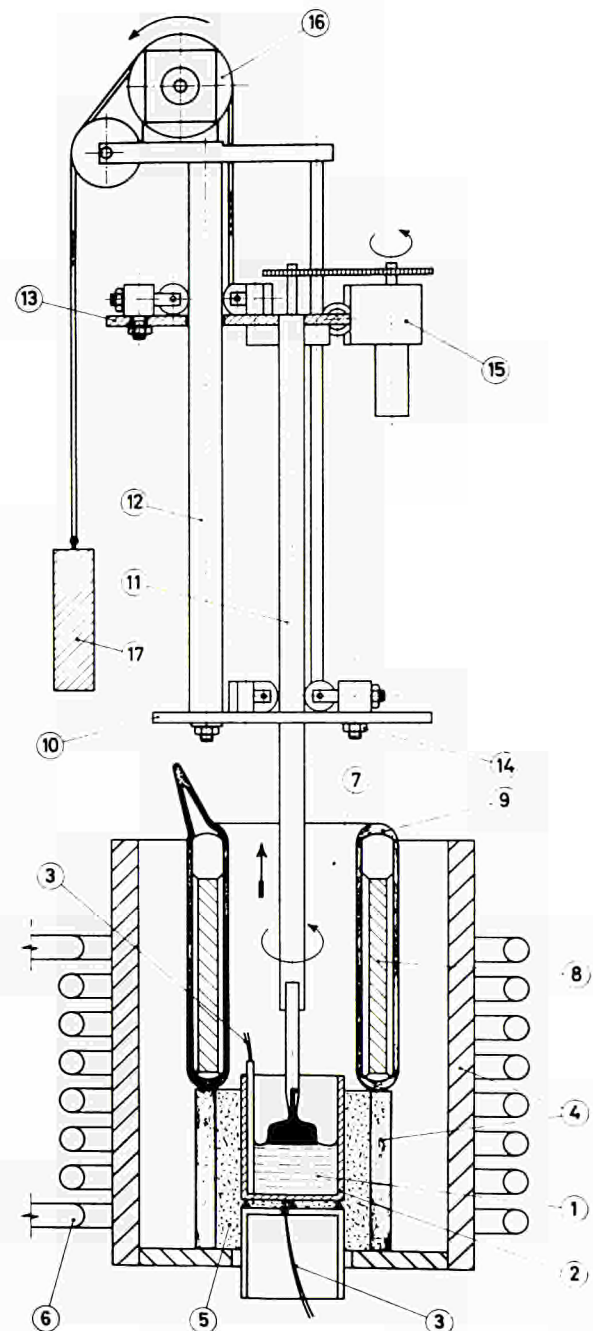


Fig. 12: Diagram of Czochralski extraction device. 1:  $\text{LiNbO}_3$  melt bath; 2: platinum crucible; 3: temperature-regulating thermocouples for bath; 4: heat-insulating refractory crucible; 5: filling of  $\text{Al}_2\text{O}_3$  powder; 6: auto-induction heater 750 kHz 12 kva.; 7: auxiliary heating furnace; 8: graphite susceptor of auxiliary furnace; 9: quartz casing of auxiliary furnace; 10: base-plate supporting extraction head; 11: support rod for seed-holding shaft; 12: extraction head support column; 13-14: ball-bearing device to guide extraction head; 15: drive motor for rotation of seed; 16: motor and gear box of crystal-lifting device; 17: counterweight.

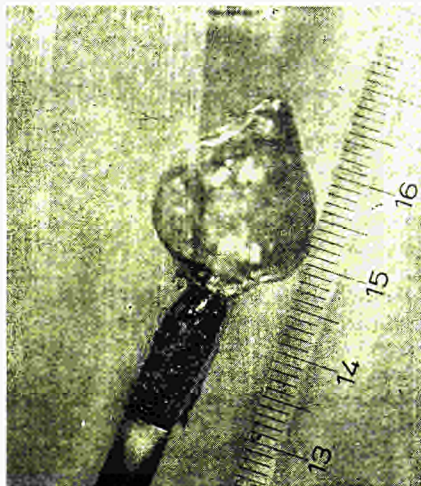


Fig. 13: A lithium niobate single crystal in growth (original photograph).

The two major applications are:

- 1) Crystallization in aqueous solution, used to produce such crystals as ADP, DKT, EDT, alkaline halides,  $\text{HIO}_3$ .
- 2) Crystallization in fused salts, also known as the fusion or flux method, principally employing the following solvents:
  - a) KF alkaline halides for barium titanate crystals ( $\text{BaTiO}_3$ ), LiCl

Fig. 14: Lithium niobate single-crystal boule (original photograph).



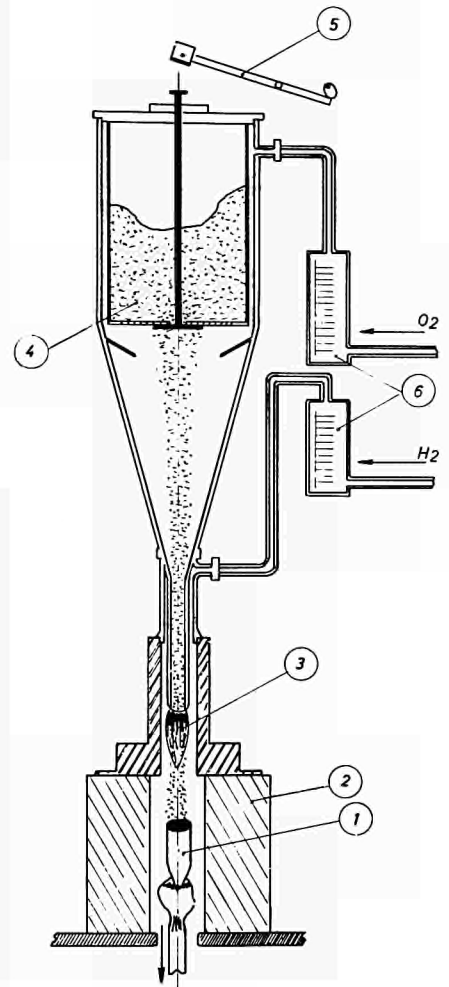
- b) alkaline tungstates and molybdates ( $\text{Li}_2\text{WO}_4, \text{MoO}_3$ ) for rutile ( $\text{TiO}_2$ );
- c) lead compounds  $\text{PbO}, \text{PbF}_2, \text{PbCl}_2$  for alumina  $\text{Al}_2\text{O}_3$ , yttrium garnets YIG ( $\text{Y}_3\text{Fe}_5\text{O}_{12}$ ), zirconia  $\text{ZrO}_2$ , nickel ferrites ( $\text{NiFe}_2\text{O}_4$ ), rare-earth orthoferrites ( $\text{YFeO}_3$ ), zinc oxide ( $\text{ZnO}$ ).

#### Hydrothermal growth method (10)

This old method, already in evidence at the beginning of the century, was an endeavour to imitate the conditions under which certain natural crystals developed. It is based on the fact that a large number

Fig. 15: Principle of the Verneuil flame fusion torch.

1: single crystal boule growing; 2: heat-insulating muffle; 3: tip of torch; 4: powder container; 5: powder feed device; 6: flow-meter measuring O and H gas input to torch.



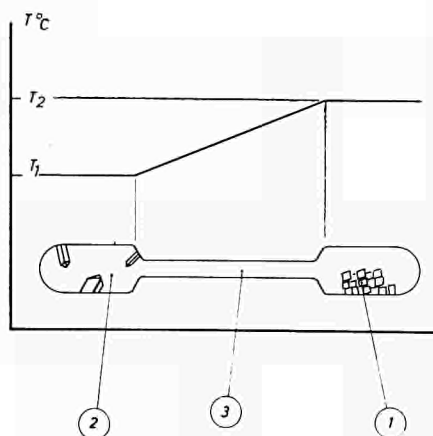


Fig. 16: Principle of transport-reaction crystallization.

1: polycrystalline compound; 2: single crystals forming; 3: diffusion zone;  $T_2$ : formation temperature;  $T_1$ : dissociation temperature.

of substances which are insoluble in water at a low temperature (below 100 °C) are soluble, or partially soluble, at temperatures above 300 °C. This technique has proved very useful in the production of quartz crystals and, more recently, of high-quality corundum and ruby crystals for laser work. Crystals obtained under these low-temperature conditions have fewer defects (dislocations, disorientation of reticular planes, internal stresses) than crystals obtained by fusion. Rutile crystals  $TiO_2$ ,  $GeO_2$ ,  $SiO_2$  have been prepared by recrystallization in a fluorinated hydrothermal solution.

Although rather slower than growth methods using a melt bath (several centimetres an hour), the hydrothermal method is nevertheless quick (about 1 mm/day) if compared with the methods of growth in solution (0.1 mm/day).

The method is relatively simple, but owing to the high pressures (up to 4 000 bars) involved at high temperatures (up to 500 °C), there is considerable difficulty in choosing materials for the autoclave, which should be made of special alloys (steels of grade z20 CN 17-2, nimonic).

Fig. 17 shows a diagram of an autoclave used to produce piezoelectric quartz. The lower part of the autoclave, in the dissolution zone, contains the parent body composed of fine quartz particles.

The upper portion, in the crystallization zone, contains the quartz single-crystal seeds, suitably oriented. A screen

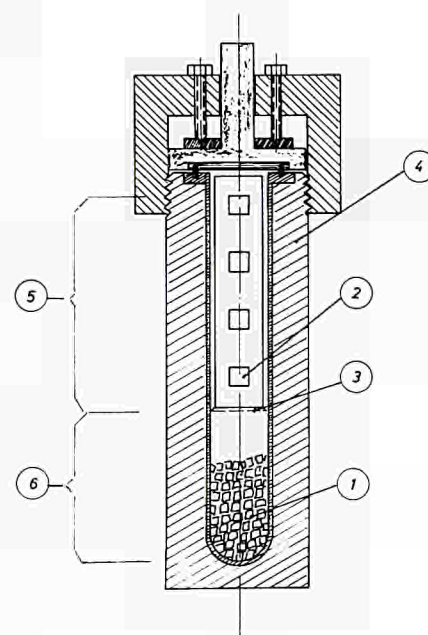
(a perforated metal disc) separates the two zones into two isothermal areas. The autoclave, filled as to 80% with a basic soda solution (NaOH 0.5 M), is placed in a furnace which gives the desired temperatures: 400 °C in the dissolution zone, 360 °C in the crystallization zone. The quartz parent body dissolves and saturates the solution. In the cooler upper part the supersaturated solution is deposited on the seeds. The convection currents set up by the temperature difference continually change the solution in contact with the parent body. This continuous cycle of solution and deposit produces large quartz crystals that may weigh up to 800 grammes.

### Conclusions

It would be fruitless to attempt, in a few pages, to draw a full picture of the art of crystallogenesi and its meteoric expansion in the last decade. One need only consider the many publications which have appeared in this field to realize the importance of the crystal as a key material in the development of electronics, optics, acoustics and magnetism and the value of its scientific and industrial uses.

Fig. 17: Hydrothermal growth of quartz crystals.

1: parent body; 2: single-crystal seed; 3: separation screen; 4: autoclave; 5: crystallization zone; 6: dissolution zone.



In thus describing so succinctly the principal methods of growing crystals (each technique now becoming a speciality in its own right), we have omitted two important chapters:

- How and why does the crystal develop? (modern theories of growth kinetics);
- What is the quality of the crystals? (i.e., is it perfect or does it contain a number of defects?)

We have seen that different processes have been used to produce a given type of crystal, and that they often produce crystals of differing quality. It is the user who, indirectly, decides which method should be employed, through his requirements as to purity, size, perfection and price.

Lastly, we should mention the wide range of new substances which have already been prepared, but whose characteristics have yet to be determined and whose properties need to be defined.

Despite the amount of work done by many laboratories, and the solutions provided to numerous specific cases, the problems to be solved before achieving the perfect crystal are still very complex.

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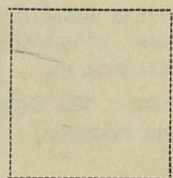
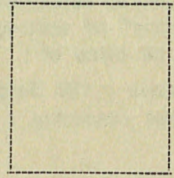
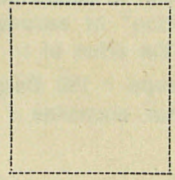
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