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FOLLOW UP ON THE CRYSTAL GROWTH EXPERIMENTS OF THE LDEF

K. F. Nielsen
Technical University of Denmark, Physics Laboratory III
DK-2800 Lyngby, Denmark.

M. D. Lind
Rockwell International Science Center
Thousand Oaks, California 91360.

The results of the 4 solution growth experiments on the LDEF have been published elsewhere (1),(2). Both the crystals of $CaCO_3$, which were large and well shaped, and the much smaller TTF-TCNQ crystals showed unusual morphological behaviour.

The follow up on these experiments (planned in 1979) was started already in 1981, when ESA initiated a "Concept Definition Study" on a large, 150 kg, Solution Growth Facility (SGF) to be included in the payload of EURECA-1, the European Retrievable Carrier. This carrier was a continuation of the European Spacelab and at that time planned for launch in 1987.

The long delay of the LDEF retrieval and of subsequent missions brought about reflections both on the concept of crystal growth in space and on the choice of crystallization materials, that had been made for the LDEF. As explained in (1) under "Historical Background" events on earth, during the flight of LDEF, caused a dramatic decline in the demand for TTF-TCNQ crystals. Already before the LDEF retrieval, research on TTF-TCNQ had been stopped, and a planned growth experiment with TTF-TCNQ on the SGF/EURECA had been cancelled.

The target of the SGF investigation is now more fundamental in nature. None of the crystals to be grown here are, like TTF-TCNQ, in particular demand by science or industry, and the crystals only serve the purpose of model crystals. The real purpose of the investigation is to study the growth behaviour. One of the experiments, the Soret Coefficient Measurement experiment is not growing crystals at all, but has it as its sole purpose to obtain accurate information on thermal diffusion, a process of importance in crystal growth from solution.

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The 4 LDEF growth reactors had all approximately the same size and were packed together in a thermostated container. The implication of this was that all the experiments had to be run at the same temperature. This temperature was determined by simple design (no cooling water) and the minimum consumption of electrical energy to the heating elements. 35^{O} C was used.

The 4 SGF/EURECA experiments (fig. 1) may be of different sizes, and they are each individually thermostated. The volumes of the reactors are increased up to 5 times the LDEF volumes, but still the simple design is maintained (no cooling water). Experiment temperatures may range from 35^{O} C to 70^{O} C, and the temperature-time profile as well as the temperature-position profile is monitored for each experiment throughout the mission.

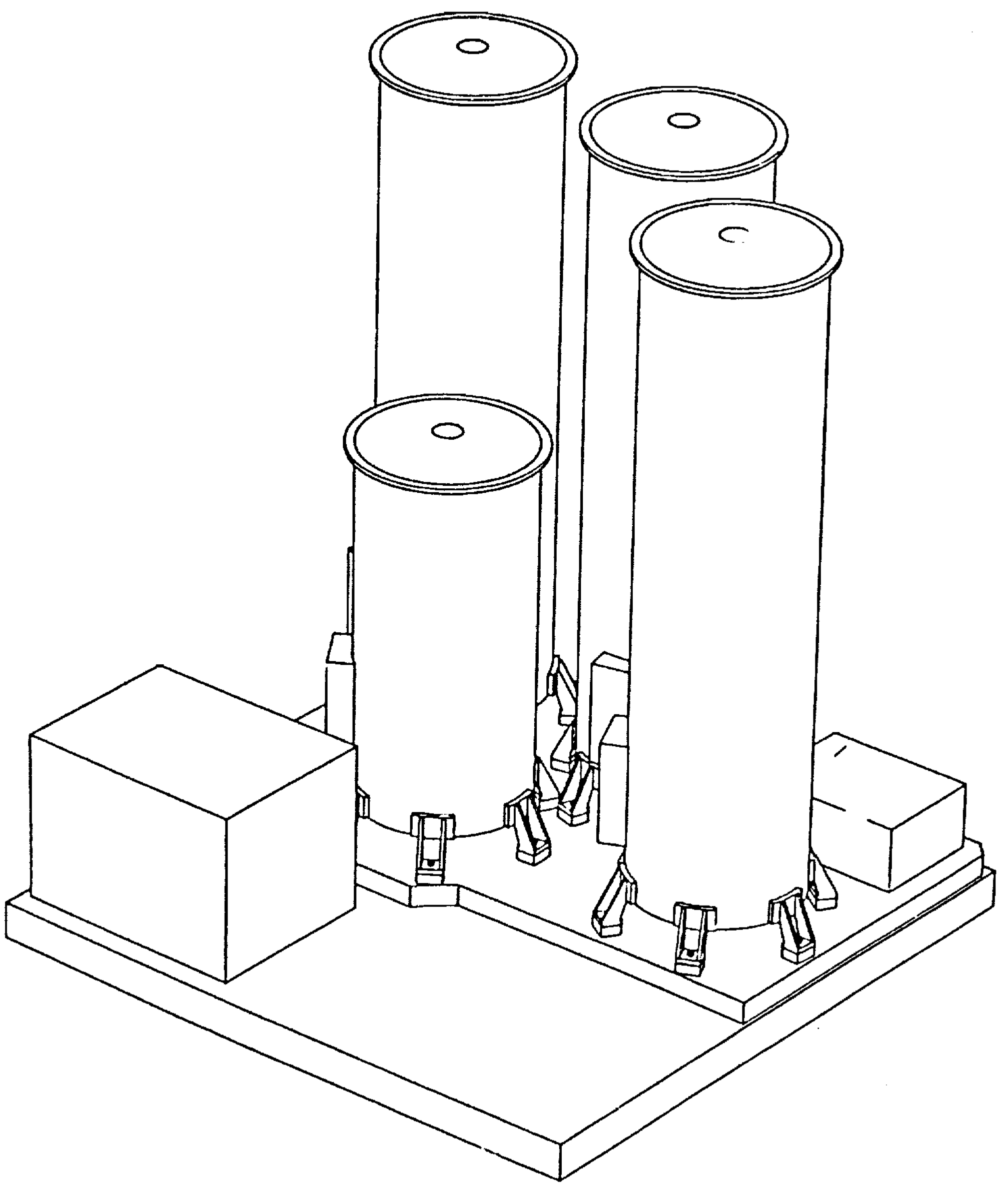

Fig. 1.

Fig. 2 is a construction drawing of a growth reactor. Basically they follow the same design principles as the LDEF reactors (2). A major improvement has been introduced in the pressure equallizing system with the bellows, designed and constructed by A.G. Contraves in Switzerland.

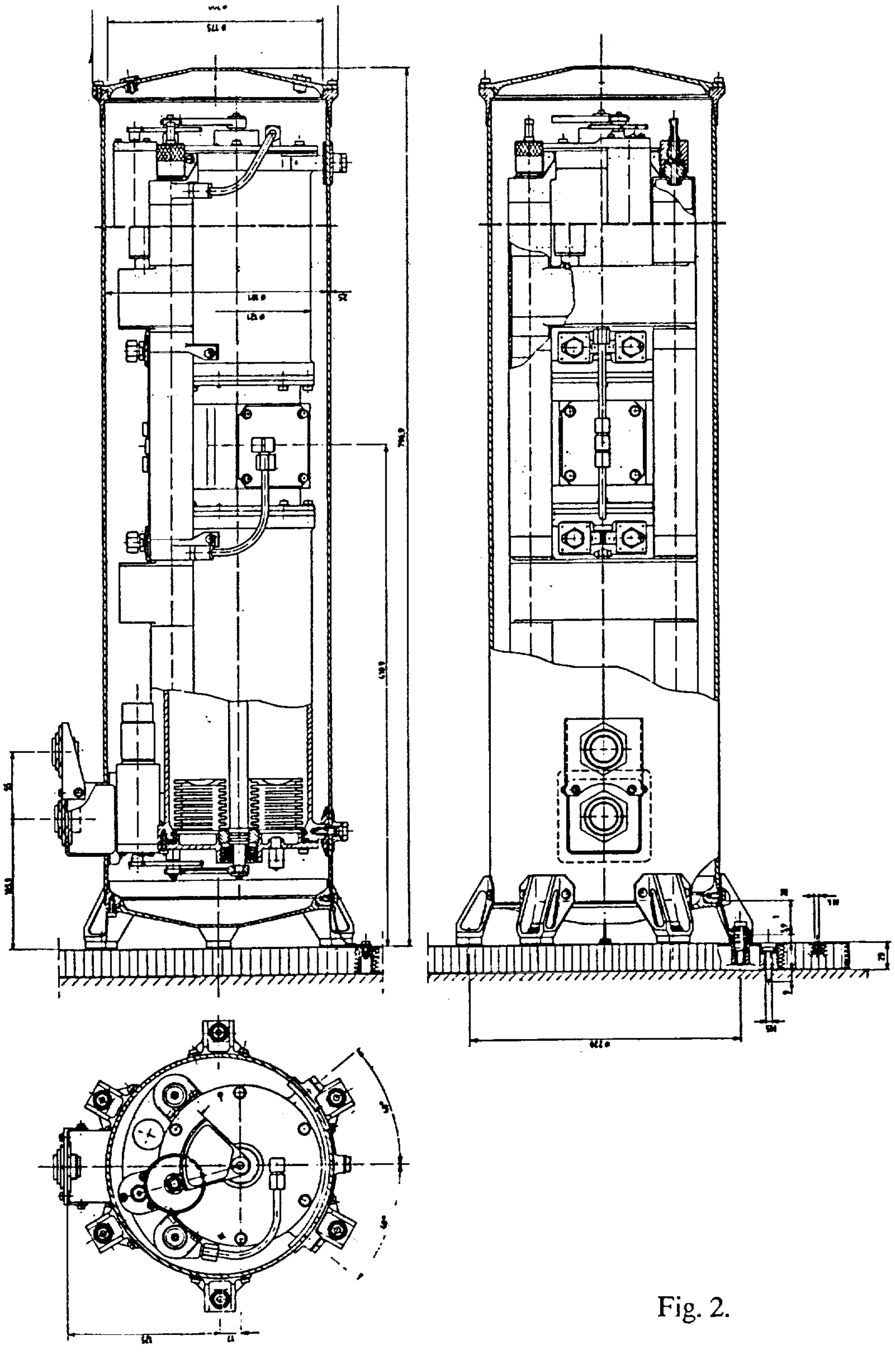

Fig. 2.

The SGF reactors and the total integrated EURECA under testing are shown in figures 3 and 4.

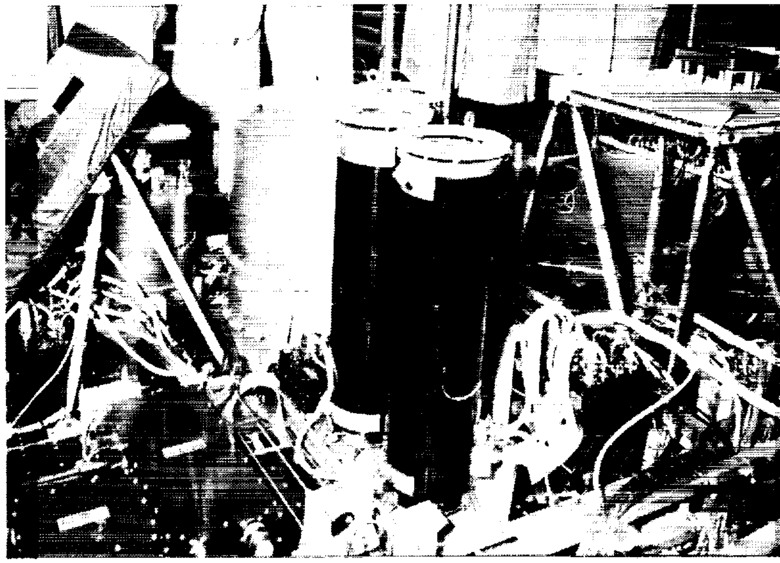

Fig. 3. The SGF reactors mounted on the EURECA.

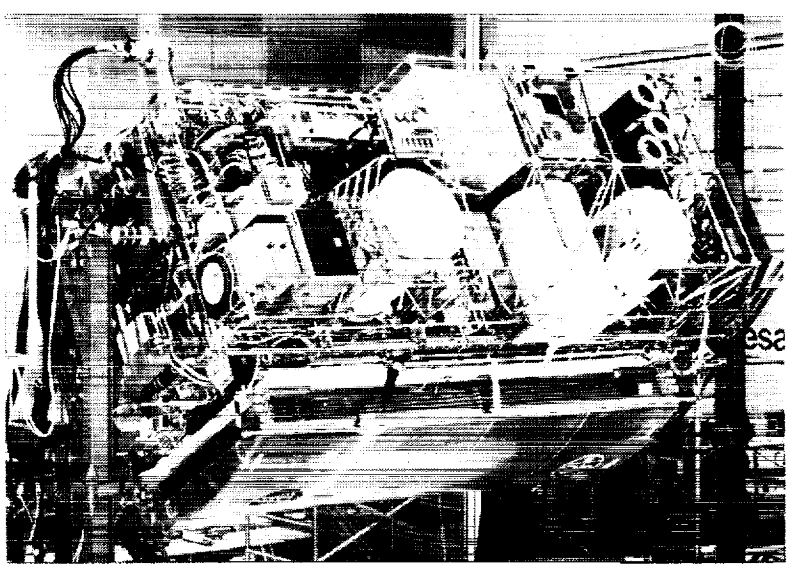

Fig. 4. Total integrated EURECA under testing.

The 4 SGF experiments have been described in detail elsewhere (3). Here only a schematic summary of the scientific background for the experiments will be given.

1. Growth of Calcium-Carbonate Crystals.

P.I. M. David Lind
Rockwell Int. Science Center
Thousand Oaks, California.

Co. P.I. Kjeld Flemming Nielsen
Technical University of Denmark
Lyngby, Denmark.

This experiment is a reproduction of the successful growth experiment on the LDEF. The unexpected and unintended long duration of LDEF prevented us from observing free $CaCO_3$-crystals obtained via homogeneous nucleation in the bulk of the solution, followed by growth of freely suspended crystals. Apart from this we are interested in a further study of the habit formation under microgravity conditions, which is unusual.

2. Formation and Transformation of Tri-Calcium-Phosphate.

P.I. H.E. Lundager Madsen
Royal Veterinary and Agricultural University
Frederiksberg, Denmark.

Nucleation and crystallization of Calcium-Phosphate takes place with an even stronger participation of water molecules than in the case of Calcium-Carbonate. Tri-Calcium-Phosphate can be described as an intermediate gel structure, from which further crystallization takes place. The study of the influence of gravitation on this gel structure and its further crystallization into 3 or 4 new crystal structures is the background for this experiment. Morphology, nucleation, and aggregation of the various structures in microgravity will be studied.

In living organisms the calcified tissues, known to be influenced by the lack of gravitation, consist mainly of Phosphates and Carbonates. Thereby the above two experiments may be viewed in a broader perspective than only that of basic science in the growth of crystals.

3. Growth of Zeolite Crystals.

P.I. Michael Stoecker
Center for Industrial Research
Blindern, Oslo, Norway.

Zeolites are crystalline Alumino-Silicates with well defined pore structures containing open channels, pockets, and pores in the same range of kinetic diameters as small molecules (3-12 Å). They are used for catalysis and ion exchange. Also here water molecules are active in the crystallization that takes place via an aqueous gel. With these crystals accurate knowledge of pore diameters is important for their application, and the material in question is Offretite, of which large, well shaped crystals are wanted. The objective is to produce channels with pore size diameters 6.4 Å and 4.3 Å (12 and 8 membered ring channels). Thus the study of morphology here is somewhat specialized.

4. Soret Coefficient Measurements (Diffusion).

P.I. J. C. Legros
Chimie Physique E.P. (CP.165)
Universite Libre de Bruxelles, Belgium.

The Soret Coefficient is the ratio of the thermal to the isothermal diffusion coefficient and is defined positive, if the denser component migrates towards the cold side. This investigation aims at the determination of the Soret coefficient in various binary organic mixtures and aqueous electrolyte solutions. The interior of this experiment is therefore totally different from the other 3 experiments. It contains an arrangement with 20 tubes with a volume of 10 ml each, and a temperature gradient of about 10^{o}C is placed over each tube.

The relation to crystal growth in this experiment is that the value of the Soret coefficient is an important parameter in the growth from solution. If the growth process involves exchange of heat, which it normally does, extra liquid flows will be generated under gravitation.

During the past year, a number of EWG's (Expert Working Groups) havebeen set up by ESA, the European Space Agency. The group on solution growth has come up with a number of ideas for further investigations under microgravity, which may be summarized as follows:

Morphological Studies	Mass Transfer Studies	Nucleation Studies	Aggregation Studies
Available theories may only be experimentally verified in micro-g environments.	Diffusion profiles may be steeper without convection.	Induction time is increased in micro-g. Early stages may be found.	Cluster formation & behaviour greatly influenced by gravitation.

The morphological, nucleation, and aggreation studies have been dealt with. Studies of mass transfer have until now only been in "natural" concentration gradients, i.e. gradients that are isothermal and where the gradient is maintained by removing solute from one side. The Soret coefficient experiment is now studying thermal gradients as well.

By removing solvent from the other side of a concentration gradient, f.inst. by evaporation, gradients may be steeper. Under microgravity conditions, gradients may become almost arbitrarily steep without convection. Thereby diffusive mass transfer may be enhanced.

One of the EWG members, Rafael Rodriguez-Clemente, has proposed a microgravity experiment to this effect. It is based on earth experiments on crystal growth from boiling solutions (4).

References:

1) K.F.Nielsen & M.D.Lind: "Results of the TTF-TCNQ and the $CaCO_3$ Crystallization on the LDEF", NASA Conf. Publ. 3134, part 3, p. 1675, Kissimmee FL, 1991.

2) M.D.Lind & K.F.Nielsen: "Crystal Growth by Diffusion in Earth-Orbit", SPIE Vol. 1557, p. 259, San Diego CA, 1991.

3) EURECA-1 Experiment Book, ESA Ref: MU/132/LI, Noordwijk, 1989.

4) R.Rodriguez-Clemente et al: "Crystal Growth from Boiling Solutions", Prog. Crystal Growth and Charact., Great Britain, Vol. 17, pp 1-40, 1988.