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"FIXBOX" - A NEW TECHNIQUE FOR THE RELIABLE CONDITIONING OF PLUTONIUM WASTE SOLUTIONS

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Annex IV

"FIXBOX" - A NEW TECHNIQUE FOR THE RELIABLE CONDITIONING OF PLUTONIUM WASTE SOLUTIONS

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Abstract

"FIXBOX" - A new technique and facility for the conditioning of plutonium waste solutions has been developed and brought into operation in the Hot-laboratory at PSI, for the solidification of the waste from the research programmes. The facility is situated in glove-boxes for handling alpha activity and gamma-shielded for conditioning of fission product-containing waste.

This report gives a brief description of the FIXBOX facility, the procedure and the first results of the cementation of plutonium waste solutions.

As a result of this solidification, the actinide waste is homogeneous and strongly bound in the cement. The presence of gluconic acid and other complexing agents in the waste solution will not disturb this process.

1 Introduction

Cementation is proposed as the best method for the conditioning of radioactive waste solutions, the most important of which are low and intermediate level concentrates from the evaporator, resulting from the reprocessing of spent fuel [1,2]. The content of this waste solution, the kind and concentration of dissolved substances, is well known and varies only slightly.

Procedures for the conditioning of waste solutions from the fuel production recovery process are widely known and used. Usually, the actinides are precipitated or coprecipitated in the form of their hydroxides and conditioned with a cement mixture to produce a certified waste disposal package.

As opposed to these standard waste solutions, many other types of radioactive waste solutions, with very different components and concentrations, are produced, e.g., in programmes of actinide research or in experiments for introduction of new technologies.

The investigation of the decomposition process of organic substances in mixed waste indicated the formation of complexing agents, which are able to form stable, soluble complexes of the actinides and lanthanides [3]. For this reason, experts classify this kind of ill-defined mixed waste, with a broad range of radionuclides and chemicals, as the group with the highest safety risk for future radioactive waste disposal.

The new FIXBOX waste conditioning technique was developed as a contribution to solving this problem.

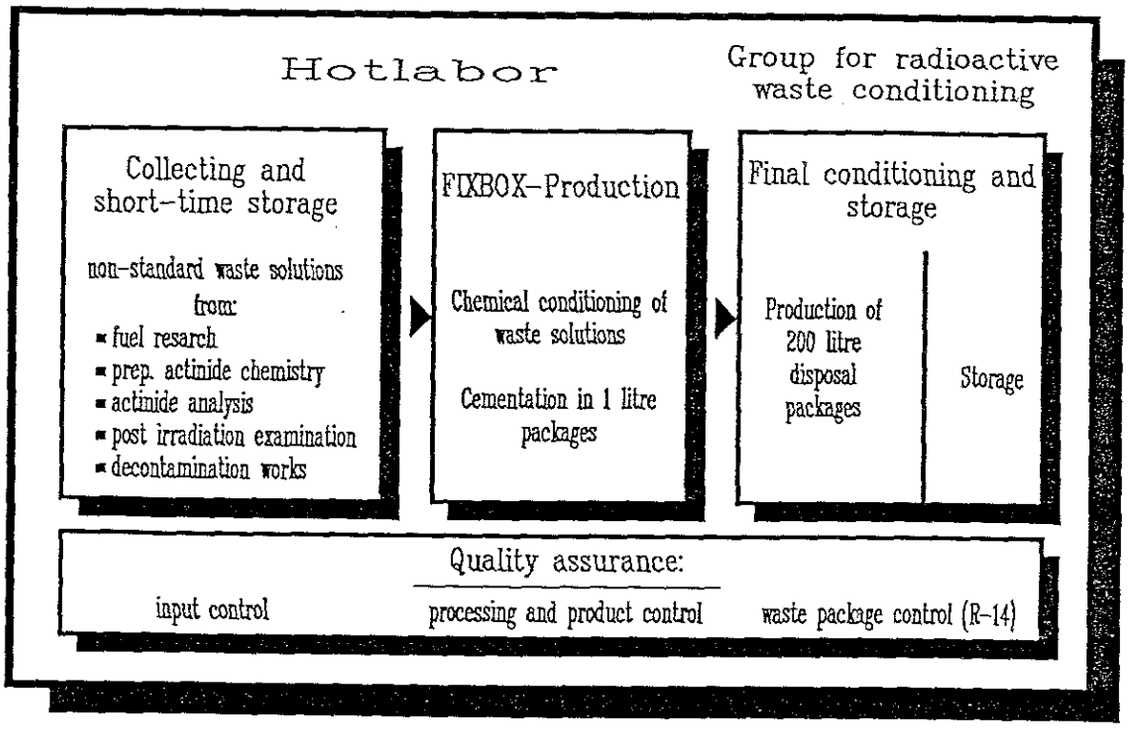

Fig. 1: PSI waste management of actinide solutions from the research programs

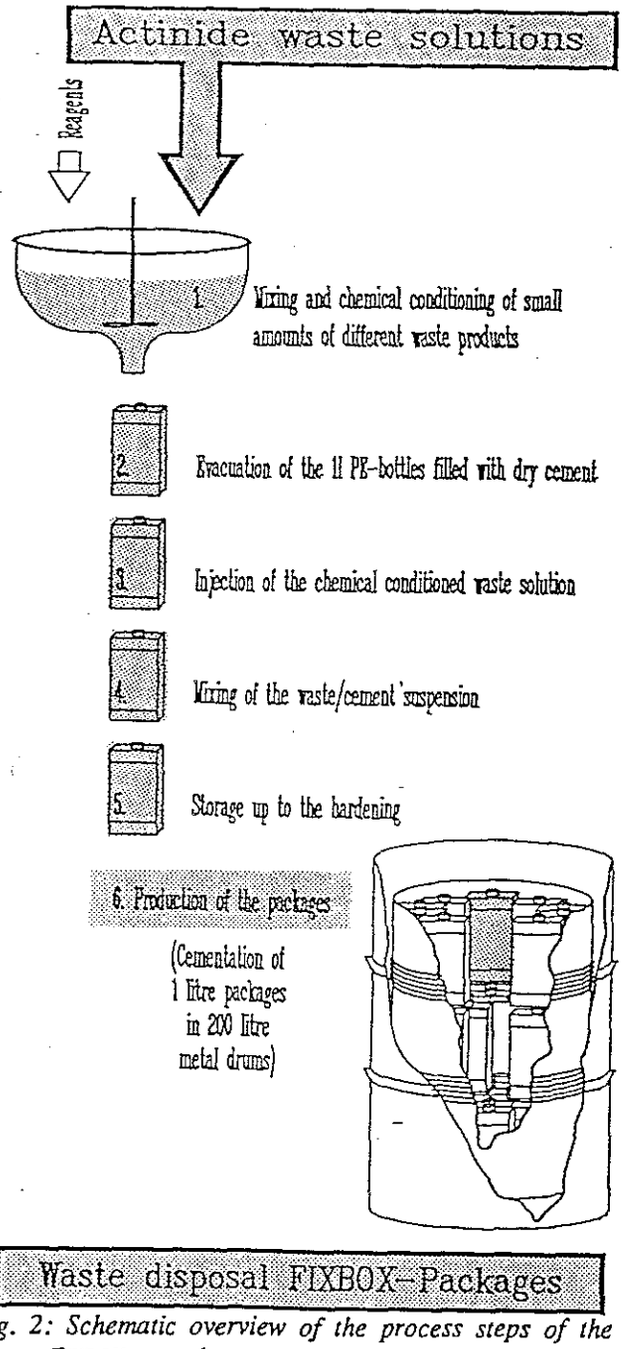

Fig. 2: Schematic overview of the process steps of the FIXBOX procedure

2 Concept of the FIXBOX waste conditioning

Due to the fact that the actinides form very stable and insoluble hydroxides which, even in extremely low concentrations, are strongly adsorbed on the surfaces of solid phases, we have developed a new method for the reliable conditioning of actinides by cementation [4].

The actinide hydroxides are precipitated immediately on mixing the waste solution with the cement grout and are distributed homogeneously onto the surfaces of the solid calcium oxides or cement particles. The cement mixture contains a surplus of calcium oxide to neutralise acid in the waste during the cement/waste solution mixing, and for the adsorption of the actinide species.

Retardation of the cement hydration and hardening is achieved by use of a plasticiser to allow complete actinide fixation on cement micro-particles. Once hardened, the cement bricks provide a chemical and physical environment in which the actinides are strongly fixed, greatly reducing the possibility of leaching. A sulphate-resistant cement with a low leacheability factor is used with a plasticiser which is a precise mixture of gluconic and tartaric acid, both thoroughly investigated, simple organic compounds [3].

Use of one litre bottles for the primary step of solidification gives the flexibility to condition small amounts unstable waste forms immediately and, in the production of the final waste disposal FIXBOX-Packages, to choose the optimal radionuclide content of the 200 litre drums.

3 FIXBOX - Procedure

The PSI waste management of actinide solutions originating from research programmes (Fig. 1), provides for the collection, short-term storage and conditioning of one litre packages in the Hotlab, and the final conditioning of the certified and quality-controlled waste disposal FIXBOX-Packages in the Department of Radioactive Waste Management.

The FIXBOX-procedure (Figs 2 and 3) includes the mixing of different types of actinide waste solutions (Fig. 2, step 1), their chemical conditioning with standard solutions (Fig. 3, *) and slurry (Fig. 3, **) to produce a 20 litre charge of actinides dissolved in an acidified aqueous solution. The solidification produces, in the first step, bottles of 1 litre (Fig. 2, steps 2-5) and, finally, a 200 litre Waste Disposal FIXBOX-Packages in a metal drum (Fig. 2, step 6).

4 FIXBOX - Facility

The FIXBOX production of the 1 litre packages is performed in a two box facility with special low pressure cells for handling of actinides and gamma-shielding for the work with fission product-containing waste. The schematic configuration of the facility and the boxes for chemical conditioning and cementation are shown in Fig. 3 - Fig. 6. The procedure is remote-controlled and mainly automated.

5 Preliminary results of cement solidification

For the first cementation experiments both pure concentrated nitric acid and acidified actual actinide waste solutions were used. The waste solutions contained fission products, iron, chromium, nitric, phosphoric, gluconic and tartaric acids, as well as other inorganic and organic substances, up to a total concentration of 0.4 g/ml.

Experiments with pure cement or cement mixed with Clinoptilolith [4] were controlled by measurements of lanthanide and actinide distribution between the supernatant water and the cement phase, carried out by alpha and gamma ray activity analysis. The radioisotopes

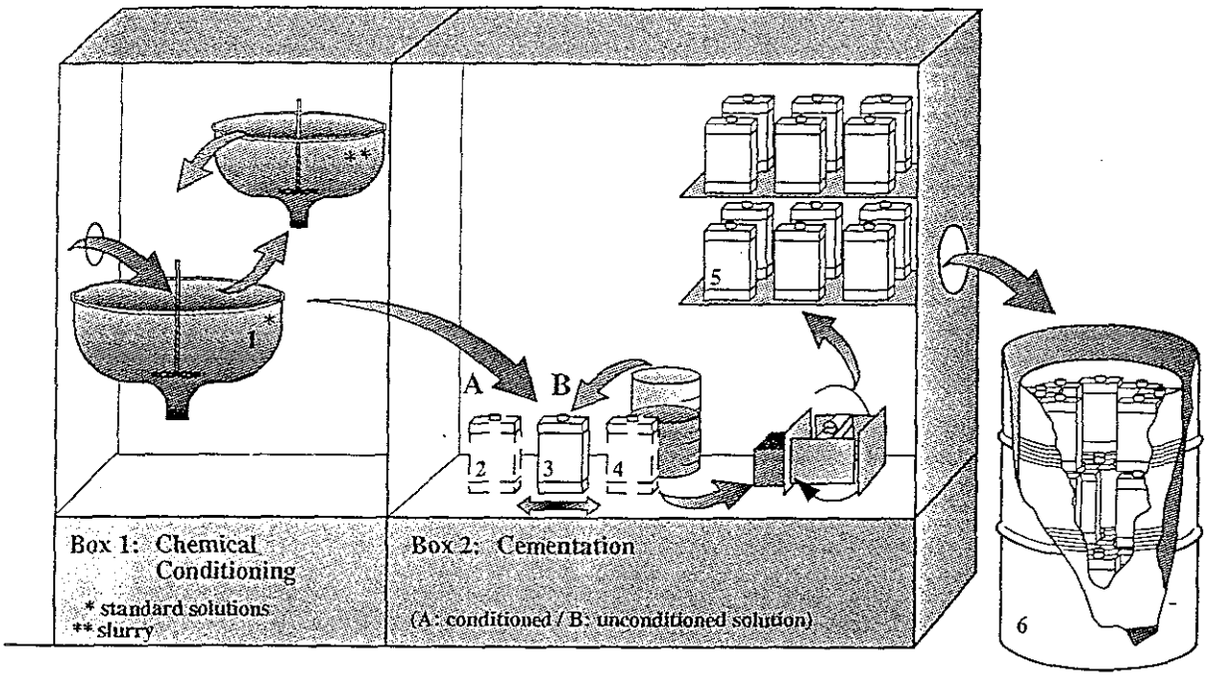

Fig. 3: FIXBOX facility: schematic configuration (see also Fig. 2)

The lanthanides are fission products and were used simultaneously as tracers for the fast determination of the trivalent actinide concentrations.

Most of the radionuclides were completely fixed within a few minutes of mixing the solution with the dry cement powder. The plutonium, americium and curium residues in the excess solution before solidification were less than 0.01% of the original concentration. This supernatant solution was subsequently incorporated in the cement during solidification. Measurements of the mechanical, thermal and chemical stability in the first months after the conditioning indicate the high quality of the cementation product. Compressive strength after one to six months curing time, and up to 5 months leaching time, is more than 40 N/mm^2.

The next test experiments will be performed with more concentrated solutions of actinides to measure very high distribution coefficients. Further systematic measurements will be made within the framework of the quality assurance for waste packages, R-14 of the Swiss Federal Nuclear Safety Inspectorate (HSK) [5], and long-term investigations for the FIXBOX-products are also in progress.

6 Conclusions

The results of the first quality tests of the solidified products and the predictions for the long-term chemical behaviour of their components indicate that the FIXBOX-procedure and facility are suitable for the conditioning of many different, and especially non-standard, types of waste solutions, including those containing relatively high salt contents and strong actinide complexing agents.

Acknowledgements

The authors are grateful to Dr.J.Nöggerath (HSK) for helpful discussions and support in setting up the FIXBOX facility and testing procedures. The control system of the facility was designed and built by Mr. M. Martin and his group of apprentices. This FIXBOX facility and the idea of the first cementation step with evacuated bottles, in particular, was introduced by Dr.M.Furrer, P.Wagner and P.Leupi [6]. We also wish to acknowledge H.P. Linder for the performance of γ-measurements and W. Görlich and his group for the fruitful collaboration.

Fig. 4: FIXBOX facility: Box 1 - chemical conditioning

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FIXBOX facility: Box 2 - cementation of chemically conditioned waste solutions

FIXBOX facility: Box 2 - handling of 11 - bottles for cementation of fission product containing Pu solutions

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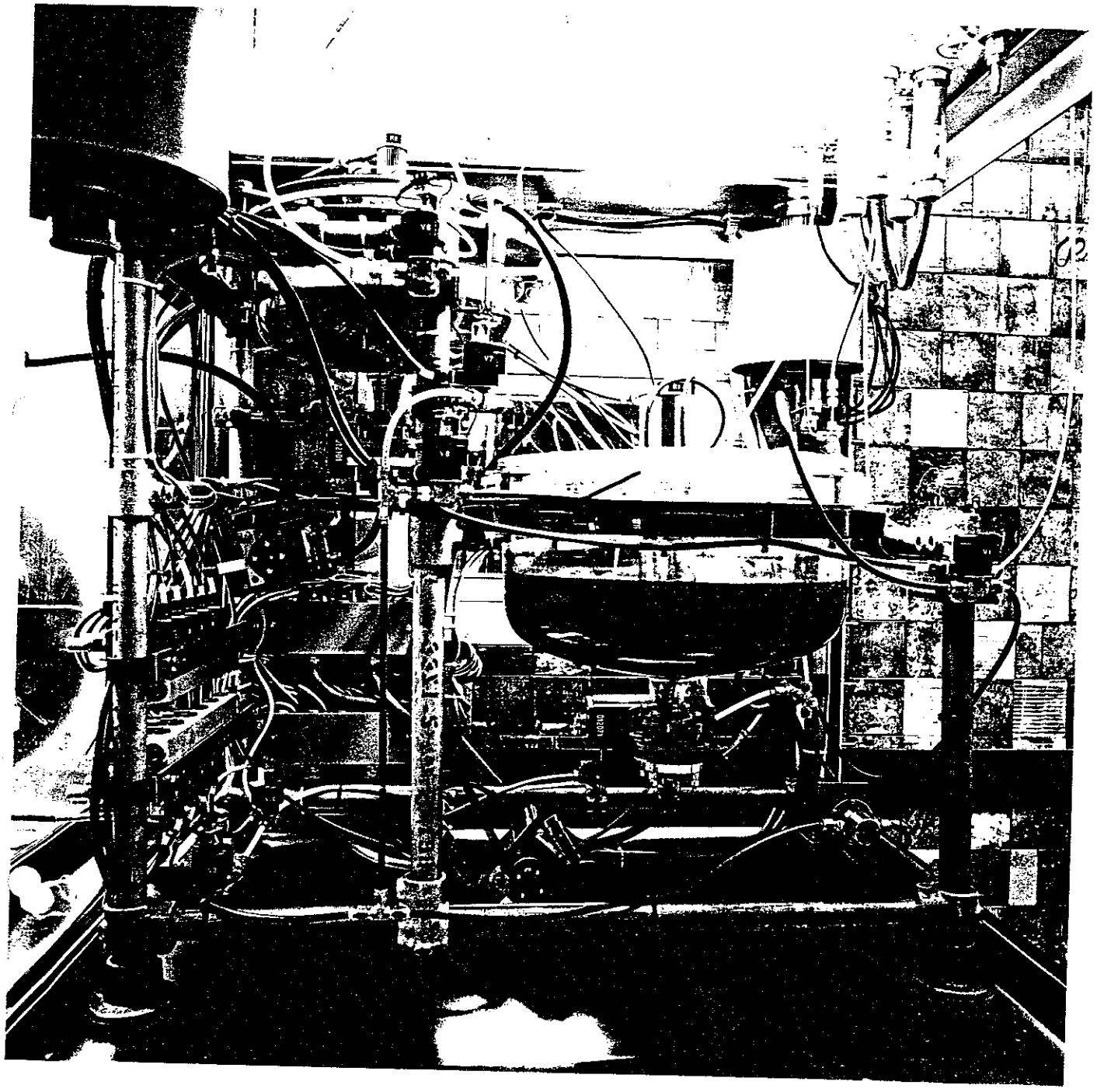

Figure 4

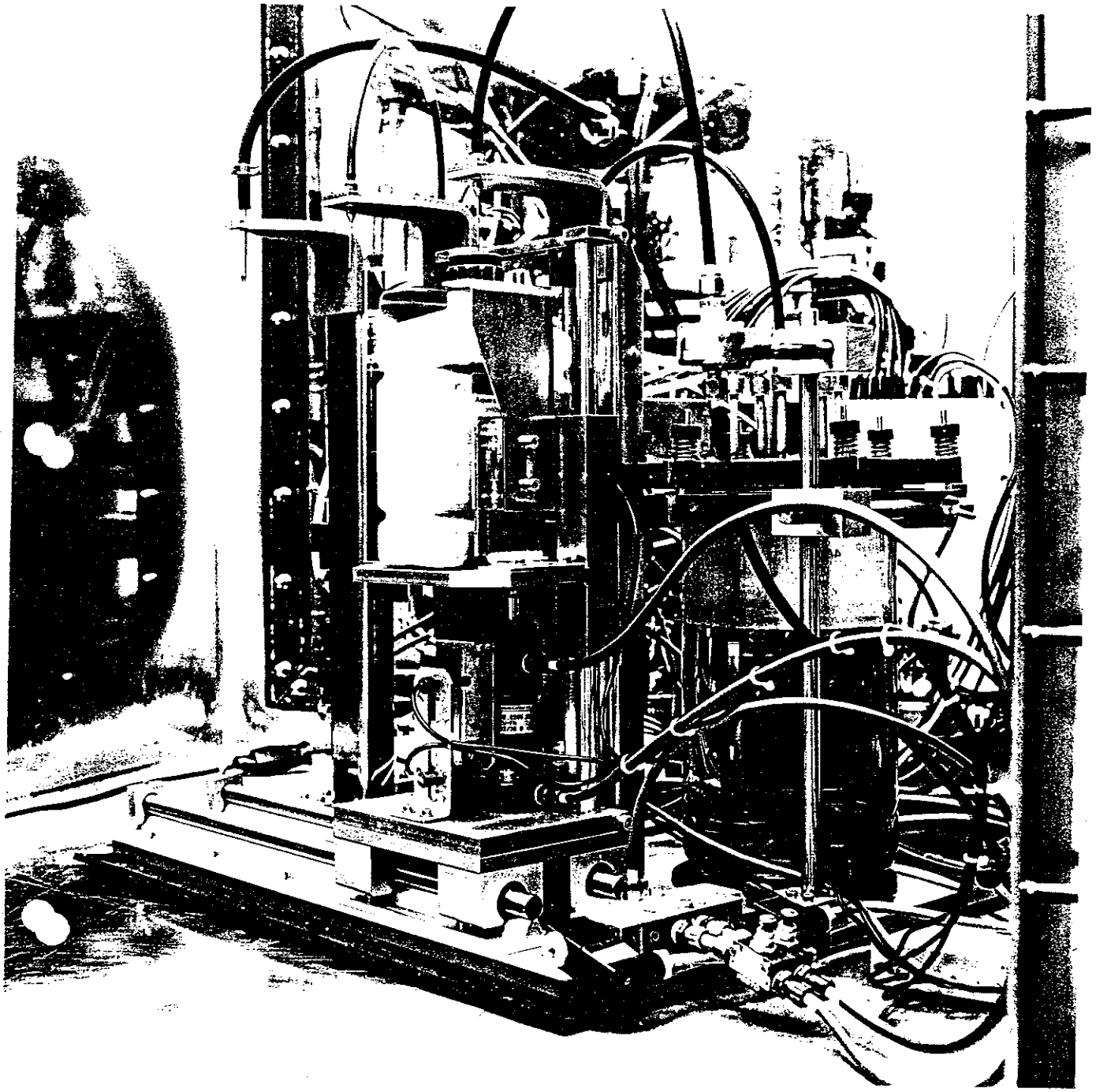

Figure 5

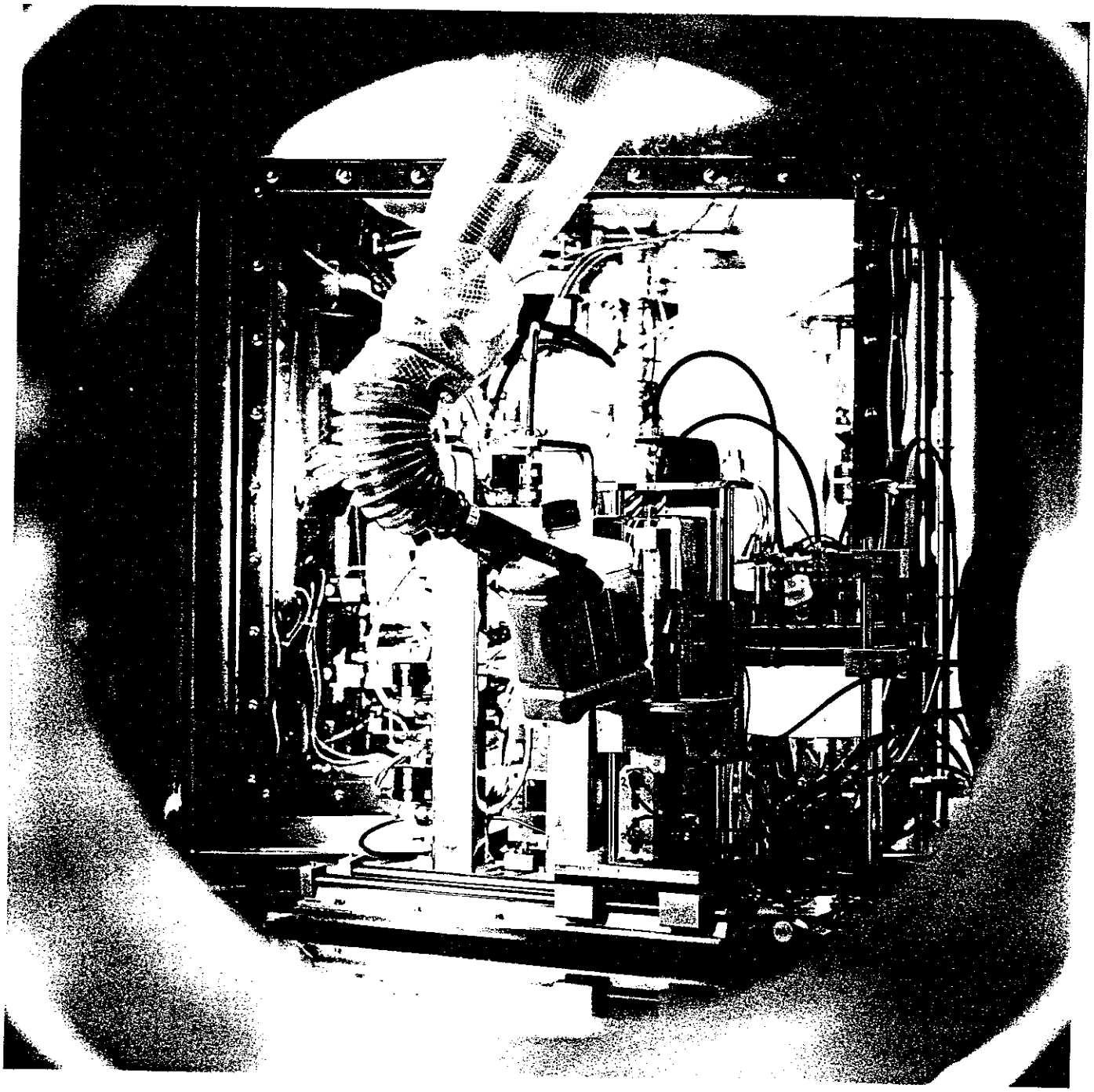

Figure 6