

NUCLEAR FUELS

Edited by

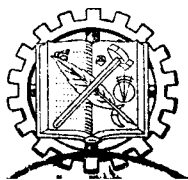
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FOREWORD

No matter how the Geneva Conference will be judged by historians of the future, there is one fact which will hardly be forgotten for some time to come: in ten days of August, 1955, an unprecedented volume of technical information on atomic energy was put into the public record. Not all the items of information were of equal value, to be sure, but relatively little was of a trivial nature.

Realizing that a verbatim record of this avalanche of data would be relatively indigestible to a large body of workers in the field, the Publishers requested me to undertake the task of organizing and editing a series of books which would present material of most urgent usefulness. Preprints of many of the conference papers were made available through the courtesy of the United Nations staff.

From a study of these papers it was concluded that six volumes of nominal length would be required. Each would cover a specific subject and include the key papers, suitably edited to eliminate duplication and secondary material, and arranged in a logical manner.

Because of the great interest in finding the raw materials for the growing nuclear appetite, Robert D. Nininger, Deputy Director for Exploration, Division of Raw Materials, United States Atomic Energy Commission, was asked to edit a volume on the geology of and the exploration for uranium and thorium.

All atomic energy projects to date have had research reactors at the base of their technical efforts. Dr. Clifford Beck, Professor of Physics, North Carolina State College, whose foresight and determination put the first research reactor on a university campus, consented to edit a volume on research reactors.

At the center of the stage, particularly for the energy-hungry nations of the world, is the power reactor, a device of many and varied forms. James K. Pickard, now a consultant engineer on atomic energy developments, for many years previously one of the stalwarts of the AEC Reactor Development Division, accepted the task of preparing a nuclear power reactors volume.

Because one of the most difficult and urgent problems in the power reactor program is assessing the hazard and providing safety in reactor operation, it appeared urgent to include a volume devoted exclusively to reactor safety. One of the pioneers in the field, Dr. C. R. McCullough of the Monsanto Chemical Company, has undertaken to edit a volume on the safety aspects of nuclear reactors.

The heart of the power reactor is the nuclear fuel, the energy source and the focal point for intense metallurgical and solid state research efforts. Dr. David H. Gurinsky and Dr. G. J. Dienes of Brookhaven National Laboratory, who have made substantial contributions to these efforts, have edited a volume on nuclear fuels.

In addition to the direct heat energy from fission, the ten per cent or so of reactor energy available as radiation is already exerting profound effect on civilization. One who has probed deeply into what might be termed the interaction of nuclear energy and agriculture, Dr. W. Ralph Singleton, Director of the Blandy Experimental Farm, University of Virginia, has edited a volume on nuclear radiations in food and agriculture.

These six volumes are not intended to cover all the technical areas of the Geneva Conference. Subjects such as radiobiology, nuclear physics, reactor physics, chemical processing—to name but a few—were not included (except where they touch on the subjects chosen) for a variety of reasons. A first consideration was that, if all subjects were properly represented, the series would presume an ungainly large number of volumes. Reducing the number of volumes could only be done by limiting coverage either to a few papers on each subject or to a superficial digest of a large number of papers. It seemed a better choice to cover fewer topics more completely. A secondary consideration was an assessment of the state of the literature—where a subject was rather well represented in the public literature, it could be argued that the need for a corresponding volume was less urgent. Certainly, for example, the literature of nuclear physics does not urgently require amplification. Moreover, although the nuclear physics papers published at Geneva were substantial, they did not disclose any striking new information. (In addition, many of the new nuclear data have been adequately disseminated by the United States Government handbooks.)

In any effort of this kind, arbitrary decisions are made in many places. Material is deleted, rearranged, abbreviated, and new material is added. It is hoped that these decisions have been wise and that the six volumes will be a fundamental reference work for the increasing number of scientists and engineers devoted to the peaceful uses of atomic energy.

J. G. BECKERLEY

PREFACE

A large body of previously classified information was disclosed for the first time at the International Conference on the Peaceful Uses of Atomic Energy held in Geneva in August of 1955. To meet the needs of this conference, many review papers were presented containing both recently released material and the related information available in the open literature. The editors' purpose in preparing this volume was to present a clear picture of the status of the field of nuclear fuels. To accomplish this end, selected material was assembled from the many excellent papers presented at the International Conference for Peaceful Uses of Atomic Energy.

It was imperative, if continuity of development was to be achieved, that some details of importance to the specialist be deleted. Consequently, this volume should be considered as an introduction, although a rather detailed one, to the field. It is hoped that this volume will also prove useful as an introduction to the specialized papers by providing a proper perspective for further study.

The volume has been subdivided into three parts: I. The metallurgy of uranium and thorium; II. Radiation effects; and III. Solid and fluid fuels. The purpose of this subdivision is (1) to acquaint the reader with the fundamental information necessary for designing a fuel system; (2) to point out the problems specific to reactor systems, i.e., radiation effects; (3) to show how the engineering requirements and the fundamentals are fused to produce specific fuel systems.

As the first topic of this volume the editors have chosen the Ames process for the reduction of uranium salt to metal since this process probably represents the most important development toward industrial scale production of fissionable material. The book does not include material on the prior activities of prospecting, mining and ore concentration. Thorium has been included as a fuel since its relation to the fissionable U-233 is the same as U-238 with respect to Pu-239.

An attempt has been made to provide complete references and to give credit to the sources from which the material was obtained. In order to make this volume available as early as possible, it was necessary to use much of the material as prepared for the Geneva Conference by the various authors. The material has been rearranged to fit a logical pattern and to avoid duplication. Although this may, in some instances, make it difficult to identify at a glance the

author of each individual paragraph, the editors believe that a valuable integration of important information has been achieved in the process.

Grateful thanks are extended to Florence Gurinsky and Margaret Dienes for their patient and invaluable assistance during the preparation of this book.

D. H. GURINSKY

G. J. DIENES

FUEL ELEMENT

ELEMENTS COMBUSTIBLES

ELEMENTOS COMBUSTIBLES

ТОПЛИВНЫЕ ЭЛЕМЕНТЫ

COMPOSITE FUEL PLATE

BRAZED FUEL-
PLATE ASSEMBLY

FINISHED FUEL
ELEMENT

JACKETED FUEL-CORE COMPACT

FUEL-PLATE CROSS SECTION

FUEL-HEATING CORE

URANIUM
DIOXIDE
POWDER

ALUMINUM
POWDER

POWDER CORE COMPACT

Completed Geneva Reactor Fuel Element and Its Components

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Part I

THE METALLURGY OF URANIUM AND THORIUM

Chapter I

THE PREPARATION OF URANIUM METAL *¹

A. THE REDUCTION OF URANIUM TETRAFLUORIDE WITH MAGNESIUM ²

A survey of the available literature in 1942 revealed that uranium metal could be prepared by any one of a number of methods. Laboratory tests on some of the more promising of these methods for production were subsequently reproduced. These tests showed, however, that the methods described in the literature either gave poor quality uranium metal or left much to be desired processwise for a large-scale low-cost operation. These results warranted an exploratory effort directed toward the development of new methods for the preparation of uranium metal. A process employing the reduction of uranium tetrafluoride with calcium was the first major step in the desired direction. This process was soon followed by the development of the method for producing uranium metal from uranium tetrafluoride with magnesium.

When the program on atomic energy in the U.S.A. was expanded early in 1942, there were two producers of uranium metal operating on a small scale in America. One producer used calcium hydride for the reduction of uranium oxide. The mixture of UO_2 and CaH_2 was placed in a steel cylinder inside a retort which was heated to 960°C . After the reaction occurred, the charge was allowed to cool under a vacuum to eliminate the hydrogen. The product of the primary reaction was a sintered clinker, which was then crushed to a powder. This powder was subsequently leached with dilute acetic acid, washed, dried, briquetted and sintered to form a fairly dense metal. This uranium was rather impure and contained considerable oxide.

The other producer employed an electrolytic process in which either KUF_6 or UF_4 was dissolved in a fused salt mixture of 80 per cent CaCl_2 and 20 per cent NaCl in a graphite crucible. This crucible served as the anode and the

* This chapter is based on the following Geneva Papers:

Section A: 817—H. A. Wilhelm, "The Preparation of Uranium Metal by the Reduction of Uranium Tetrafluoride with Magnesium."

Section B: 407—L. Grainger, "Production of Uranium Metal."

uranium was electrolyzed out in the form of a granular deposit on a central molybdenum cathode. A temperature of about 900°C was employed for the fused salt electrolyte in order to produce coarser, less pyrophoric metal. The uranium was subsequently leached, dried, briquetted and melted and cast in beryllia crucibles to give good quality metal.

Neither of these two methods met all of the desired requirements as processes for large-scale low-cost production of high purity uranium metal. Production was stepped up, however, by both of these methods beginning early in 1942 and plans for even larger expansion of the electrolytic plant were shaping up when the practicability of the uranium tetrafluoride-calcium process was demonstrated.

Efforts on the UF_4 -Ca process were expanded and a few tons of the metal were produced in the chemistry laboratories at Iowa State College during the latter half of 1942 while a separate building on the campus was being remodeled and furnished to serve as a small production plant. Meanwhile, efforts were also directed toward the experimental development of the process employing magnesium in place of calcium; and as a result early in 1943 all uranium metal production was converted from the calcium to the magnesium reduction of the tetrafluoride.

This first uranium metal production plant employing the UF_4 -Mg process served not only to supply metal for experiments and for charging atomic piles but also served as a pilot plant. The process proved to be one that could be readily expanded and it gave high purity uranium at a low cost. Over a thousand tons of uranium metal billets were shipped from the production plant on the college campus before industry took over uranium metal production entirely.

1. The reaction. There are certain advantages in using magnesium instead of calcium for the reduction of UF_4 . The purity of the uranium metal product depends largely on the purity of the materials that go into the charge and the materials that the metal contacts before solidification. Commercial-grade magnesium generally has fewer contaminants than commercial distilled calcium with respect to undesirable elements. Furthermore, the weight of calcium required to reduce a pound of uranium is about 1.6 times as great as the weight of magnesium required for the same weight of uranium produced. In addition, the cost per pound of the calcium may be five to ten times the cost per pound of magnesium. These factors, as well as relative availability, favored magnesium and made it desirable to explore the conditions for using magnesium instead of calcium in the reduction of UF_4 .

Some of the requirements for the magnesium reduction can be introduced by first considering the process of reducing UF_4 with calcium. If a mixture of UF_4 and excess Ca metal (at room temperature) is ignited at some point by means of a hot wire or a fuse, the reduction reaction $(2\text{Ca} + \text{UF}_4 \rightarrow 2\text{CaF}_2 +$

U) is propagated rapidly throughout the entire charge. The heat of the reaction is sufficient to heat both uranium metal and calcium fluoride to a temperature well above their melting points. The liquid products then separate readily because of the difference in their densities.

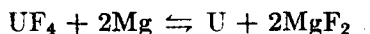
The heat generated by the reaction between UF_4 and magnesium metal is about 45 kcal less per gram-atom of uranium formed than by the corresponding reaction employing calcium metal. The melting point of the MgF_2 differs only slightly from that of CaF_2 . Experimentally, it has been found that the heat generated during the reduction with magnesium is not adequate, by itself, to give the necessary fusion of the products. If heat is added, however, to the charge before or during the reaction, the necessary temperature for fusion and separation of the products may be attained in the magnesium process.

This heat can be added by mixing in the charge an agent which reacts with the magnesium and thus generates excess heat on its own to serve as a thermal booster to the main reaction. If such a charge containing adequate booster at room temperature is ignited by a hot filament, the reactions will proceed spontaneously and give massive metal well separated from the slag. A mixture containing seven (7) mols of UF_4 to one (1) mol of KClO_3 and a small excess of magnesium will behave in this manner.

A more convenient method of accomplishing this same objective is to pre-heat the charge of UF_4 and Mg to ignition. The heat added to the charge before ignition and the heat of the reduction reaction together is sufficient to yield the products in the desired molten condition.

In the calcium reduction, the charge can be placed in an open vessel and very little calcium metal will be lost by vaporization during the reaction. However, in the magnesium reduction the boiling point of the magnesium metal is well below the melting point of the slag, making it essential that the reaction by this process be carried out in a closed vessel. The vapor pressure of excess magnesium metal at the peak temperature is well above one atmosphere and this pressure tends to drive the chemical reaction in the desired direction.

The reaction between uranium tetrafluoride (green salt) and magnesium metal,



when carried out under proper conditions in a closed vessel generates sufficient heat to reach a temperature at which the products of the reaction are molten. The dense liquid uranium metal collects to form a regulus, with the molten magnesium fluoride slag on top. After allowing the products to cool to near room temperature, the reaction vessel can be opened and the massive metal recovered.