Chemical Processing and Equipment

VOLUME SIX

Chemical Processing and Equipment

UNITED STATES OF AMERICA

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SELECTED REFERENCE MATERIAL

UNITED STATES ATOMIC ENERGY PROGRAM

Chemical Processing and Equipment

UNITED STATES OF AMERICA

Foreword

Interchange of scientific and technical knowledge will greatly facilitate the work of the scientists and engineers whose skills will be devoted to the future development of the peaceful uses of atomic energy.

The United States has made available to the world's scientific community a large body of such data. In honor of this historic Conference and to stimulate further exploration and development of the beneficial applications of nuclear energy, the United States Atomic Energy Commission has prepared this special collection of technical data for the use of the delegates and the nations represented.

The purpose of this collection is to provide information concerning the ways that we have found in which fissionable materials can be put to work in nuclear reactors for research purposes and for the production of power and radioisotopes.

It is our sincere hope that this material will be of practical value to the men and women of science and engineering in whose hands the great power of the atom is becoming a benign force for world peace.

Lewis L. Strauss

Chairman, U.S. Atomic Energy Commission

SELECTED REFERENCE MATERIAL ON ATOMIC ENERGY

VOLUME ONE Research Reactors
VOLUME TWO Reactor Handbook: Physics
VOLUME THREE Reactor Handbook: Engineering
VOLUME FOUR Reactor Handbook: Materials
VOLUME FIVE Neutron Cross Sections
VOLUME SIX Chemical Processing and Equipment
VOLUME SEVEN Eight-year Isotope Summary
VOLUME EIGHT Information Sources

Acknowledgment

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Chemical Processing of Reactor Fuel Elements at the Idaho Chemical Processing Plant

COMPILED BY NATIONAL REACTOR TEST STATION
PHILLIPS PETROLEUM COMPANY

Chemical Processing of Reactor Fuel Elements at the Idaho Chemical Processing Plant

It is not possible completely to burn up the fissionable material from the fuel elements in a nuclear reactor because the build-up of fission-product poisons, the depletion of the fissionable material, and the radiation damage to the elements eventually require their replacement. Economical operation of the reactor requires that the unconsumed fissionable material be recovered from the spent fuel elements. This section describes a successfully operating plant for the decontamination and recovery of fissionable material from fuel elements. Its purpose is to familiarize the reader with the problems connected with recovery operations and to give details of the process operations and plant facilities. As reprocessing operations must always be considered an integral part of any nuclear reactor research or power program, it is hoped that this information will be of value to those planning nuclear projects in the future.

The Idaho Chemical Processing Plant (ICPP) is designed to recover fissionable material from spent reactor fuel elements from various experimental and power-producing reactors in the United States. The plant is located at the National Reactor Testing Station near Idaho Falls, Idaho. The initial process development for the plant processes was carried out by the Oak Ridge National Laboratory (ORNL). The present operating contractor is the Phillips Petroleum Company of Bartlesville, Oklahoma.

This account is confined to describing the facilities, equipment, and process initially provided for uranium recovery from enriched uranium–aluminum alloy fuels such as those used in the materials-testing reactor (MTR) and the bulk-shielding reactor. Much of this equipment is, of course, adaptable to the processing of other types of fuels.

The high value of the uranium in the fuel elements demands that the process be capable of essentially complete uranium recovery. In addition, the process is required to effect nearly complete separation of the fission products from the uranium.

This plant is the first Atomic Energy Commission processing facility that has been designed for direct, rather than remote, maintenance. No provisions have been made for remotely controlled removal or repair of the process equipment. Thus, in the event of equipment failure or the necessity of modification, chemical solutions must be used to decontaminate the equipment before personnel can enter the process areas to carry out maintenance or construction work. As the radioactive solutions processed contain as much as 150 curies of radioactivity per liter, effective procedures for decontamination are an absolute necessity for the successful operation of this type of plant.

PROCESS

The process consists of three main steps: (1) dissolution of the uranium-aluminum elements in nitric acid, (2) adjustment of the dissolver solution to a composition suitable for solvent extraction, and (3) separation of the uranium from the aluminum, fission products, and transuranic elements with which it is associated in the fuel elements. The separation is accomplished by continuous liquid-liquid extraction employing methyl isobutyl ketone (hexone) as the solvent.

A schematic flow sheet of the process is shown as Fig. 1.

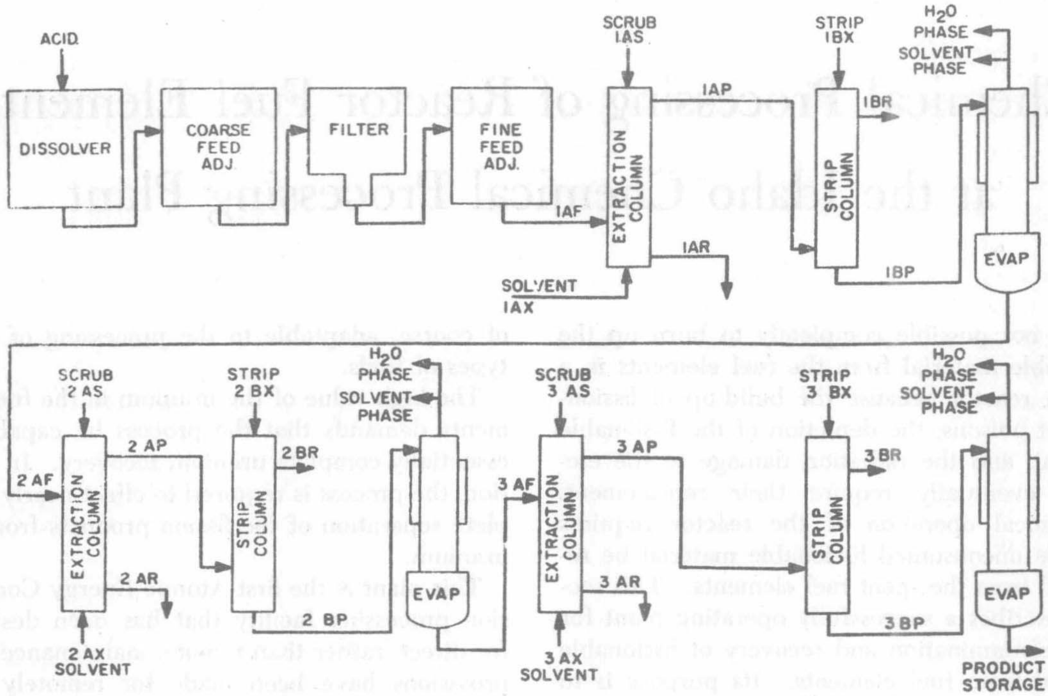

Fig. 1 Simplified feed preparation and extraction flow sheet.

Process Requirements. The process is required to recover greater than 95 per cent of the unconsumed uranium, produce a uranium product essentially free of plutonium, and reduce the fission-product activity to a level that will allow further processing of the uranium without shielding. In the case of MTR fuel, the fission-product concentration must be reduced by a factor of approximately 10^6. These requirements have been successfully met in all processing runs.

Fuel Cooling. At the time of discharge from a reactor, the fuel elements are intensely radioactive, owing primarily to approximately 100 short-lived fission products. It has been estimated that an MTR element will have about 2×10^8 curies of activity associated with it. This activity may be greatly reduced by storing (cooling) the irradiated elements and allowing these fission products to decay. Four days of cooling reduces the activity by a factor of about 10^3, and 135 days by a factor of 10^4. However, the activity that determines the minimum permissible cooling time is U^{237}, which is formed by neutron capture in U^{236}. It constitutes only a small fraction of the total activity at the time the irradiated fuel is discharged, but, since it is not possible to separate the isotopes of uranium during chemical processing, the fuel assemblies must be cooled until such time as the U^{237} activity in the recovered uranium is reduced to a low enough concentration to permit product handling and further processing without shielding.

Fuel Dissolution. The first chemical operation after cooling is the dissolution of the assemblies in nitric acid. Nitric acid dissolves finely divided aluminum quite readily, but, owing to the formation of passive surfaces of aluminum oxide, thick pieces of the metal are incompletely dissolved. The presence of mercury catalyzes the reaction of nitric acid with aluminum and allows complete dissolution, presumably owing to amalgamation and galvanic effects. Dissolving procedures for all fuel elements that contain aluminum include the use of mercuric nitrate as a catalyst. Dissolution of uranium metal in dilute nitric acid involves the primary reaction

$$U + 4HNO_3 \rightarrow UO_2(NO_3)_2 + 2NO + 2H_2O$$

The dissolution of aluminum in nitric acid involves a number of reactions which may be approximately summarized in the over-all reaction

$$Al + 3.7HNO_3 \xrightarrow{Hg} Al(NO_3)_3 + 0.15NO + 0.10N_2O + 0.175N_2 + 1.85H_2O$$

All the above reactions and others occur in the course of a normal dissolving, and it is difficult to predict accurately the composition of the gaseous reaction products. However, the acid consumption is always about 3.7 moles per mole of aluminum dissolved.

In addition to the gaseous reaction products shown above, volatile fission products are liberated during dissolution.

MTR elements are dissolved in a batch operation as shown on the chemical flow sheet (Fig. 2). The elements are charged to the dissolvers and are dissolved in nitric acid (3.7 moles per mole of metal) containing a weight of mercuric nitrate equivalent to 1 per cent of the element weight. The dissolution is carried out at boiling temperature. MTR elements dissolve rapidly; and to control the reaction rate, only one-half of the acid is added initially and the balance is metered in over a 2-hr period. The dissolver batch is digested for a total period of 7 hr at boiling, at which time the dissolution is 100 per cent complete.

Feed Preparation. The solution from the dissolvers is adjusted to a composition suitable for extraction-column feed in the feed-preparation equipment. These operations are carried out in batches following the chemical flow sheet for MTR elements shown in Fig. 2. The equipment for these operations is shown in Fig. 3.

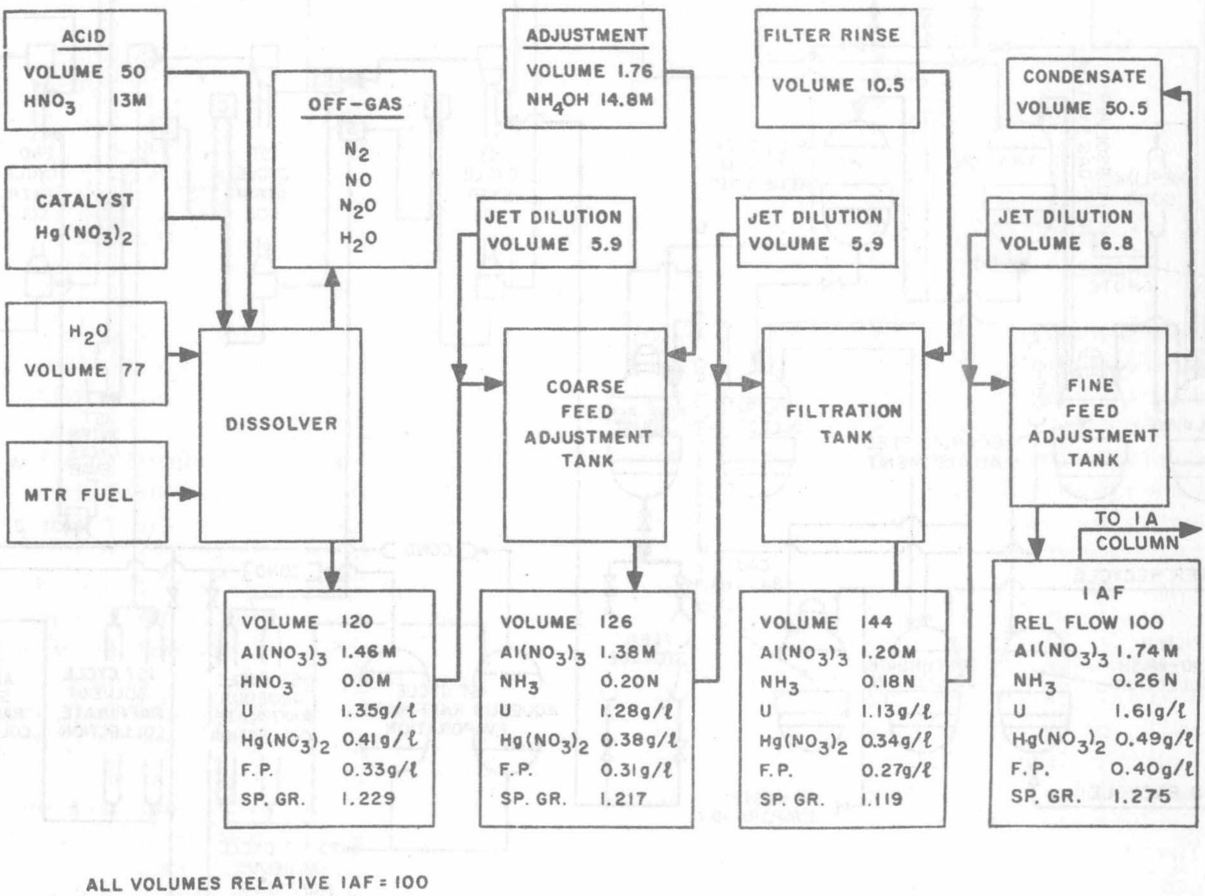

Fig. 2 MTR dissolution and feed preparation flow sheet.

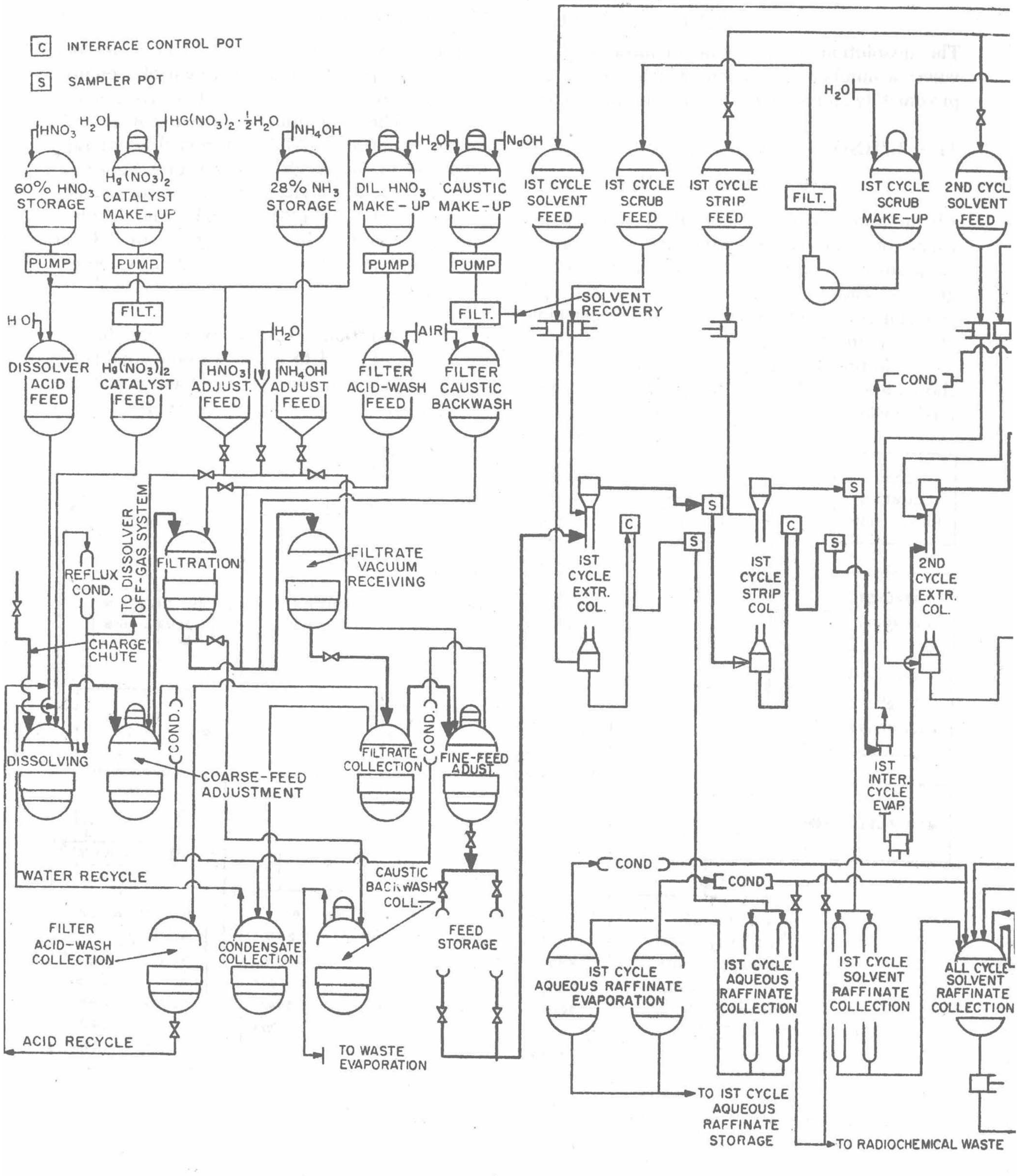

Fig. 3 Over-all process equipment flow

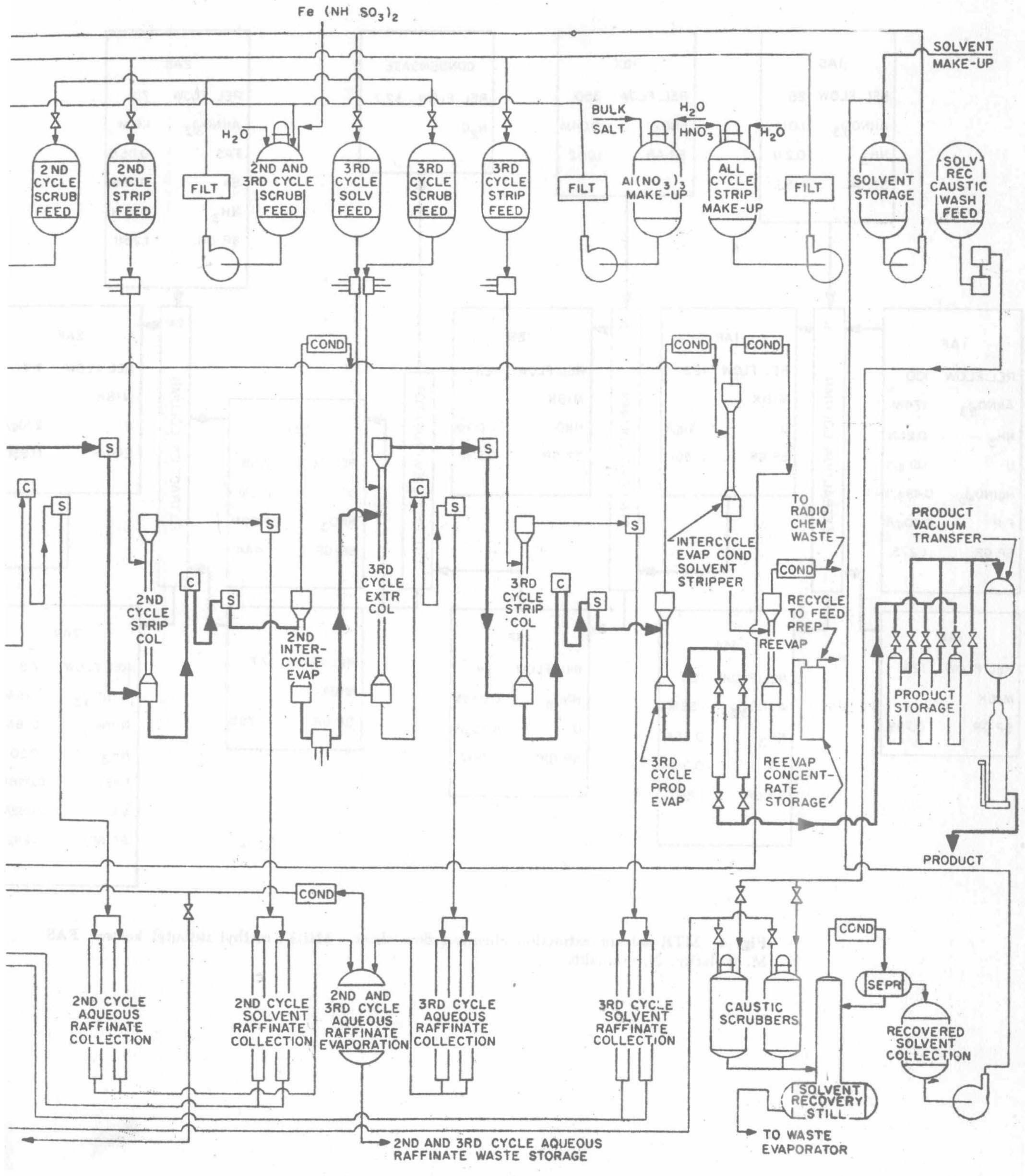

diagram for the Chemical Processing Plant.

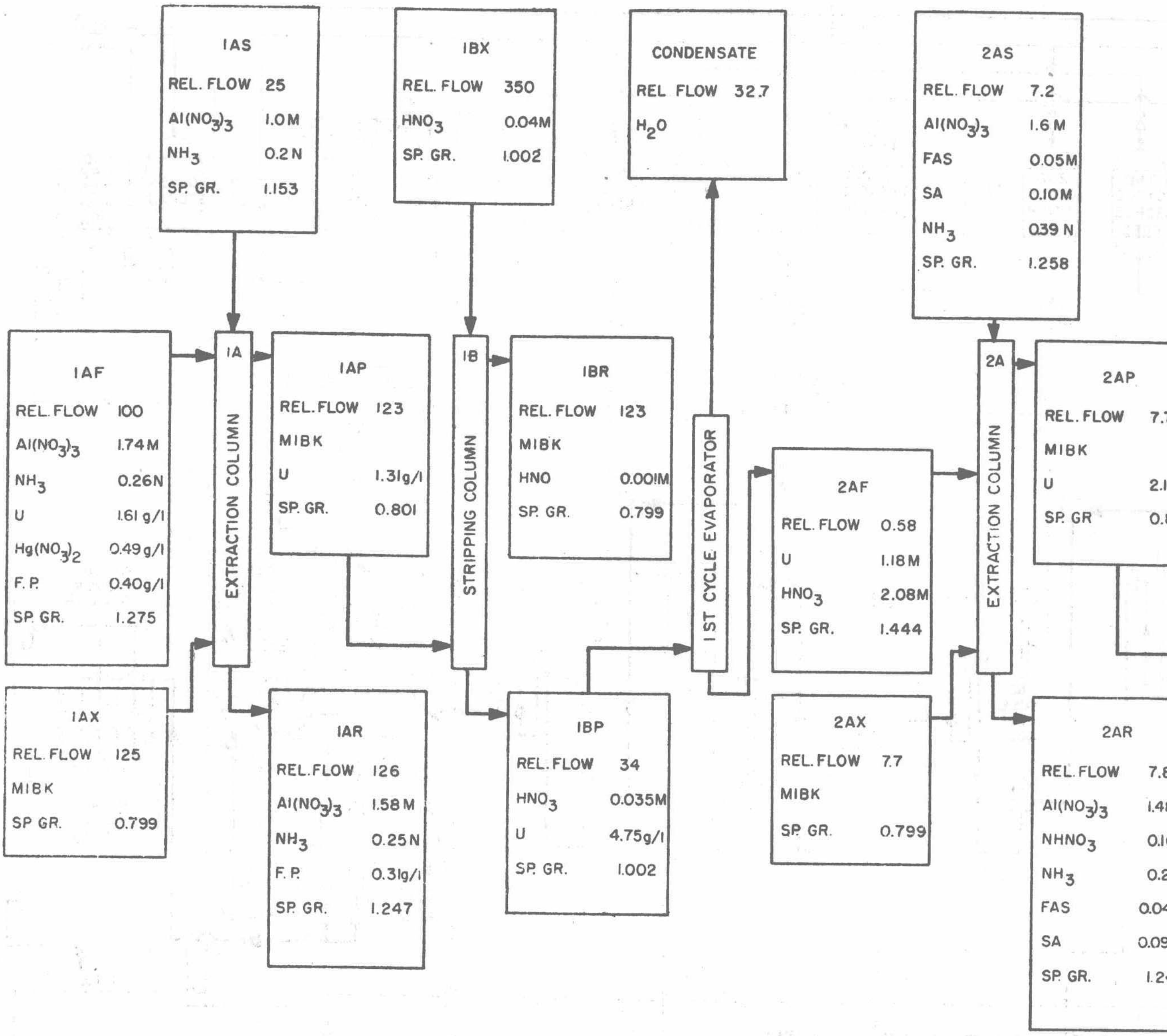

Fig. 4 MTR solvent extraction chemical flow sheet. MIBK, methyl isobutyl ketone; FAS, M, molarity; N, normality.