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NUCLEAR EFFECTS ON CHEMICAL ENGINEERING

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INTRODUCTION

This is the atomic age. Newspapers, popular magazines, and technical journals remind us of this fact every day, and maybe twice on Sundays when the special supplements and comics have their say. In atom-conscious sections of this country we may even hear fair, warmer, and roentgens per hour in every weather report. As chemical engineers, our reactions to this arrival of the age of the atom and nucleus are in many cases little different from those of the lay public. This may seem paradoxical, for after all, we have been educated in the physical sciences and should have a fair understanding of the implications of nuclear energy. The situation arises, however, from the simple fact that, although we are told this is the atomic age, there are still many amongst us (chemical engineers) who have not felt the impact of atom splitting either in business or in everyday life. Our closest contact with nuclear energy may well be during the inhalation of an infinitesimal trace of radioactivity resulting from the fallout of an atomic or hydrogen bomb which was set off in the Pacific Ocean or New Mexico or even Siberia; and in this instance we do not experience any noticeable effect. Powerful as it is and as far-reaching as it may be in its political and economic importance, nuclear energy is still somewhat remote in the daily life of the average chemical engineer.

In considering the various ramifications of nuclear energy, it is not the purpose of this paper to discuss the political effects of atomic bombs or the effect of radiation on our genes, but rather to note how chemical engineering and the professional life of the chemical engineer are being or will be changed by the developments in the nuclear field. The handwriting is on the wall and we or our successors are bound to feel the influence of nuclear energy in the field of chemical engineering. This, of course, should not be surprising because chemical engineers have already played an important part in this new area. It is estimated that some two to three thousand chemical engineers now derive their employment in the nuclear field, and it is expected that this number will increase steadily in the next few years. Chemical engineering and nuclear energy are, therefore, just as surely entwined in the future as has been chemistry and engineering in the past.

For purposes of this discussion the changes which are being brought about in chemical engineering by nuclear energy will be considered from two points of view. The first will deal with the changes in the curricula and education of chemical engineers and the second will treat the new industrial applications and techniques for which nuclear energy has been primarily responsible. Obviously, the two approaches go hand in hand with neither logically preceding the other.

NUCLEAR EFFECTS ON CHEMICAL ENGINEERING EDUCATION

It is naturally understood that many changes have been made in the courses and subject matter of chemical engineering during the past decade or two; however, our concern of the moment is only with

those which have resulted from the development of the atomic energy program. It should also be clear that not all schools have incorporated all of the following material into their teaching programs, for progress and change require time.

1. Nuclear Physics

The embryo chemical engineer is not long on the campus today before his teachers of physics and chemistry are leading him through the realm of molecules and atoms to the nucleus. Although he is probably unaware of it, considerably more emphasis is placed on nuclear physics than was done with the generation of engineers before him. It is not possible to explain the whole field of nuclear physics in a few short paragraphs, but it is rather easy to present a few simple examples of the kinds of things on which the modern student's attention is being focused. A survey of the kinds of particles of which matter is composed is usually covered early in order to give a feel for the ultimate structure of atoms and nuclei. Table I lists the most important particles which have been either discovered experimentally or postulated theoretically to explain the behavior of matter. Some of the particles are probably familiar and some are new to those who took their physics and chemistry more than a decade ago. The table is not meant to be complete for every few years there are additions, and as yet, there appears to be no end to the number and kinds of particles which physicists will discover. Of the many kinds of particles those with which the engineer is most often concerned are electrons, protons, neutrons, and alpha particles.

TABLE I

ELEMENTARY PARTICLES OF MATTER

Particle	Mass, amu*	Charge, e**	Comments
Electron	0.000549	-1	Beta particle
Positron	0.000549	+1	
Proton	1.0076	+1	Hydrogen nucleus
Antiproton	1.0076	-1	
Neutron	1.0089	0	Important in fission
Antineutron	1.0089	0	
Deuteron	2.0142	+1	Heavy hydrogen nucleus
α -particle	4.0028	+1	Helium nucleus
Neutrino	0	0	Needed for momentum balance
π -mesons	0.144 to 0.149	+1,-1,or 0	Binding material for
μ -mesons	0.113	+1,-1,or 0	neutrons and protons

* 1 amu (atomic mass unit) = 1.66×10^{-24} gm

** e (electron charge) = 4.8×10^{-10} esu

Any treatment of the behavior of particles leads directly to a discussion of radiation. As shown in Table II, radiation falls into two general classes, corpuscular (particle) and electromagnetic (no mass). The properties and sources (i.e., accelerators, radioactive isotopes, fission reactions, etc.) of the various kinds of radiation are considered, with the interaction of radiation and matter constituting one of the most fundamental aspects of nuclear science. For the average engineer the treatment must necessarily be brief, but if there is exceptional interest in the nuclear field, the student can take an elective course dealing with radiation and matter. On the theoretical side he can elect a course in quantum mechanics which helps to explain many of the phenomena, though this would rarely be done by an undergraduate chemical engineer.

Nuclear reactions are taken up after the properties of particles and radiation are understood. Table III presents several typical nuclear reactions. It will be quickly noticed here that conventional chemical reaction nomenclature is replaced by a new scheme of subscripts and superscripts. The superscript on the right side of each nuclide is its mass number, A , and is the sum of the number of protons and neutrons in the nucleus. The subscript on the left of the nuclide is the atomic number, Z , which is the number of protons in the nucleus, or the positive charge of the nucleus which, in a neutral atom, is balanced by the negative charge of the surrounding electron cloud. For the reactions of ordinary interest the sum of the superscripts and subscripts respectively must be the same on both sides of an equation when proper account is made for the coefficients modifying each term. This means that the

TABLE II

TYPES OF RADIATION

	Energy, Mev	Comments
<u>ELECTROMAGNETIC</u>		
Gamma	0.01 to 5.0	Extremely penetrating and
X-ray	10^{-4} to 10^{-3}	will ionize.
Ultraviolet	10^{-6} to 10^{-5}	Stopped by optically opaque
Visible light	10^{-6}	substances and insufficient
Infrared	10^{-7} to 10^{-6}	energy to ionize.
Cerenkov	10^{-6}	Secondary due to fast particles.
<u>CORPUSCULAR</u>		
Alpha	2 to 400	Principal particles given off
Beta	10^{-5} to 250	during radioactive decay.
Deuteron	0.07 to 3000	
Proton	0.07 to 3000	
Neutron	0.02 to 10	Liberated during fission.
Heavy charged particles	up to 6000	May be product of fission or accelerator.

TABLE III

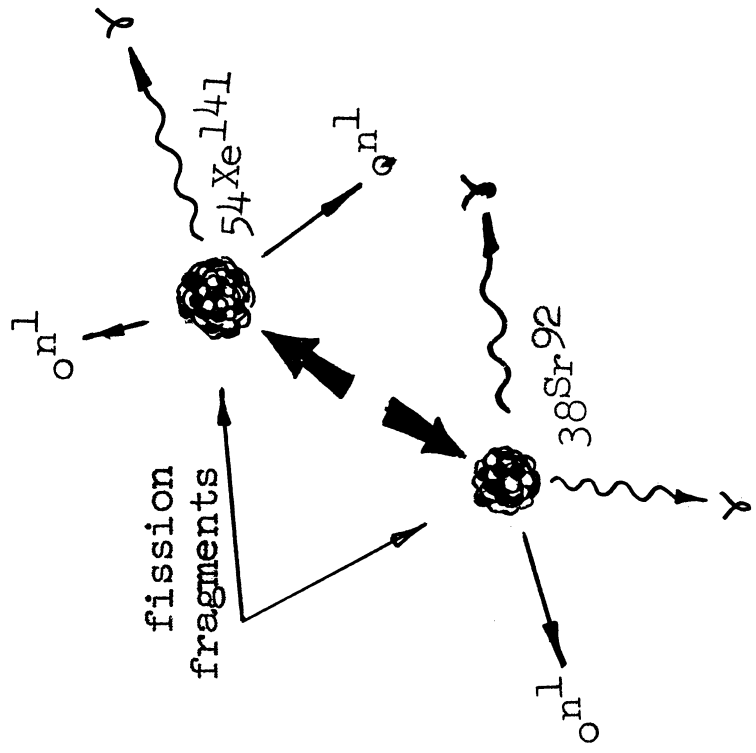
TYPICAL NUCLEAR REACTIONS

Reaction	Comments
1) ${}_{92}\text{U}^{235} + {}_0\text{n}^1 \rightarrow {}_{54}\text{Xe}^{141} + {}_{38}\text{Sr}^{92} + 3{}_0\text{n}^1$	Fission
2) ${}_{92}\text{U}^{238} + {}_0\text{n}^1 \rightarrow {}_{92}\text{U}^{239} + \gamma$ $\quad \quad \quad \searrow$ $\quad \quad \quad \rightarrow {}_{93}\text{Np}^{239} + {}_{-1}\beta^0$ $\quad \quad \quad \quad \searrow$ $\quad \quad \quad \quad \rightarrow {}_{94}\text{Pu}^{239} + {}_{-1}\beta^0$ $\quad \quad \quad \quad \quad \searrow$ $\quad \quad \quad \quad \quad \rightarrow {}_{92}\text{U}^{235} + {}_2\text{He}^4$	Scheme for production of plutonium
3) ${}_{7}\text{N}^{14} + {}_0\text{n}^1 \rightarrow {}_{7}\text{N}^{15} + \gamma$	(n, γ)
4) ${}_{7}\text{N}^{14} + {}_0\text{n}^1 \rightarrow {}_{6}\text{C}^{14} + {}_1\text{H}^1$	(n, p)
5) ${}_{3}\text{Li}^6 + {}_0\text{n}^1 \rightarrow {}_1\text{H}^3 + {}_2\text{He}^4$	(n, α)
6) ${}_1\text{D}^2 + {}_3\text{Li}^6 \rightarrow {}_2\text{He}^4$	Fusion

total numbers of protons and neutrons remain unchanged during nuclear reaction. As an example, in the fission reaction of U^{235} the mass number balance is $235 + 1 = 141 + 92 + (3 \times 1)$, and the atomic number or proton balance is $92 + 0 = 54 + 38 + (3 \times 0)$.

Figure 1 illustrates schematically one of sixty possible nuclear reactions which occur when fission occurs in U^{235} . This figure shows two fission fragment nuclei being material products, three prompt neutrons which are available for sustaining fission and gamma radiation. Certain fission fragments are neutron producers. Such neutrons are termed "delayed" neutrons and provide one means for mechanical control of thermal and intermediate neutron energy reactors.

The conditions under which the various nuclear reactions proceed are worthy of considerable attention, particularly in the case of the fission reaction which is more or less responsible for the growth of the nuclear field. Understanding the sustaining of a fission reaction requires a knowledge of fast and slow neutrons and their diffusion, neutron production and absorption, nuclear properties of materials, and criticality. An intensive study of these factors can be given only in a special course which again would be elected by very few undergraduate students. In addition to fission, particular attention is paid to disintegration reactions of single isotopes, which are responsible for radioactivity. This is exemplified in Table III by ${}_{93}\text{Np}^{239}$ which decays to ${}_{94}\text{Pu}^{239}$ while emitting a beta particle (electron). The half-life of ${}_{93}\text{Np}^{239}$ is 2.3 days while that of ${}_{94}\text{Pu}^{239}$ is 24,000 years, so the relative stability of these two isotopes can easily be compared.



incident neutron \rightarrow

U235 nucleus

Key:

- Proton
- Neutron
- ~ Gamma Ray

Figure 1. Representative Fission Reaction

For ordinary chemical reactions, the periodic chart of the elements has been most useful. In the same way nuclear reactions can be better understood and correlated on a chart of the nuclides. The General Electric Chart is typical of several which have been developed and a section of this is shown in the upper part of Figure 2. By means of the lower part of Figure 2, one can see how a given nucleus may be transformed to another via various nuclear reactions.

In the course of studying particles, radiation, and nuclear reactions, it is expedient to include some discussion of the instruments used for detection. Table IV lists the more important instruments and techniques used to detect and measure radiation. The principles behind the instruments are discussed briefly and an occasional laboratory experiment may be run to demonstrate their use. For example, a simple ionization chamber or Geiger-Muller experiment may be carried out on the radiation from a luminous-dial wrist watch.

The coupling of instrumentation for particle detection and measurement with more conventional instrumentation and control systems for processes requires extensions of knowledge and experience for the process engineer.

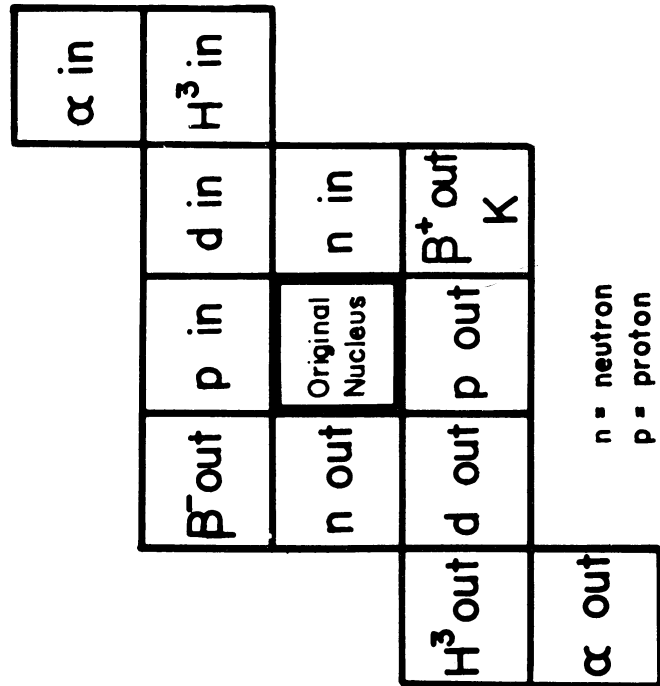
Consider a simple system of a reactor, heat exchanger and pump loop for transport and transfer of nuclear process heat for steam generation. The systems control and instrumentation to couple heat generation and conversion with nuclear measurements impose new problems and extension of knowledge. Such a systems control is illustrated in the Figure 3.

TABLE IV

INSTRUMENTS AND TECHNIQUES FOR DETECTION OF PARTICLES AND RADIATION

Instrument or Technique	Comments
Cloud chamber	Observe nuclear reactions in vapor
Bubble chamber	Observe nuclear reactions in liquid
Ionization chamber	Detect radiation by ions formed
Pulse instrument	Detect radiation in pulses
Proportional counter	Pulse heights depend upon energy
Geiger-Muller	All pulses same magnitude
Scintillation counter	Radiation detected through light emitted
Photoelectric cell and photomultiplier tube	Absorption of radiation photons causes electrons to be discharged
BF ₃ proportional counter	For detection of neutrons
Fission ionization chamber	For detection of neutrons
Neutron thermopile	For detection of neutrons
Photographic emulsion	Detects all forms of radiation
Foil activation	Determine radioactivity induced by neutrons
Chemical dosimeters	Measure radiation by chemical reaction
<u>Associated electronic devices:</u>	
Linear amplifier	Amplify extremely small currents or pulses
Scaling circuits	Select certain percent of pulses
Discriminator	Select certain energy pulses

Relative Locations of the Products
of Various Nuclear Processes



- n = neutron
- p = proton
- d = deuteron
- α = alpha particle
- β^- = negative beta particle
- β^+ = positive " "
- K = K-electron capture

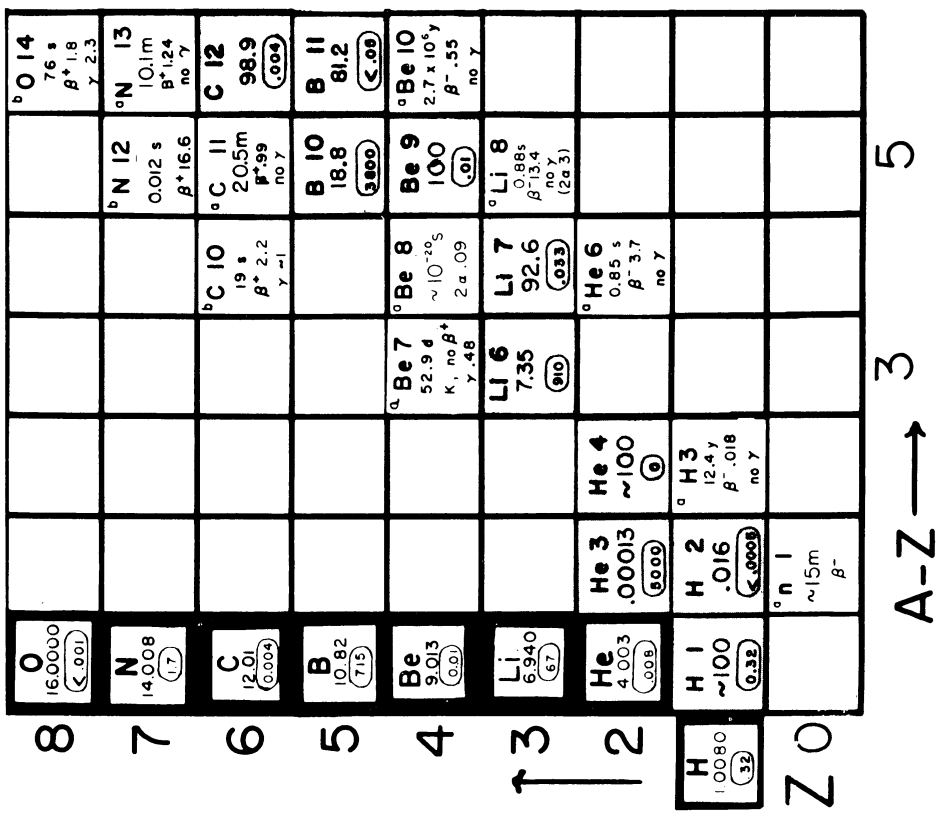
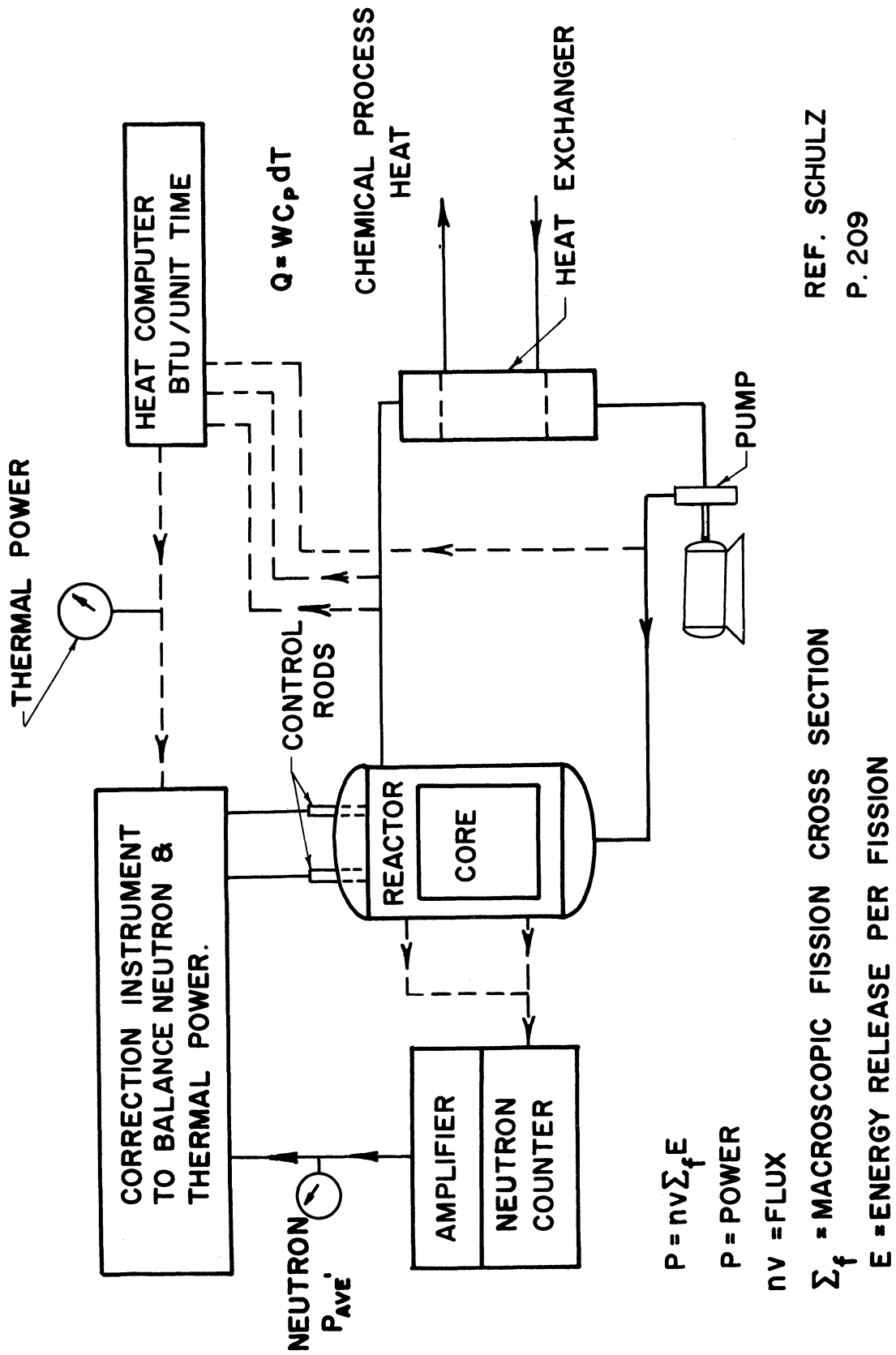


Figure 2. Nuclide Chart



REF. SCHULZ
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Figure 3. Control System for Process Heat Reactor

This diagram illustrates a typical nuclear-heat system wherein a continuous calibration of reactor power level is required. A heat computer can be used to measure the heat extracted by the heat exchanger. In essence, such a measurement is achieved by computing the heat extracted by determining coolant flow rate and temperature differences. The neutron power is determined by one or more neutron measuring devices. The neutron measurements are amplified and correlated with the heat or thermal power by a comparator (which corrects and calibrates neutron power with heat power). The output of the comparator feeds back and corrects the neutron detection. The resultant measurements can be used for adjustment and control movements. Similarly, in an electrical powerplant, the feedback from turbine throttle can be coupled into the nuclear system for adjustment of reactor power level.

2. Thermodynamics

The engineer further requires insight into thermodynamics in terms of nuclear energy. Although it was undoubtedly introduced earlier, the Einstein concept that energy is merely another manifestation of matter will be emphasized, and the first law of thermodynamics re-stated to say that it is the sum of mass and energy which is always conserved. A simple calculation can be made from the well-known equation, $E = mc^2$, to show that the mass change in an ordinary chemical reaction is so negligible as to be beyond detection. Thus, if a fuel with a heating value of 20,000 Btu/lb is burned, the loss of mass from the system containing the fuel and its required air is

$$m = \frac{E}{c^2} = \frac{(20,000 \text{ Btu})(980 \text{ gm-cm/gm force-sec}^2)}{(3 \times 10^{10} \text{ cm/sec})^2 (454 \text{ gm/lb})(9.486 \times 10^{-11} \text{ Btu/dyne-cm})(980 \text{ dyne/gm force})}$$
$$= 5.2 \times 10^{-10} \text{ lb}$$

It is clear that this is far beyond the ability of the finest balances to detect, so that it is justifiable to say that in chemical reactions there is no change of mass. In nuclear reactions the associated energy changes are of the order of a million times those of chemical reactions. Appreciable energy changes are, therefore, encountered and must be taken into account. A very interesting aspect of the Einstein relation is that if both the system and its surroundings to which energy is transferred are considered, there is no loss of mass whatsoever. This is because the mass loss of the system due to its energy loss is exactly matched by the mass gain of the surroundings due to the energy gain of the surroundings. Except for energy in transit, the total mass of the universe is constant. From another point of view, one can say that for ordinary so-called "pure energy" transfers there is always an accompanying transfer of mass, though the transfer of mass is normally of minute proportions. This obviously is the reason the Einstein relation has not been given much attention in thermodynamics courses until the development of the nuclear energy program where nuclear reactions involving tremendous energy transfers are common rather than rare.

3. Unit Operations and Control

The chemical engineer also feels the impact of nuclear energy in the field of unit operations and process control. Special requirements have forced the chemical engineers working in the nuclear field to develop new equipment and new procedures as solutions to its problems. Table V presents some of the more important of these operations which are gradually being absorbed into modern chemical engineering courses. In many cases the operations are being adapted to conventional processes. It can be expected that within a few years most of these operations will be as familiar to chemical engineers as ordinary heat transfer.

TABLE V

CHEMICAL ENGINEERING OPERATIONS AND EQUIPMENT
DEVELOPED IN THE NUCLEAR FIELD

Operation or Equipment	Comments
Barrier diffusion	Principle means of obtaining U ²³⁵
Gas centrifugation	For general isotope separation
Gas thermal diffusion	For general isotope separation
Pulse columns	For liquid-liquid extraction
Ultra-low-temp. distillation	For separating heavy hydrogen
Ultra-high-temp. distillation	For purifying metals
High decontamination evap.	For removing radioactivity from solutions
Liquid metal heat transfer	For heat transfer at high temperature
Electromagnetic pumps	For moving liquid metals
Pressurized water systems	For improving heat transfer
Remote control handling	For operations behind shielding
Radiation sensing gauges	For liquid-level, thickness, and flow meas.
Micro-analytical control	Process control with micro amounts
Neutron-analytical control	Control standards based on neutron absorption
Materials of construction resisting radiation damage and with low neutron absorption	Materials required for the cores of nuclear reactors and for use in regions of intense radiation
Special storage systems	For storing radioactive materials safely
Radiation chemistry and irradiated catalysts	Special attention is given to radiation as a potential process variable
Ion exchange	Unusual applications in intense radiation

NUCLEAR EFFECTS ON PROCESS INDUSTRIES AND DESIGNS

The chemical engineer practicing his profession in any one of the many industries contributing their efforts to the enhancement of nuclear energy, finds it necessary to broaden his background and fields of interest as well as probe into the depths of knowledge. In general, areas requiring extension of knowledge might be classified as follows:

- a. Nuclear physics
- b. Operational mathematics
- c. Reactor theory
- d. Interactions of radiation and matter
- e. Nuclear instrumentation and control
- f. Shielding
- g. Nuclear process design

Such increase in depth of knowledge and broadening of background and experience are essential for the achievement of nuclear processes which are technically feasible, "safe in operation," and possess economic potential with maximum flexibility.

To illustrate such "nuclear" effects on systems and processes that involve radiation, neutrons, particles and neutron reaction rates in an overall system, the following areas have been selected for discussion.

1. Preparation of Fissile and Fertile Materials

Uranium and thorium are found in nature in concentrations comparable to the range of concentrations of chemical waste streams. As an example, in many uranium deposits, such as the carnotite ores, the uranium concentration will range from .1 to 1%. Processes must be

developed wherein this low grade material is upgraded to a purity of 99.99% and in which isotopes are separated in relatively pure form. Present day practice consists of the following steps.

a. Ore Concentration.--In ore concentration, the chemical engineer has been required to develop process systems in which micro-chemistry is conducted in production-scale flow systems where many degrees of conversion are required in the separations technologies employed. As an example, in a solvent extraction system, the solvent extraction columns for the recovery of uranium must provide a product of uranium which has a purity of 99.99% or better while the losses of uranium from the system must be accountable within 0.1%.

While such processing improvements and higher degrees of purification are being effected, the chemical engineer must give consideration to radioactive contaminants in certain components existing in the ore. A good example is the uranium ores obtained from the Belgium Congo which contain significant quantities of radium so that, while upgrading the concentrate, consideration must be given to the hazards of radiation and the provisions of structural shields for the protection of health and radiation safety.

Figure 4 illustrates some of the nuclear effects involved in the treatment of uranium ores which contain limited quantities of radioactivity. The presence of radiation not only permits discovery of new ore deposits but must be considered in terms of radiation health, control of radioactive particulates, partial shielding as radioactive materials become more concentrated, methods for remote mechanical handling, methods for decontaminating equipment, structures, clothing

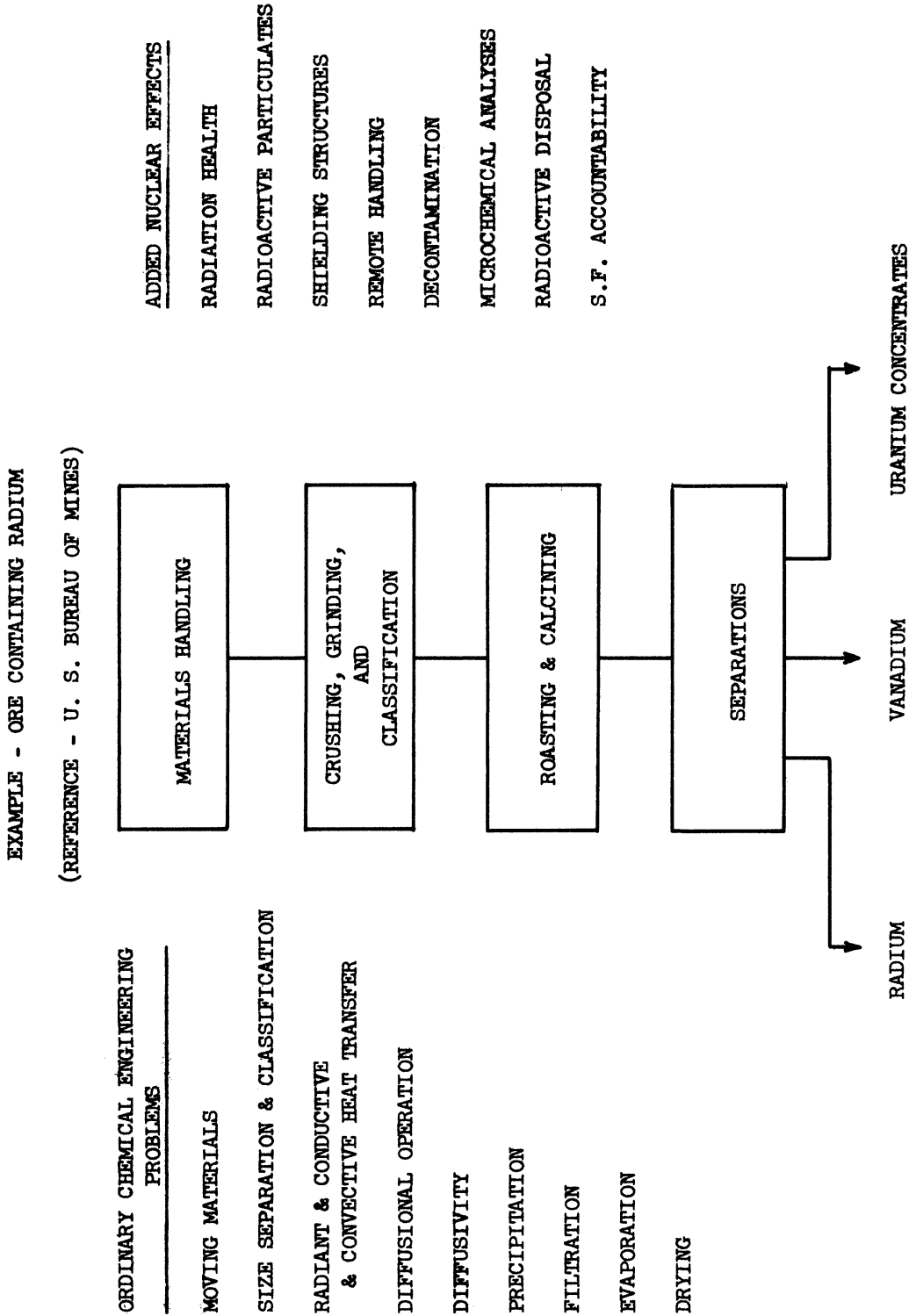


Figure 4. Nuclear Effects on Chemical Engineering - Milling and Concentration of Uranium.

and wastes. In addition to the problems of radiation, the process concepts must be extended to provide means for essentially complete recovery of all the uranium in as pure a form as possible.

b. Conversion of Uranium to Nuclear-Grade Metal, Metal Oxides, Metal Fluorides, and Other Compounds.--The processes which have been developed for such concentration involve basically reaction rate data, solvent extraction, evaporation, denitration, hydrofluorination, and reduction operations as well as fluorination reactions. Such concentrations have a two-fold application: (1) the preparation of a material in such chemical form that isotopic separations of uranium-235 from uranium-238 will result; (2) the production of nuclear-grade structural materials such as natural uranium fuel for power reactors, breeder blankets for fast reactors, and special shielding materials.

Figure 5 illustrates some of the nuclear effects in achieving feasible process designs for feed materials preparation and nuclear-grade natural uranium for reactors. This figure illustrates a typical dissolution, solvent extraction and conversion process for production of uranium as metal, oxide, or hexafluoride. Since the product has high value the process requires complete accountability of uranium in all process streams (which in essence is microchemical control in production processes), separation factors for recoveries of 99% or higher, unusual control of waste streams, and special materials of construction.

In each of these cases, the processes must be conducted under conditions so that the product possesses purities normally foreign

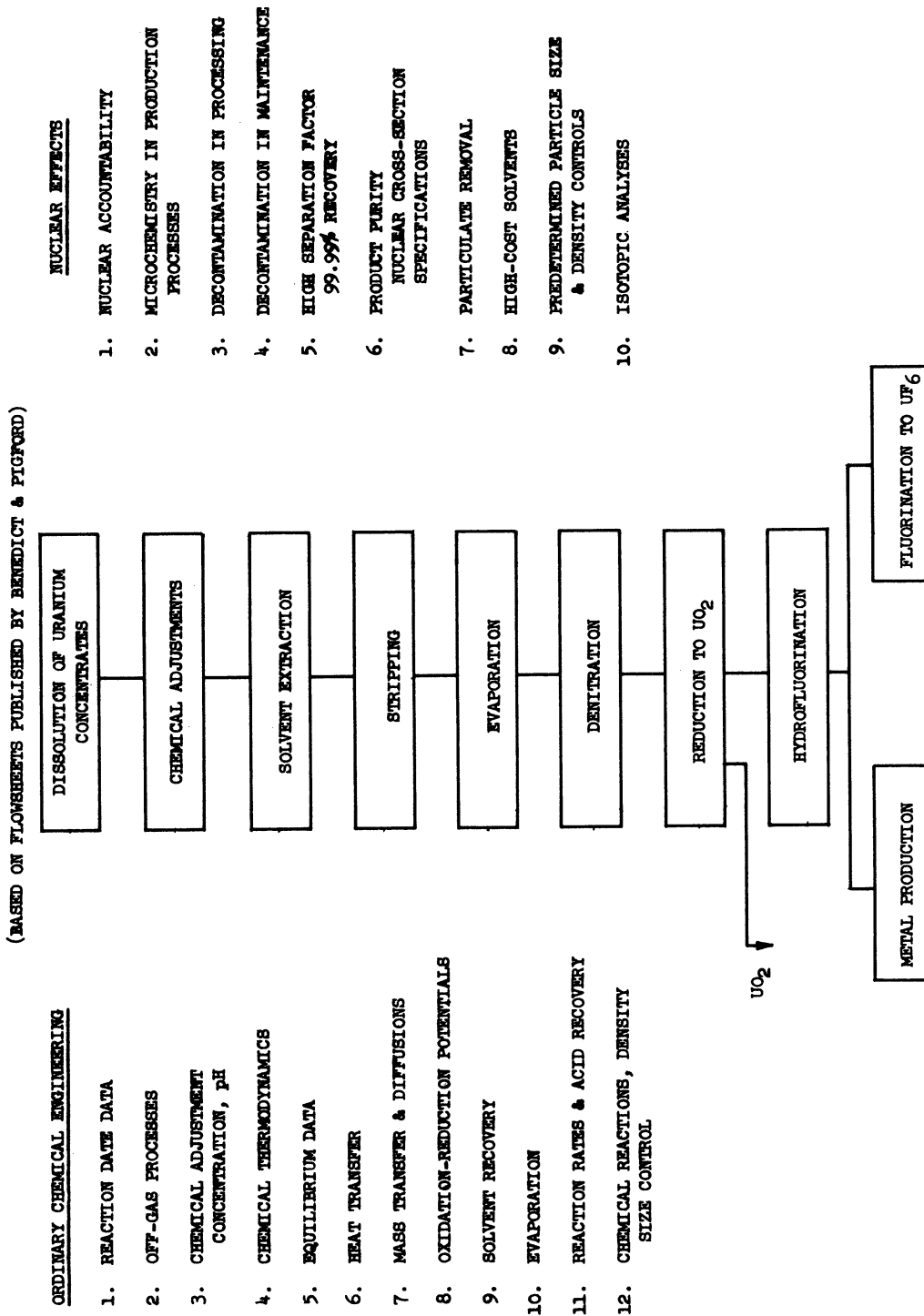


Figure 5. Nuclear Effects on Chemical Engineering - For Uranium Purification.

to chemical engineering processes. The recoveries must be complete so that waste solutions do not prompt pollution problems in either atmosphere or in liquid form. Since the value of the products lies somewhere in the range from \$2 to \$100 per pound, unusual precautions must be taken in process inventory, process recovery, and product handling systems.

c. Gaseous Diffusion.--Most power reactors in the United States, at least, are directed toward a degree of isotopic enrichment of the fissile species, uranium-235, so that smaller, more compact reactor systems can be achieved. Isotopic separations in the main consist of the conversion of uranium to uranium hexafluoride which, as a gas, can be cascaded through several thousand diffusion cascades to enrich uranium-235 from 0.71% to some specified enrichment lying somewhere between 1 to 93.4% as uranium-235. The diffusion cascades, as a consequence, require special materials of construction, special diffusion barriers, and numbers of cascades ranging in the thousands.⁽¹⁾

Figure 6 serves to illustrate some of the special nuclear problems which must be considered in view of process engineering in isotopic separations employing gaseous diffusion and resultant conversion of uranium hexafluoride to fuel form.

In addition to applying the nominal chemical engineering principles to the gaseous diffusion cascade attendant with the numerous problems of pumps, controlled atmosphere, materials of construction, etc., nuclear problems have been imposed. As the uranium becomes enriched in the isotope U-235, there is an increasing problem of geometry so that the system will be maintained as a subcritical system throughout the process. The chemical engineer, therefore, must be in a position

ORDINARY CHEMICAL ENGINEERING

1. DIFFUSION RATES
2. SEPARATION FACTORS
3. MINIMUM VS. PRACTICAL STAGES
4. STAGE EFFICIENCIES
5. HEAT TRANSFER
6. CORROSION
7. GAS HANDLING
8. FLUID MECHANICS
9. RATE THEORY
10. MATERIALS HANDLING

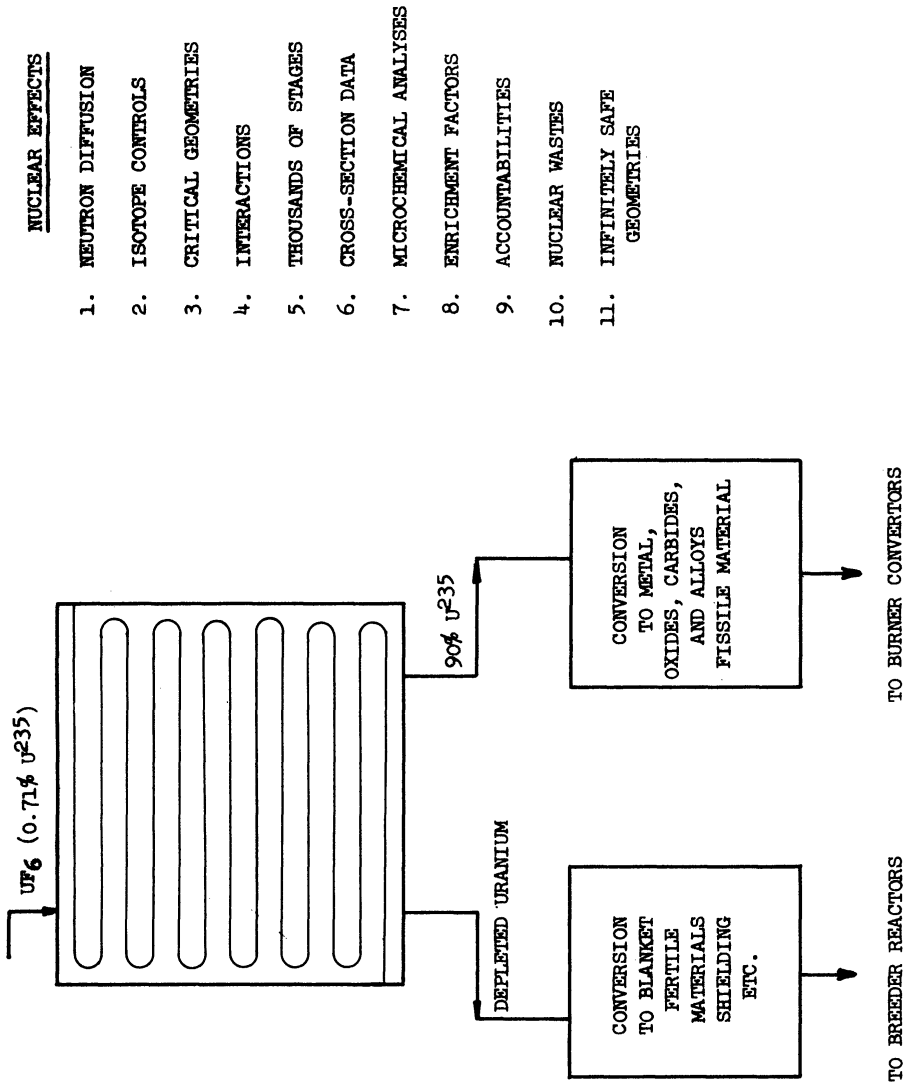


Figure 6. Nuclear Effects on Chemical Engineering - Isotope Enrichment and Conversion to Reactor Fuel.

to calculate critical geometries, critical mass, neutron interreactions, nuclear diffusional phenomena, and many other problems unique to the nuclear energy program.

The conversion of uranium hexafluoride to metals, metal oxides, metal carbides and other uranium compounds imposes new problems on the chemical engineering process systems employed for such conversion. Since 1 gram of uranium-235 is equivalent to about 3 tons of coal, or 600 barrels of fuel oil, the chemical process must be one which accounts for every gram of uranium-235 in the system. For fully enriched uranium systems, the processes must be conducted in equipment which is either "geometrically safe" at all times or in "unsafe geometries" wherein uranium-235 concentrations are controlled in each step of the process. The new knowledge, therefore, involves a high degree of recovery coupled with criticality controls. Special processing techniques and methods must be employed for the physical handling, packaging and shipment of an enriched product for reactor fuel fabrication.

d. Fabrication of Reactor Fuel Elements and Reactor Fuel Solutions.--Since the nuclear power programs include reactors which have fuels in solid form, as aqueous salt solutions of uranium, and liquid metal solutions of uranium, the processing techniques for assemblage and preparation of reactor fuels has had considerable effect on the chemical engineering process system. These include: (1) selection of fuel form, such as uranium metal vs. uranium dioxide or some other compound; (2) the selection of structural cladding material because of the chemical instability of uranium and uranium compounds

in reactor coolants, as well as the containment of fission products in a power reactor; (3) the analysis of impurities which have high probabilities for neutron absorption in both uranium fuel-bearing materials and fuel structural materials; (4) chemical and metallurgical bonding of fuel structural materials to fuel materials for optimum heat transfer; (5) control of atom concentrations as a function of temperature limits of the system, solubilities as temperature functions, and thermal stress problems in transient behavior systems.

In the areas of nuclear energy which involve the conversion of fissile and fertile materials to reactor fuel form we see that the chemical engineer in process designs and process developments must give consideration to an increased number of new variables which basically involve nuclear interreactions, as contrasted to chemical reactions, such as radiation problems, shielding, higher degrees of purity than heretofore required, and new principles of health and safety.

2. Reactor Power Systems and Reactor Developments

Nuclear reactors are assemblages of fissile and fertile materials coupled with coolants, reflectors, moderators, neutron and biological shields, wherein the assembled system produces heat with unlimited temperature ranges, neutrons, radioactive fission products, beta particles, gamma rays, neutrinos, and new fissile species. Such nuclear heat source devices are useful in the generation of power, chemical process heat, radioisotopes, fissionable materials production and an unusual flexible combination of useful purposes. Alvin Weinberg, Oak Ridge National Laboratories, has indicated that there are possibly

20 to 25 thousand possible reactor concepts. He further states that there are several hundred physical concepts worthy of consideration, of which perhaps 10 to 20 may result in economical power plants and heat sources. In order to reduce the number of theoretical concepts to practically feasible systems which possess economic potential, it becomes necessary to combine conventional engineering disciplines with comprehensive understanding of nuclear phenomena. Many chemical engineers have adapted their backgrounds and knowledge with experience and qualification to make major contributions in this field.

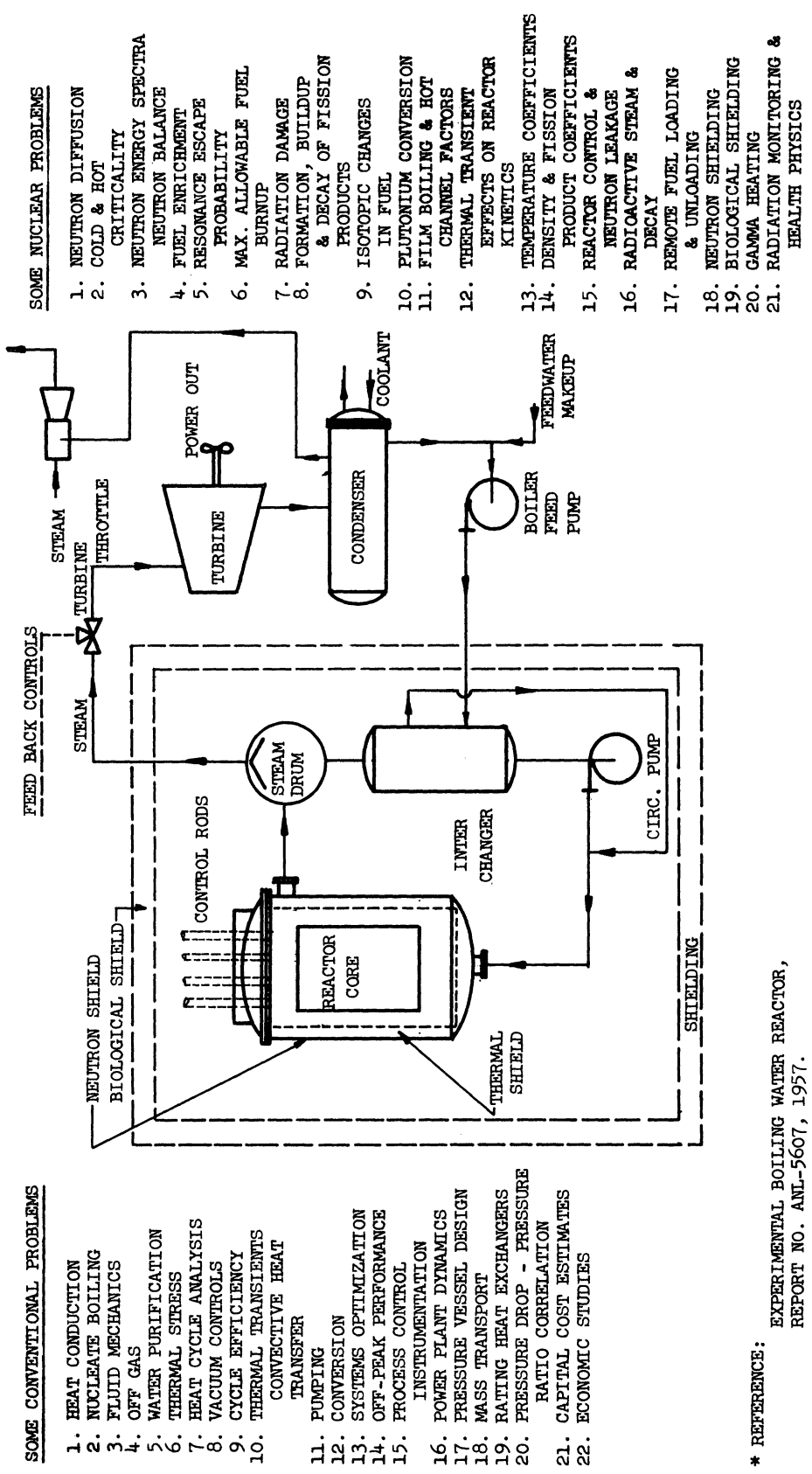
It is interesting to note that the generalized diffusion equations and slowing down models have had to rely fundamentally upon the early developments of heat conduction equations and principles of mass transfer. As an example, the time-rate of change of neutron density in a given volume is equivalent to the number of neutrons absorbed in a given volume per second plus the net number of neutrons escaping from a given unit volume per second, less the number of neutrons which are produced in that given volume. Mathematically, we write such an expression as follows:

$$-\frac{\partial n}{\partial t}(\vec{r}, t)d\vec{r} = \sigma_a C(\vec{r}, t)d\vec{r} + \vec{\nabla} \cdot \vec{j}(\vec{r}, t)d\vec{r} - S(\vec{r}, t)d\vec{r}$$

Solutions to such general mathematical expressions are dependent upon the relationship between neutron currents and the gradient of flux in terms of Fick's Law and bear a similarity to the equations developed for heat, mass and momentum transfer. By the insertion of proper boundary conditions and separations of variables, it is possible to obtain solutions which predict the gradients of neutron flux and the

critical parameters of the system. In order to solve such problems, it requires that the neutron balances and fluxes be determined in proper energy groups, dependent upon the selection of the materials comprising a nuclear power system. Upon obtaining reference design considerations which define the dimensions of a power reactor fuel, the fuel concentration, and the selection of reflectors, it is necessary to couple such nuclear variables with variables of heat transmission, variations in coolant flow and changes of fuel with time. Although it is not possible to discuss these interrelationships in detail and since they vary from one reactor concept to another, Figure 7 indicates a typical power reactor system using the boiling water principle as developed by the Argonne National Laboratories and as now being developed extensively by the General Electric Company for application to power reactors. It should be noted from this figure that this is a typical boiling water heat cycle for the generation of power in which the heat source component is a reactor and conventional saturated steam turbomachinery comprises the powerplant. The conventional engineering problems involved in the design of such a powerplant are indicated in the figure and include considerations of heat conduction, nucleate boiling, fluid mechanics, methods and processes for purification of water, thermal stress considerations, heat cycle analysis, process controls and instrumentation, and other major problems. In addition to these conventional problems, the "reactor process engineer" must depend in the main upon new knowledge which must be coupled with the conventional process variables in which he is concerned with the effects of criticality and excess multiplication constants in terms of all of the

A TYPICAL POWER REACTOR SYSTEM*
 EXAMPLE: BOILING WATER - STEAM CYCLE POWER PLANT
 NUCLEAR EFFECTS ON



SOME CONVENTIONAL PROBLEMS

1. HEAT CONDUCTION
2. NUCLEATE BOILING
3. FLUID MECHANICS
4. OFF GAS
5. WATER PURIFICATION
6. THERMAL STRESS
7. HEAT CYCLE ANALYSIS
8. VACUUM CONTROLS
9. CYCLE EFFICIENCY
10. THERMAL TRANSIENTS
CONVECTIVE HEAT
TRANSFER
11. PUMPING
12. CONVERSION
13. SYSTEMS OPTIMIZATION
14. OFF-PEAK PERFORMANCE
15. PROCESS CONTROL
INSTRUMENTATION
16. POWER PLANT DYNAMICS
17. PRESSURE VESSEL DESIGN
18. MASS TRANSPORT
19. RATING HEAT EXCHANGERS
20. PRESSURE DROP - PRESSURE
RATIO CORRELATION
21. CAPITAL COST ESTIMATES
22. ECONOMIC STUDIES

* REFERENCE: EXPERIMENTAL BOILING WATER REACTOR,
 REPORT NO. AML-5607, 1957.

SOME NUCLEAR PROBLEMS

1. NEUTRON DIFFUSION
2. COLD & HOT
CRITICALITY
3. NEUTRON ENERGY SPECTRA
NEUTRON BALANCE
4. FUEL ENRICHMENT
5. RESONANCE ESCAPE
PROBABILITY
6. MAX. ALLOWABLE FUEL
BURNUP
7. RADIATION DAMAGE
FORMATION, BUILDUP
& DECAY OF FISSION
PRODUCTS
9. ISOTOPIC CHANGES
IN FUEL
10. PLUTONIUM CONVERSION
11. FILM BOILING & HOT
CHANNEL FACTORS
12. THERMAL TRANSIENT
EFFECTS ON REACTOR
KINETICS
13. TEMPERATURE COEFFICIENTS
14. DENSITY & FISSION
PRODUCT COEFFICIENTS
15. REACTOR CONTROL &
NEUTRON LEAKAGE
16. RADIOACTIVE STEAM &
DECAY
17. REMOTE FUEL LOADING
& UNLOADING
18. NEUTRON SHIELDING
19. BIOLOGICAL SHIELDING
20. GAMMA HEATING
21. RADIATION MONITORING &
HEALTH PHYSICS

Figure 7. A Typical Power Reactor System.

variables of the system. There have been listed 22 areas to which a process engineer must give consideration when dealing with the development of power reactors themselves. As a result of coupling conventional engineering and fully realizing the necessary effects from nuclear technology, it becomes possible for the chemical engineer to give consideration to multiple purpose nuclear heat power systems, as illustrated in Figure 8. This is an example of a typical high temperature, nuclear heat source adapted for possible application in a typical chemical or petroleum plant wherein it is desirable to utilize nuclear process heat for chemical reactions while producing high temperature, high pressure process steam and the necessary power requirements for a given number of chemical process systems. In order for such a nuclear heat power concept to have utility in chemical and petroleum product manufacture, it becomes necessary to optimize simultaneously a complete economic balance around the chemical plant while giving consideration to the interreactions of neutrons, gamma radiation, and fission fragments for certain specific types of useful chemical products.

3. Production and Processes for Nuclear Materials

Another very important area for the successful growth of nuclear technology which has had significant effects upon the chemical engineering profession is the processing of special nuclear materials and the production of nuclear-specification products for use in reactor programs. These processes and productions might be identified as follows.

a. Processes and Production of Zirconium and Hafnium.--

Zirconium metal has useful applications as fuel element cladding material in many types of reactor systems. In addition to useful chemical, physical, mechanical and metallurgical properties zirconium has an

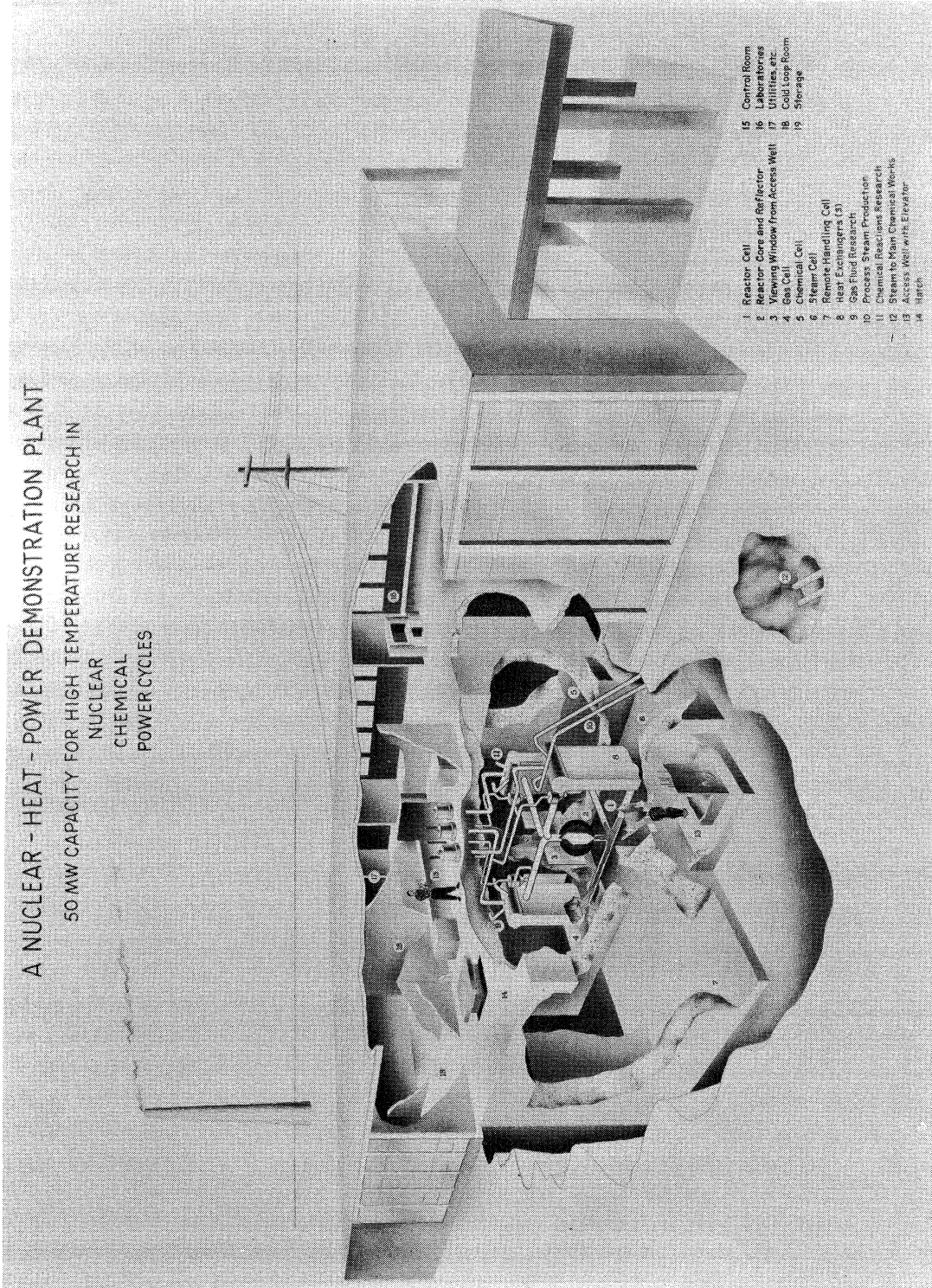


Figure 8. Nuclear - Heat - Power Demonstration Plant

absorption cross section in the thermal neutron energy range of 0.18 barns. To achieve a reactor material which has an absorption cross section in this range, it has been found necessary to develop chemical processing techniques for the selective separation of hafnium from zirconium. Although a number of separation techniques have been employed for such separations, the solvent extraction of thiocyanates with hexone (ref. Benedict and Pigford, p. 169) illustrates the nuclear effects upon chemical technology. This method, in principle, consists in separating hafnium from zirconium by means of such a solvent as hexone so that the hafnium concentrates preferentially in the organic phase while leaving the zirconium in the aqueous phase. By giving consideration to the nuclear properties of the system, it became possible to produce commercially relatively large quantities of hafnium-free zirconium which contains less than 0.005% of hafnium. Thus, a process was effected whereby the zirconium became a useable nuclear fuel construction material and the hafnium possessed properties as excellent control rod material for reactors.

b. Production of Deuterium and Heavy Water.--Since heavy water is an excellent moderator for those types of reactors where a high degree of thermalization is desired, such as a natural uranium reactor, the process engineer was confronted with the problem of developing process systems whereby a high degree of purity of either deuterium and/or heavy water could be obtained for use in such types of systems. Processes have been developed for the production of heavy water at production rates and costs which make certain types of natural uranium reactors feasible.

c. Process and Production of Special Solvents.--In

solvent extraction technologies using aqueous systems for the recovery of uranium, thorium, plutonium, and radioactive nuclides the nuclear technology has imposed the problem on the chemical engineer to develop special types of solvents of high purity, such as diethyl ether, hexone, tributyl phosphate, special kerosene fractions, chelating agents, and special materials with variable oxidation reduction potentials.

4. Reprocessing and Recovery of Nuclear Fuels

The power reactor programs throughout the world involve numerous types of fuels concepts for reactors in which each specific type requires special consideration of metallurgy and materials engineering, unique to a given specific type of reactor. In addition, the reactor types are:

Fissioning reactors or burnup reactors

Convertor single-region reactors

Fast breeder reactors

Thermal breeder reactors

Consequently, the process engineer who is required to develop systems by which fissile and fertile materials can be recovered, recycled and/or sold, and dispose of radioactive fission products in gaseous, liquid, and solid form is confronted with combining special nuclear problems with conventional chemical engineering. Each type of reactor fuel, therefore, possesses challenges of unique problems in which the impact of nuclear technology can control the selection of process variables, capital and operating costs. There are numerous chemical engineers practicing professionally today who have acquired unusual and unique

backgrounds for the development of process systems for the separation and recovery of various materials discharged from nuclear reactors.

Floyd L. Culler (ref. General Economics of Chemical Reprocessing Using Solvent Extraction Processes, 1958 Nuclear Congress, Preprint 22) has discussed a number of aqueous chemical systems which involve the coupling of dissolution and solvent extraction for the separation of fissile and fertile materials from fission products.

S. Lawroski, of Argonne National Laboratories, has presented information describing special volatility techniques for conversion of uranium to uranium hexafluoride, permitting either recycle to gaseous diffusion or reconversion back to fuel form.

Ercole Motta, North American Aviation, has discussed aspects of pyrometallurgical techniques wherein high degrees of decontamination have been achieved by special metals extraction processes.

The Brookhaven National Laboratories have developed unique systems for the recovery of fissile and fertile materials from liquid metal-fueled reactors.

Although it is not possible to discuss the enumerable multiple materials and metallurgies confronting such engineers, an example will illustrate the nature of the impact of nuclear technology on process considerations. Figure 9 indicates in general the type of remotely operated reprocessing system for a specific type of fast power breeder reactor system. Such a system is considered extremely important to the fissile and fertile materials power economy of this nation and of modern civilization. Chemical and nuclear engineers have coupled their efforts in several locations throughout the country whereby

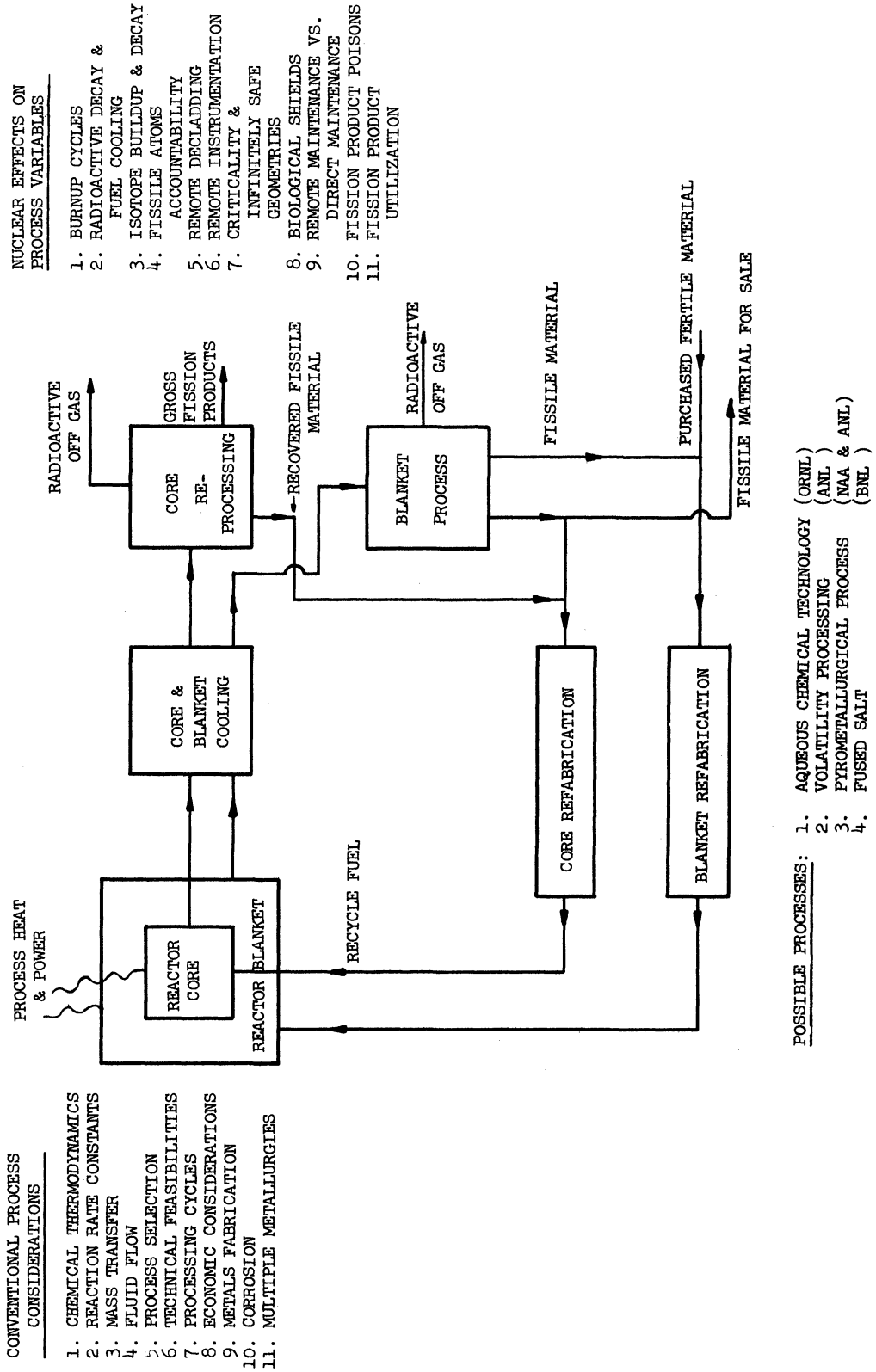


Figure 9. Remotely Operated Reprocessing Systems - Impact of Nuclear Technology.

reprocessing systems and unique chemical and metallurgical processes are combined with nuclear considerations.

A typical fast reactor system is comprised of a highly enriched core surrounded by a depleted blanket. Such a fast reactor system requires rapid recycle with minimum inventory of fissile materials external to the reactor. Thus, the processing engineer is confronted with the recovery of fissile and fertile material with minimum cooling and decay time, introducing special engineering problems.

The objective is to recycle and return the fissile materials from the reactor core with optimum removal of fission products and to combine such core fissile material with that portion of the fissile materials produced in the reactor blanket to sustain power reactor operations.

The conventional process considerations cover the range from chemical thermodynamics to the technical and economic feasibility evaluations of selecting a particular process suitable for a given type or types of reactor. In order to select such optimum systems, it becomes necessary to couple with such conventional process considerations detailed and complete evaluations of reactor burnup cycles, radioactive decay and fuel cooling, special isotopic buildups resulting from neutron capture (such as protoactinium in the thorium series and neptunium in the uranium series), a high degree of accountability of fissile atoms, possible methods for remote decladding, remote instrumentation, and unusual precautions for determining geometries in which critical configurations are not possible until the materials are

returned to the reactor. In addition, the design criteria must be established to give consideration to health and safety, in-process inventory control, optimization of equipment designs in terms of normal process variables, coupled with nuclear variables, and the processing concepts for the separation and disposal of radioactive fission products. Again, we see that it is not possible to achieve a practical and workable system or to make engineering applications of technical data for reprocessing reactor fuels unless the background knowledge, experience and qualifications of the engineers are extended to include the nuclear parameters.

CONCLUSIONS

The achievement of a national and probably a world-wide energy and power economy will depend largely upon the abilities of scientists and engineers to convert data, information, conceptual engineering and operational principles of numerous individual processing systems integrated into an overall energy production and conversion system whose technical and economic feasibilities depend upon extensions of knowledge. Such data, concepts and demonstration programs must be reduced to a production technology which is compatible with our social, economic and political changes.

Such reduction to production technologies can be achieved only through coupling science with engineering in all phases of developments. Achievement of this kind cannot be attained by our producing only highly educated and trained scientists, mathematicians and engineers whose breadth of knowledge is limited to only narrow fragments of specialization in one area of knowledge. Such high degrees of

specialization must be knit together into overall systems concepts. Hence, in addition to the highly specialized scientist and engineer on one hand, and the general engineering practitioner on the other, we have a need to develop, educate, and train technical men whose backgrounds, knowledge and experience cross the spectrum of the disciplines of nuclear science and engineering, economics, social and health problems. Such a man might be termed the "nuclear process engineer."

REFERENCES

1. Benedict, Manson and Pigford, Thomas, Nuclear Chemical Engineering, McGraw-Hill, 1957.
2. Bonilla, Charles, Nuclear Engineering, McGraw-Hill, 1957.
3. Glasstone, Samuel and Edlund, Milton C., Elements of Nuclear Reactor Theory, D. VanNostrand, 1952.
4. Holmes, D. K. and Meghreblian, I. V., "Notes on Reactor Analysis - Part II," August, 1955, CS-54-7-88, Technical Information Service.
5. Farbman, G. H., "Developments in Commercial Atomic Powerplants," Preprint 127, 1958 Nuclear Congress.
6. Flinn, S. W. and Petric, M., "Performance and Potential of Natural Circulation Boiling Reactors," Preprint 98, American Society of Mechanical Engineers.
7. Jealous, A. C. and Klotzbach, R. J., "Reprocessing Costs for Fuels from a Single Region Aqueous Homogeneous Reactor," AIChE Preprint 66, 1958 Nuclear Congress.
8. Perry, J. H., Editor, Perry's Chemical Engineering Handbook, McGraw-Hill, 1950.

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