Energy and Technology Review

Lawrence Livermore Laboratory

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Hydrogen

HYDROGEN PRODUCTION

THE COVER

A condenser packed with glass beads is being readied to separate liquid zinc chloride from the gaseous products formed in the zinc chloride hydrolysis reaction at 900 K. This reaction is one of a series required to produce hydrogen from water with our zinc selenide thermochemical cycle. The main reactions are illustrated in the schematic flow chart; the flow of selenium is shown in red, zinc in blue, sulfur in green, and water and hydrogen products in white. For further information on our work on hydrogen production, see the article beginning on p. 1.

ABOUT THE JOURNAL

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Hydrogen Production by Thermochemical Decomposition: The Zinc Selenide Cycle

The hydrogen on which fertilizer, molded plastics, and petroleum-refining industries depend comes now almost entirely from natural gas and light petroleum fractions. As these resources dwindle, other sources of hydrogen must be developed to support the ever-increasing demand. Water is a plentiful source. Hydrogen can be extracted from water by electrolysis or by a method that now promises to be more economical and energy-efficient: thermochemical decomposition. A number of thermochemical cycles under development use process heat directly to cleave the water molecule, release the hydrogen and oxygen, and regenerate the original reagents. Our experimental work has shown that the zinc selenide cycle offers many advantages in terms of favorable reaction rates and separation efficiencies as well as freedom from undesirable side reactions. These laboratory results imply that the zinc selenide thermochemical cycle may be an economical means of producing hydrogen.

Supplies of some of the most vital necessities—food and fuel, for example—depend on a potentially scarce chemical: elemental hydrogen. Chemical, bound hydrogen is plentiful, but elemental hydrogen is almost nonexistent in nature.

Chemicals synthesized from elemental hydrogen are varied and important: ammonia, an important constituent of fertilizers; methanol, a major in-

dustrial solvent; formaldehyde, a raw material for a broad array of molded plastics.

The annual volume of elemental hydrogen used in this country today is 110 cubic kilometres (10 Tg), and all of it must first be liberated from some compound. About 99% of it now comes from natural gas and light petroleum distillates, sources that will surely disappear within the next few decades. Moreover, the demand for hydrogen is increasing. For the past 30 years the average yearly increase, both in the U.S. and worldwide, has been about 13%. (Our hydrogen use is about one-third of the worldwide total.) If we postulate continued growth of conventional hydrogen uses at the minimum, 8% per year, demand will reach 5.4 times the current usage by the year 2000.

In addition, many new uses of hydrogen may be expected over the same period. It may be used to gasify or liquefy coal or refine oil shale, be blended with natural or manufactured gas, or be used to power vehicles, either directly or in the form of methanol. With these additional uses we may need 20 times as much hydrogen in the year 2000 as we do today.¹ Yet another possibility would expand the uses of hydrogen still further: conversion to a hydrogen economy in which hydrogen would replace almost all other fuels.

These expanded needs for hydrogen cannot be met by current methods. Since at present 6% of our natural gas goes for hydrogen production, generating 20 times as much would take more natural gas than we now produce. Clearly, it is es-

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essential to develop alternative sources of hydrogen before liquid and gaseous fossil fuels run out.

One possibility is coal gasification, an option that is being actively pursued at this and other laboratories. Since it is not certain that this process can produce all the hydrogen we will need, it is prudent to investigate the one virtually inexhaustible resource—water.

The traditional method of extracting hydrogen from water is by electrolysis, and this is still one possible alternative. But because of the inherent inefficiencies of electric power generation, the overall efficiency of electrolytic hydrogen production can never be more than about 40%—and is limited to about 30% at present.

Another theoretically possible method would be direct thermal decomposition. This is not now a practical alternative, for two reasons. First, temperatures above 2500 K are required to obtain significant decomposition (there are no present nonfossil heat sources in this temperature range, and materials problems would be severe). Second, the problem of preventing the resulting hydrogen and oxygen from recombining into water is considerable. If we interpose suitably chosen chemical steps between the cleavage of the water molecule and the release of free hydrogen, however, we can lower the reaction temperature and eliminate the recombination problem as well. This is the thermochemical cycle approach for hydrogen production.

We have investigated a number of such cycles and have focused most of our experimental research on the zinc selenide cycle. Our measurements strongly suggest that this cycle is quite feasible. We have also extended the laboratory discoveries to estimate its economics and have found it to be competitive.

THERMOCHEMICAL HYDROGEN PRODUCTION

There are a great many possible thermochemical cycles, most of which have never been explored. Each of them uses thermal energy from any convenient nonfossil source—a nuclear reactor or solar collector, for example—to break up the water molecules. Each of them is a closed loop,

regenerating its own reagents and passing them through the process over and over. Several of them have passed the scientific feasibility threshold and are in active development. The box on p. 3 lists those cycles confirmed by laboratory experiments and outlines their principal reactions.

The maximum reaction temperature in each of the cycles listed in the box is between 1100 and 1200 K. This temperature is not intrinsic to thermochemical cycles; it is the operating temperature of the hottest available nonfossil source, a high-temperature gas-cooled nuclear reactor.⁶ High-temperature solar collectors are not yet competitive with nuclear power for this application, although simpler solar energy systems may be useful in lower temperature portions of the cycle—for example, in boiling down dilute solutions to recover reagents.

Development of a hotter prime source of process heat would make additional and far simpler thermochemical cycles practical and might improve the efficiency of existing cycles. Fusion reactors may provide such a source sometime after the year 2000.⁷

A listing of three to five major reaction steps (as outlined in the box) gives only an indication of the complexity of a thermochemical system. Choosing the best cycle and optimizing it in terms of efficiency and economy are challenging research goals. The criteria that must be considered include:

- Favorable thermodynamics.
- Fast reactions.
- High reaction yields.
- Simple separations.
- Process design factors, such as efficient use of heat, avoidance of scarce or hazardous reagents, and simplicity of construction.

Favorable thermodynamics implies a negative and very small free-energy change for each reaction step. If the free-energy change were positive, the equilibrium constant of the reaction would be unfavorable, and much energy would be wasted in separating or concentrating the reaction products. If the free-energy change were too negative, on the other hand, one or more of the reaction products would be too stable and hard to regenerate into the original reagents.

HYDROGEN-PRODUCING THERMOCHEMICAL CYCLES WHOSE CHEMISTRY HAS BEEN VERIFIED IN THE LABORATORY

A thermochemical cycle is a series of chemical reactions that produces hydrogen gas from water and regenerates its reagents without loss. There are many theoretically possible thermochemical cycles. Those listed below have been proved scientifically feasible and are now under active development in various laboratories.

Zinc Selenide Cycle² (see Fig. 1 for the corresponding batteries)

Sulfur-Iodine Cycle³

Hybrid Sulfur Cycle⁴ (part electrochemical)

Iron Chloride Cycle⁵

Fast reactions are desirable, especially for reactions involving gaseous reagents, because they enable us to use smaller and less expensive reaction vessels. The requirement that the free-energy change be nearly zero makes it harder than usual to achieve fast reactions, however, because it allows very little driving force for the reactions. Reactions in which

the energy barriers are low enough permit a very small free-energy change to drive them rapidly.

High reaction yields are important to minimize recycling of unreacted reagents or diversion of material into undesirable by-products.

By using simple separations, we can avoid another area in which hidden costs can be substan-

tial. Operations such as boiling, compression, and filtering can involve expensive process equipment and energy losses, and they need to be minimized.

Finally, careful process design can do much to provide the optimum tradeoff between efficient heat-energy use and economy of construction and operation. Its contributions include new and innovative process equipment, plant layout, and siting.

THE ZINC SELENIDE CYCLE

Although the ideal thermochemical cycle almost certainly remains to be discovered, the zinc selenide cycle that we have been investigating is one of the more advanced. Through research funded by DOE's Division of Basic Energy Science, we have already demonstrated that each of its steps meets the criteria outlined above and that the efficiency of the entire cycle can be about 42%, better than even the most advanced electrolysis systems.

As may be seen in Fig. 1, the zinc selenide cycle consists of a number of interlocking chemical cycles, with water the only input material and hydrogen and oxygen the only products. The basic process consists of forming zinc selenide in Battery A (reaction 1), separating it in Battery B, and hydrolyzing it in Battery C (reaction 2). Decomposition in Battery E (reaction 5) of the hydrogen selenide formed in the hydrolysis step liberates hydrogen and regenerates selenium for making zinc selenide. An auxiliary cycle in Battery D (reaction 3) converts the zinc chloride (also formed in the hydrolysis step, Battery C) into zinc oxide for input to Battery A. Another auxiliary cycle in Battery F (reaction 4) decomposes the zinc sulfate formed in Battery A into zinc oxide, oxygen, and oxides of sulfur. A separator (Battery G) extracts the oxygen, returns sulfur dioxide to Battery A, and through additional iterations, which are not shown, decomposes the sulfur trioxide into more sulfur dioxide and oxygen. Other minor loops recycle hydrochloric acid (Batteries C and D) and excess selenium (Batteries A, B, and C).

Each of the steps in these many loops is a separate physical-chemical process, with its own set of reaction rates, equilibrium constants, and optimum con-

ditions. Each must be adjusted to fit into the integrated whole, optimized to yield only those products required and to produce them in the amounts needed to satisfy the succeeding steps in the cycle. Reaction vessels, pumps, pipes, and separators must be so sized that they are neither too big, and therefore expensive, nor too small, thereby creating a bottleneck.

We are pursuing basic research on the zinc selenide cycle to determine reaction rates, equilibrium yields at various temperatures, and reaction mechanisms. Our objectives are twofold: to define the controlling parameters of the zinc selenide cycle and to gain more basic information about thermochemical reactions in general. Specific information on reaction rates and yields will be invaluable to systems designers planning production-scale facilities. The reaction-mechanism information may be even more important; such information sheds light on whole classes of reactions, making it easier to select appropriate reactions and, perhaps, easier to devise additional thermochemical cycles.

Our laboratory studies have concentrated on the most crucial or least known reactions.

The ZnO/Se/SO₂ Reaction. This reaction, the most critical reaction in the zinc selenide cycle, was first discovered at LLL.⁸ Major points of concern are reaction rate, yield, and side reactions. The reaction is quite complex, involving solid, liquid, and gaseous phases:

We have conducted a series of sealed bulb tests ranging in time from 15 minutes to several hours at temperatures of 573 to 800 K and sulfur dioxide pressures of 100 to 800 kPa (1 to 8 atm). Reaction rates become rapid above about 700 K, but an undesirable side reaction that produces zinc sulfide occurs below about 723 K and sets this temperature as a lower limit. Between 723 and 800 K, ΔG_T° , the free energy change, for the reaction is calculated to range from -20 to 0 kJ/mole, becoming positive above 800 K (see Fig. 2). This then sets our working temperature range.

Our data to date indicate that the most important

Fig. 1. Simplified schematic flow diagram of the zinc selenide thermochemical cycle for extracting hydrogen from water. The reaction numbers refer to those listed in the box on p. 3. The various colors indicate the different internal cycles involving zinc, selenium, and sulfur. Broken lines indicate partial flows; solid lines trace paths that contain all of a particular component. Note that all the reagents except water are regenerated, and the only products are hydrogen and oxygen.

parameter for reaction rates within this temperature regime is sulfur dioxide pressure. Using the highest sulfur dioxide pressures attainable (800 kPa) in sealed silica glass bulbs we find the fastest rates to occur at about 723 K with reaction yields of about 66% in 20 minutes, tapering off to 95% in 2 hours and 99.5% in 4 hours. We have found also that the intimacy of mixing of selenium with zinc oxide is yet another important parameter. In future work we expect to develop an understanding of the mechanism

of this reaction and to define the optimum reaction conditions.

The ZnSe/HCl Reaction. We have found in small batch-type experiments that zinc selenide is rapidly hydrolyzed (greater than 95% in 5 minutes) by either dilute or concentrated aqueous hydrochloric acid.⁹ Care must be taken in an actual plant to minimize water content, since all water added for this step will need to be removed later by boiling. Care must also be taken to obtain a clean separation of the

Fig. 2. Change in free energy for the main reaction and the side reaction as a function of temperature. ΔG° is a measure of the tendency of the reaction to go to completion. Above zero, the reverse reaction is favored; the more negative, the more complete the reaction when equilibrium is reached. As our data show, reaction 1 does not proceed above 800 K; and below 723 K, a competing side reaction is possible. These results imply operation in the temperature band 723 to 800 K.

hydrogen selenide product from hydrochloric acid.

We also investigated alternative methods of hydrolyzing zinc selenide, but found them not as suitable as hydrolyzing with hydrochloric acid. As shown in Fig. 3, hydrolyzing with various concentrations of sulfuric acid is neither as fast nor as complete as with hydrochloric acid. Furthermore, this hydrolysis produces zinc sulfate rather than zinc chloride, and the sulfate requires more energy to convert it to zinc oxide.

The $\text{ZnCl}_2/\text{H}_2\text{O}$ Reaction. We have only recently begun to study this reaction.² In an earlier version of the zinc selenide cycle, we converted zinc chloride to zinc sulfate by reaction with sulfuric acid, and then decomposed the zinc sulfate at high temperatures to form the desired zinc oxide product.

We found, however, that significant gains could be made in cycle efficiency and economics by converting zinc chloride directly to zinc oxide using steam hydrolysis.

We have studied this reaction in the laboratory using a transpiration technique in which a carrier gas, nitrogen, is saturated with water vapor at 3.3 kPa (25 Torr) partial pressure and bubbled through molten zinc chloride at temperatures of 673 to 950 K. For a contact time of a few seconds, we find that equilibrium is readily attained above 800 K. Two other techniques have also been applied to the study of ZnCl_2 hydrolysis: one is illustrated in Fig. 4 and the other on the cover.

The ZnSO_4 Decomposition Reaction. A substantial literature exists on the zinc sulfate decomposition reaction,² and we have not investigated it in the laboratory thus far. The reaction is strongly endothermic and requires the highest temperature heat input for the cycle. Based on literature data, we expect zinc sulfate to decompose at 1160 K to produce 100 kPa (1 atm) of gaseous products containing a six-to-one molar ratio of sulfur dioxide/sulfur trioxide with the balance oxygen.

The decomposition of zinc sulfate occurs

Fig. 3. Completion of reaction 2, hydrolysis of ZnSe , as a function of time. Hydrolyzing with hydrochloric acid is faster and more complete than hydrolyzing with sulfuric acid.

Fig. 4. Basic components of an experiment to measure the rate of the zinc chloride steam-hydrolysis reaction. Absorption of ultraviolet light by zinc chloride vapor in the cylindrical optical-absorption cell enables the scanning spectrophotometer (in the background) to monitor the progress of the reaction and continuously record zinc chloride concentration as a function of time at one temperature. A fragile seal just above the absorption cell keeps the zinc chloride and water vapors separated until both parts of the tube have reached the desired operation temperatures (about 400 and 900 K for the upper and lower parts of the tube, respectively).

stepwise. Initially it is decomposed to an oxysulfate, probably $\text{ZnO} \cdot 2 \text{ZnSO}_4$, which then further decomposes to zinc oxide. From the literature we would expect decomposition to occur in about 15 minutes at 1160 K; however, heat and mass transfer effects appear to limit the decomposition rate of zinc sulfate in the work cited. This is slower than it should be because the zinc sulfate has been heated as a stagnant bed of material in the presence of a slowly flowing carrier gas stream over the surface of the

bed. We would expect to decrease the decomposition time to about 1 minute by using a dispersed flow of zinc sulfate particles and radiation heat transfer directly to each particle.

The H_2Se Decomposition Reaction. At room temperature hydrogen selenide is a metastable gas; thermodynamically it should decompose to selenium and hydrogen, but the decomposition kinetics are negligibly slow. We have studied the mechanism and kinetics of hydrogen selenide decomposition in

the temperature range between 673 and 748 K (see Fig. 5) and find the decomposition rate to be 60% in 5 minutes at 748 K; it is even more rapid at higher temperatures.¹⁰ Maximum yields at these higher temperatures are about 65%. Recycling, therefore, will be necessary for complete decomposition. If a suitable catalyst could be found so that we could operate at a lower temperature, we could attain higher yields because of hydrogen selenide's lowered stability.

ECONOMIC ANALYSIS

We have performed a preliminary economic analysis of the zinc selenide thermochemical cycle, assuming a production plant that uses process heat at 1200 K from a very-high-temperature nuclear reactor and turns out 27.3 Mg of hydrogen per hour. In 7000 hours (a year's operation with a capacity factor of 80%) such a plant would produce

Fig. 5. Temperature dependence of hydrogen selenide decomposition rate. The decomposition is 60% complete in 5 minutes at 748 K. Equilibrium yield at 748 K is about 65%, as determined by previous investigators.

Table 1. Breakdown of capital investment for a plant producing 27.3 Mg/hr ($2 \text{ km}^3/\text{yr}$) of hydrogen from water by the zinc selenide thermochemical cycle. The cost of building the reactor, \$520 million, is not shown in this table.

Item		Cost ^a (millions of 1976 dollars)
Battery		
A	ZnSe/ZnSO ₄ generation	20
B	ZnSO ₄ separation	152
C	ZnSe hydrolysis	141
D	ZnCl ₂ steam hydrolysis	215
E	H ₂ Se decomposition	24
F	ZnSO ₄ decomposition	218
G	SO ₂ /SO ₃ /O ₂ separation	63
Off-site, general		7
Off-site, direct		15
Subtotal		855
Contingencies and indirect costs ^b		373
Total capital investment		1228

^aIncluding 105% for structures, installation, etc.

^bTaken at about 44% of capital investment.

2 km^3 of hydrogen, or about 2% of our current needs. The results are presented in Table 1, based on tentative process design concepts with costs reported in mid-1976 dollars. The invested capital for the plant would total about \$1.2 billion. In addition, the plant would require a dedicated reactor costing about \$520 million.¹¹

The annual operating cost of such a plant, exclusive of nuclear heat cost, would be about 23% of the \$1.2-billion initial capital investment, or \$280 million. This 23%, mainly amortization costs, assumes major equipment lifetimes of 10 to 20 years. This part of the cost of hydrogen would be $\$0.124/\text{m}^3$, or $\$9.86/\text{GJ}$ (based on a heating value of hydrogen of $12.9 \text{ MJ}/\text{m}^3$). The annual operating cost of the reactor comes to about \$105 million (figured at about 20% of initial capital investment of the reactor), bringing the cost of process heat at the reactor—assuming 80% operation at its rated capacity of 3426 MW(t)—to about $\$1.24/\text{GJ}$.

To estimate how much the process heat cost adds

to the cost of the hydrogen produced by such a plant, we need to know how much heat is required per mole of hydrogen produced. Since our cycle is not yet optimized, the energy needs summarized in Table 2 are necessarily tentative. They are solidly conservative, however; an optimized design would be more efficient.

In constructing Table 2 we have assumed an efficiency of 34% for generating the auxiliary power needed for pumping and other process machinery. We see that the overall heat required to produce 1 mole of hydrogen at 3 MPa by this process is 679 kJ. Since the theoretical decomposition energy of water into hydrogen and oxygen gases is 286 kJ/mole, we obtain an overall cycle efficiency of 286/679, or 42%.

At a thermal efficiency of 42%, a plant producing 27.3 Mg/hr of hydrogen would need only 2430 MW of process heat. If we assume that this heat is available at the same rate (\$1.24/GJ) derived for the 3426-MW reactor, this translates into about \$2.94/GJ of produced hydrogen. The overall cost, plant plus nuclear heat, is therefore \$12.80/GJ energy equivalent (hydrogen heating value).

This is a very promising initial cost estimate for

an unoptimized process in the early stages of its development. Published cost figures for other thermochemical cycles range from \$4.70 to \$11.70/GJ.¹² We believe most of these estimates are optimistic, whereas ours is conservative. We expect that hydrogen can be produced for about \$10/GJ by the best thermochemical cycles when they are fully developed, and that this process will be competitive with other production methods by the year 2000.

Key Words: ammonia fertilizer; coal gasification; coal—hydrogenation; coal—liquefaction; hydrogen—applications; hydrogen economy; hydrogen production; natural gas; power resources—natural gas; thermochemical cycle.

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Table 2. Tentative hydrogen production process energy balance (kJ/mole of hydrogen), based on unoptimized zinc selenide thermochemical cycle.

Reaction No.	Battery	Process step	Auxiliary power ^a (heat equivalent)	Heat input	Heat output
(1)	A	ZnSe/ZnSO ₄ generation	1	9	218
	B	ZnSe/ZnSO ₄ separation		110	76
(2)	C	ZnSe hydrolysis		207	
(3)	D	ZnCl ₂ hydrolysis		234	
(5)	E	H ₂ Se decomposition	30		39
(4)	F	ZnSO ₄ decomposition	70	325	
	G	SO ₂ /SO ₃ /O ₂ separation	129	10	117
		Total	230	899	450

Net nuclear heat requirement = 230 + 899 - 450 = 679 kJ/mole H₂.

Thermal efficiency (η) = 286/679 = 0.421, or 42%.

^aBased on 34% electric power generating efficiency.

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Safeguards Research: Assessing Material Control and Accounting Systems

The Laboratory is working for the Nuclear Regulatory Commission to improve the safeguarding of special nuclear material at nuclear fuel processing facilities, to provide a basis for improved regulations for material control and accounting systems, and to develop an assessment procedure for verifying compliance with these regulations. Early work included setting up a hierarchy of safeguard objectives and a set of measurable parameters with which systems performance to meet those objectives can be measured. Present work has focused on developing a computerized assessment procedure. We have also completed a test bed (based on a plutonium nitrate storage area) to identify and correct problems in the procedure and to show how this procedure can be used to evaluate the performance of an applicant's material control and accounting system.

The growth and acceptance of nuclear power have been limited by society's fears of the risks accompanying the use of such power. The main areas of concern have been reactor safety, management of nuclear wastes, and safeguards. LLL has research projects under way in all three areas for the Nuclear Regulatory Commission (NRC).¹ This article focuses on our work to develop ways of assessing safeguards for special nuclear material (SNM) at nuclear fuel processing facilities.

Safeguards are intended to prevent the malevolent use of nuclear materials or the sabotage of nuclear facilities. Traditionally, SNM safeguarding has had two components: physical security, and material control and accounting. The NRC has given LLL the responsibility of developing a methodology to assess material control and accounting systems and Sandia Laboratories the responsibility of evaluating physical security at fixed sites and transportation systems.

In the area of nuclear material control, the NRC is interested in reviewing and updating its safeguard regulations, and it is studying the possibility of making these regulations performance-oriented rather than very detailed and specific, as is the case today. Instead of requiring, for example, that the analytical chemistry laboratory maintain adequate reference standards, calibrate the measurements system, and control measurements performance, the NRC might simply require that a material control system be designed to detect the theft of a specified amount of SNM. However, assessing performance-oriented regulations is more difficult since it requires a methodology by which the performance of a material control system can be measured. Quantifying system performance would also contribute to the development of specific regulations.

We are therefore working on two closely related tasks: (1) providing a logical basis for the creation of improved regulations and (2) developing a means to test compliance with these regulations. The first task required us to develop a hierarchy of safeguard

¹Contact Arturo Maimoni (422-8575) for further information on this article.

objectives and a set of measurable parameters for these objectives, which are briefly described below. Most of our latest efforts have been directed at developing a usable assessment procedure and associated computer codes to test compliance.

SAFEGUARDS HIERARCHY

We have developed a seven-tier hierarchy to show calculable relationships between overall safeguard objectives and measurable functions of a material control system. This breaks down general objectives—such as protection of the public against the consequences of a theft of SNM—into successive levels of more specific objectives until the latter are reduced to physically measurable quantities that can be assigned performance measures, or probabilities.

To illustrate this hierarchy (see Fig. 1), consider the relationship between the objective of protecting against theft of SNM (level II) and the functions of a material control system. The general objective can be broken down into the more specific objectives of reducing the adversary's probability of success given an attempted theft and reducing the adversary's incentive to make that attempt. In turn, his probability of success can be broken down into limiting his choice of actions and guarding against those actions. One subobjective of the latter is then to protect against unauthorized acquisition, and one means of doing so is to detect abnormalities in the SNM processing stream. This detection, in turn, could involve monitoring SNM quantities and records. With this last step, we can calculate probabilities, conditional on the attributes we give the adversary, for the objectives based upon the performance—or failure—of specific components in a material control system.

Performance measures provide a value that indicates how well an objective is met. Values at one level of the hierarchy can then be aggregated to the level above as sums or products of probabilities or through more complex mathematical functions. Ultimately, aggregation could continue until a value can be given to how well the material control system meets the basic safeguard objective of protecting the public against the consequences of an SNM theft.

This will be a very complicated process, however, because of the number of variables—for example, characteristics of the different types of adversary—that must be accounted for.

ASSESSMENT PROCEDURE

Our main effort this past year has been to develop the methodology and computer software needed by the NRC to assess material control and accounting systems. In developing a usable assessment procedure, we are interested in finding ways to measure the capability of proposed or existing systems to detect a theft attempt. These ways should be flexible and, for proposed material control systems, should provide a consistent means of assessing the applicant's compliance with regulations at all stages of development, from preliminary design and approvals through construction to initial and ongoing plant operations.

From a detailed description of a fuel processing facility, the assessment procedures being developed at LLL systematically generate a set of possible actions by an adversary that could lead to the diversion of SNM and calculate the probabilities of interrupting those actions. To develop this procedure, we have drawn upon modeling techniques from a number of disciplines, since we need to describe such varied factors as the adversary's characteristics, the effect of process upsets on SNM inventory, the material control and accounting system, and societal perceptions of risk from nuclear incidents.

The assessment procedure will draw upon two types of data: applicant information and the NRC/LLL data base. Applicant data include the plan of the facility's physical plant, operational procedures, descriptions of SNM processing, and details of the material control and accounting system. The NRC/LLL data base will contain the mathematical models necessary to evaluate an applicant's submittal, for example, models of the performance of the analytical chemistry measurements.

There are five steps to our procedure:

- Identify targets from which attractive nuclear materials could be diverted.
- Determine what adversary actions and condi-

Fig. 1. Single line from our hierarchy of safeguard objectives at nuclear fuel processing facilities (corresponding to the blue line in the overall hierarchy schematic shown at the bottom of the figure). We have broken down the problem of protecting the public against the consequences of an SNM theft into seven levels of safeguard objectives. Our goal was to ultimately reach a level for which we could calculate probabilities for these objectives based upon the performance of specific components in a material control and accounting system. The schematic of our complete hierarchy indicates the extent of this breakdown.

tions of the material control system could allow a successful diversion, that is, generate event sets.

- Time-order the adversary's actions and their resulting stimuli (physical, chemical, or informational changes being monitored by the material control system).

- Determine the response of the material control system to those stimuli and estimate the probability of outcomes for attempted thefts based on such responses.

- Score the overall effectiveness of the material control and accounting system.

Target Identification. In our context, a target is a given type of SNM, the location in the plant where this material may be obtained, and the conditions that will cause the SNM to appear at this location. We first identify all locations of SNM in the plant and rank them according to the attractiveness of that material to the adversary. Attractiveness is simply a measure of the effort required to prepare the SNM for malevolent use: plutonium-239 would thus be more attractive than uranium ore. Our algorithm for this ranking considers the material's chemical species, plutonium concentration, radiation hazard, and physical state.

After ranking the material, we must identify all normal and abnormal material access points. Abnormal access points are those into which SNM does not normally flow but into which it could be made to flow by applying pressure or a vacuum or by changing the plant's configuration. We have developed a computer program to analyze a plant's piping and instrumentation diagram to identify these potential diversion flow paths.² At this early point in our procedure, therefore, a plant's vulnerability to diversion can be preliminarily assessed and, in the case of a proposed plant, probably lessened by minor design changes.

Event-Set Generation. An event set is defined as a set of adversary actions and corresponding conditions in the material control system that could lead to the successful diversion of SNM from a given target. To generate these sets, we begin with directed graphs—referred to as digraphs—that depict the interrelationships of processing-stream variables and external events.³ We model the adversary's

movements, the SNM flow, and the signals and actions resulting from various responses of the material control system, including its decision logic. Digraphs can also show the effects of normal equipment failures and of countermeasures taken by the adversary.

A simplified digraph for a pressure-operated switch in a floor mat, typical of what might be present in a proposed material control system design, is shown in Fig. 2. Each box is referred to as a node; the arrows connecting the nodes are referred to as

Fig. 2. Digraph (directed graph) of a floor mat pressure switch. The initial action of the adversary crossing the floor mat results in pressure being applied on the mat; this flow of information is denoted by the plus sign for the edge (arrow) between the first two nodes (boxes). The applied pressure will register on the mat's pressure sensor (plus) unless the adversary first unplugs the mat or the plant has a power failure (zero). The pressure signal in turn will be transmitted to the decision logic of the material control system unless, again, there is a power failure or the adversary cuts the conduit to the logic. Once the logic receives the signal, it would have to decide whether the signal was normal (someone was supposed to be there at that time) or abnormal and thus whether or not to start countermeasures.

edges. A plus sign on an edge denotes the flow of information, a zero the absence of that flow. For each node we are looking for those conditions or acts that would allow the adversary to interrupt the information flow. We note from this digraph that the adversary's presence could go undetected if he unplugs the pressure mat or if there were a power failure. Furthermore, his presence could be detected but not responded to if he had cut the conduit from the pressure sensor to the control system's decision logic. Thus, for further analysis, we would need data on the probabilities of detection and of false alarms and on the system's susceptibility to tampering.

For a more realistic but still simplified example of event-set generation, consider Figs. 3 and 4. The tank in Fig. 3 contains a plutonium nitrate solution whose level (L) is monitored by an air bubbler system that measures the pressure difference be-

tween the tank's bottom and top. An operator periodically samples the solution and sends the sample to a laboratory for analysis. The pressure in the sample-handling glove box is also monitored, and an observer oversees the entire area.

To construct the digraph in Fig. 4 for this setup, we first define a diversion scenario: material removal at the glove box leading to an SNM diversion at the facility (blue boxes). The digraph traces some of the adversary's actions and responses of the material control system that would allow or prevent such a diversion. For example, the adversary may cut the gloves to remove the plutonium solution, affecting the pressure in the glove box. He may try to interfere with the air bubbler system by substituting another liquid for the plutonium nitrate being removed or by pulling a partial vacuum on the differential pressure indicator. The observer may react to these actions and immediately ap-

Fig. 3. Hypothetical plutonium nitrate storage area at a fuel processing facility. An air bubbler system monitors the level (L) of the plutonium nitrate in the tank by measuring the pressure difference between the tank's bottom (P_1) and top (P_2). The solution is sampled periodically at a glove box with the samples being sent to a laboratory for analysis. An observer monitors the glove box area. Figure 4 shows a digraph representing this arrangement.

prehend the adversary at the glove box (minus sign) or react by sending an alarm to the material control system logic (plus sign), which then notifies physical security. A prompt response from security could also prevent any diversion from the facility. A variety of conditions (zeros) could lead to the adver-

sary's success.

Once a set of digraphs for the material control system has been developed—one for each material access point—the digraphs are converted into fault trees, which are then examined to obtain the event sets. A partial listing of event sets for our example is

Fig. 4. Digraph for the plutonium nitrate storage area shown in Fig. 3. Some adversary actions—and corresponding responses from the material control system—are traced. Plus signs on the edges (arrows) connecting the nodes (boxes) indicate a flow of information. Zeros indicate the lack of such a flow. Minus signs indicate a response to the information flow.

given in Fig. 5. A sequence of adversary actions might be to fill a container at the sampler, pull the container back through the glove, clamp the glove, cut the glove to remove the container, and exit with the plutonium nitrate solution. The conditions that would allow such a sequence to go undetected are failure of the tank's level sensor and no observer detection. Since these two conditions are independent of one another and do not share a common cause (such as a power failure), the diversion probability for this event set comes simply from the probabilities of failure of the sensor and observer. We anticipate that the outcomes of most event sets will derive from the probabilities of such independent conditions. More complex event sets will be further analyzed in the next step.

Sequence/Stimuli Generation. A sequence is a time-ordered series of actions needed to remove SNM and to conceal its removal. Stimuli are signals feeding information to the material control system. In this step, we characterize the duration of the adversary's acts in an event set, the time separation between them, and the stimuli they generate. NRC

criteria might eliminate certain event sets from such analysis on the basis of the difficulty of the diversion attempt or the inconsequential risk involved. Event sets whose outcomes still cannot be determined will be analyzed in the next step.

Response and Outcome Determination. The computer code developed for this step is the Material Control System Simulator (MCSS). With this code we will model the pertinent portion of the plant's SNM processing stream, the characteristics of all applicable components, and the decision logic of the material control system. We will then superimpose the adversary actions and control-system conditions of each sequence determined in the preceding step and compute the response and outcome probabilities. A Monte Carlo simulation can be used whenever required. MCSS will explore how the outcomes of attempted diversions are influenced by various uncertainties, such as the effects of precise timing of the process, adversary acts, and random stimuli levels.

The results from this analysis, together with those from the two preceding steps, will define the

Fig. 5. Examples of some of the event sets that could be generated from the Fig. 4 digraph. (The event set discussed in the text is shown in the righthand column.) Each vertical column consists of a series of adversary acts and the corresponding material control system conditions necessary for those acts to be successful. Once the important conditions for an SNM theft are so determined, we must then calculate their probabilities.

response of the material control and accounting system to SNM diversion attempts. This information must then be combined with data about the performance of the physical security system to determine the outcome for a given diversion attempt. We are beginning to work out the details of this interface between our material control models and Sandia's security models.

When assessing the SNM safeguards at an existing or proposed fuel processing facility, the NRC will probably conclude its evaluation by aggregating the results at this point. With the results of the assessment, the NRC would make a decision regarding the effectiveness of the material control and accounting system of the applicant. However, when formulating new regulations—especially performance-oriented ones—it would continue the aggregation process.

System Effectiveness. The final step of our assessment procedure is to score the overall effectiveness of the material control and accounting system. From the previous steps, we will have the probabilities of outcomes of individual sequences. These will then be aggregated, based on NRC criteria, into an overall measure (or measures) of system performance. The theoretical structure for performing the aggregation is now being developed. An example of the criteria that the NRC might use to formulate improved regulations and system-acceptance criteria is the tradeoffs between the operating costs of a safeguards system and the degree of acceptable theft risk posed by different adversary types. Ultimately, society's perception of the risk from a safeguards incident must be incorporated into the performance levels required by NRC regulations.

Basic to our assessment procedure is information about the performance of material-control system components and about adversary perceptions. To be useful in our assessment, the complex characteristics of the components of the material control system that might be proposed by an applicant have to be expressed as simple functional representations (models). Typical components requiring modeling include monitoring equipment such as pressure sensors, components that deter adversaries such as

physical barriers or locks, and humans as observers or operators. The modeling required to characterize the performance of proposed online material diversion detectors is complex and requires knowledge of the dynamics of the chemical processes and measurements system. We need to know how well these components perform in the context of their environments, the probabilities of detection and false alarm, and pertinent time-response information. Means for modeling the decision logic supplied by the applicant (including reaction components) are also needed. The framework for these models is being developed now.

Models of the adversary are essential to many aspects of our analysis. We need to know, for example, how many adversaries may be involved in a theft attempt, what their skills are, what information is available to them, and what risks they may be willing to take. We are developing such adversary models in close cooperation with the NRC.

TEST BED

To test our assessment procedure, we have written the design specifications for the material control and accounting system of a plutonium nitrate storage area. This test bed is based to a great extent on the storage area at Allied-Gulf Nuclear Services, Barnwell, South Carolina, but with considerable departures. It includes a processing stream within a material access area, detailed operational procedures, structured material control decision logic, and well-characterized monitoring components.

The test bed includes examples of most of the protection mechanisms an applicant might use to meet future performance-based regulations. It will allow us to identify and correct problems in our assessment procedure and will serve as an example of how one can use this procedure to evaluate applicant submittals. The assessment will focus on timely detection of diversion. We are now developing detailed system digraphs for the test bed and plan to complete the assessment by the end of January 1978. The scope of this work—and of assessing fuel processing facilities in general—is evident from the fact that we have identified 131 access points to

SNM in the test bed's storage area. For just one of these access points, we have generated 2180 event sets that involve 20 to 30 adversary acts and material control system conditions. The vast number of possibilities and the complexity of each scenario underscore the utility of our computerizing the assessment procedure, which provides quantitative information about the importance of the basic events and allows analysis to be performed for collusion requirements, common mode failures, etc.

SUMMARY

The assessment procedure and codes we are developing for the NKC will allow the Commission to systematically evaluate the adequacy of safeguards at nuclear fuel processing facilities. Although each step in the procedure is designed to provide input to the next step, individual steps can also be used as standalone tools to examine particular cases of plant vulnerability. We believe that our methodology will be useful and flexible. With it the NRC will be able to efficiently assess a proposed facility during its design stages, study the effect of proposed modifications to existing plants, and develop improved regulations.

Key Words: material control system; NRC; nuclear materials management; safeguarding nuclear materials.

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Solar Power for Industrial Process Heat

American industry annually burns the equivalent of 30 million barrels of oil just to heat water. Generating steam takes roughly another 650 million barrels. Solar energy could supply much of this hot water and steam, saving enormous quantities of nonrenewable fossil fuel, if solar collectors were less expensive to build.

In our solar power development, we have been concentrating on ways to reduce the capital cost. The cost of expanding our prototype shallow solar ponds—now at the end of their second year of operation near Grants, New Mexico—to a full-scale operating system is within about 25% of the breakeven price based on current oil prices. The experimental solar steam generator we are about to begin testing should cost from one-tenth to one-fifth as much as other currently funded DOE solar steam systems. We can achieve these economies by constructing the system from inexpensive materials and by eliminating the need to track the sun.

An enormous amount of solar energy—about 600 times the country's total annual energy consumption—falls on the U.S. each year. The amount of this vast resource that is being directly used is negligible. Existing schemes for collecting and converting solar energy are too expensive to compete with conventional fuels.

Contact William C. Dickinson (422-1465) for further information on this article.

Fig. 1. One of three shallow solar ponds built under our supervision near Grants, New Mexico, and operated there over the past two years by the Sohio Petroleum Company. We have designed a demonstration facility, based on operating experience with these modules, that would supply about 25% of the hot water needed by Sohio's uranium ore processing plant, saving about 950 m^3 (6000 bbl) of oil every year. Later expansion could double this savings. If the economics appear favorable, construction of this facility could start this spring.

To be competitive, a solar energy system must cost no more than the fossil fuel it will save over its operating lifetime; hence its allowable cost depends on the price of fuel. With oil selling for \$14 per barrel, the installed cost of a solar collector must be below about \$40 per square metre. The major goal of our research has therefore been to keep costs down.

Another criterion that directed our research is scale. Residential solar units are small. Important though it may become eventually, residential solar power requires the modification of millions of existing dwellings before it can make any significant impact on the nation's energy use. However, because industrial process heat applications tend to be very large, they can begin to effect substantial savings with relatively few installations.

Industrial heating processes account for about 25% of the nation's fuel bill. ¹ About 2% of this (5%

of the total national energy budget) is for processes that take place below 175°C , mostly for heating water and generating steam. ² Our research has concentrated on developing cost-competitive solar collection systems that operate in this relatively low range.

We are developing two solar energy collection systems, a shallow solar pond concept (Fig. 1) that produces hot water in the 30 to 60°C range, and a concentrator system to produce steam up to temperatures of 175°C (Fig. 2). Three of our prototype shallow solar ponds have been operating at the Sohio Petroleum Company's uranium ore processing plant for the past two years. We are also constructing a pair of experimental solar concentrators and will soon begin preliminary testing.

SHALLOW SOLAR PONDS

In exploring ways to use solar energy to heat

Fig. 2. Artist's conception of an inflated cylindrical concentrator for producing industrial process heat. Our working model is 1 m in diameter with a central working section 4 m long. The upper half of the inflated outer cylinder is clear; the lower half is aluminized to form a concentrating reflector. The receiver tube is the vessel in which the water is heated to approximately 170°C. The thin plastic sheath around the pipe aids in reducing convective and radiative heat losses from the pipe.

water, we were guided by two criteria: economy and ease of manufacture in the large sizes consistent with industrial applications (typically 10 000 m² or larger). We also recognized the need to seek cost-effective materials—those that strike an economical balance between long life and low initial cost. These considerations led us to develop an 80-year-old design, the shallow solar pond, with modern plastics and construction techniques.

Our shallow solar pond consists of a long plastic bag (4 by 60 m) filled with 5 to 10 cm of water. The bag lies on an insulating bed and is glazed over with a low fiberglass roof similar to those on greenhouses. Figure 3 illustrates many of these construction features. The plastic bag prevents heat loss

through evaporation, the glazing cuts down on convective and radiant heat losses, and the insulation reduces heat conduction to the ground.

In operation, the bags are filled every morning at sunrise, absorb heat through the day, and are drained into an insulated tank when the water temperature reaches its peak in the late afternoon. The peak temperature may reach 60°C or more in summer and 30°C in winter, depending on the initial temperature of the water, the outdoor temperature, and the cloud cover. If the stored water is not hot enough for its intended use, the rest of the heat can be supplied by a conventional oil-fired boiler. The number of pond modules depends on the volume of hot water required; each module will heat about

Fig. 3. Construction details of a shallow solar pond module. The PVC water bag is 4 m wide and 60 m long and is filled to an average depth of 10 cm. Thus it holds 24 Mg (24 tonnes) of water. The plastic bag prevents heat loss through evaporation, the fiberglass top glazing reduces convective and radiant heat losses, and the foamed glass bottom insulation reduces heat conduction to the ground.

24 Mg (24 tonnes) of water each day.

In 1975, under our supervision, the Sohio Petroleum Company built three prototype shallow solar pond modules to our specifications at their uranium mill (Fig. 1). Tests with these modules over the past two years, plus three years of experiments at LLL enable us to predict shallow solar pond performance accurately and provide a fairly good understanding of the effects of varying design and materials. For example, clear acrylic plastic might be considered an attractive choice for glazing, since it is more resistant to ultraviolet light than is fiberglass. However, it tends to crack badly at the corners due to the high stresses there and actually has a shorter life.

Based on this experience, we have designed an 8000-m² demonstration facility to supply hot water at the rate of 800 m³ per day to the Sohio mill. This will reduce the mill's fuel consumption by about 25% or 950 m³ (6000 bbl) of oil per year. If it operates as expected, its capacity can readily be doubled, by adding more ponds, to provide almost all the plant's energy requirement in the summer and a good fraction in the winter. This represents the most economically attractive system for Sohio,

since a larger system would be partly idle during the summer.

We estimate that this system will cost \$50 per square metre. With fuel oil at \$88 per cubic metre (\$14/bbl), the most that Sohio can afford to invest while still maintaining their normal 15% rate of return is \$40 per square metre. Sohio is therefore postponing construction until either the rising cost of oil or federal solar-energy-incentive legislation makes up the difference.

Several solar-incentive proposals are now before Congress, and if favorable legislation is enacted, construction on the Sohio project could begin this spring. If it proceeds on that schedule, it will be the first large-scale solar industrial-process-heat system to be constructed in this country without a direct government subsidy.

A second shallow solar pond facility has been proposed by the Army for Ft. Benning, Georgia, to supply hot water to three barracks complexes and the post laundry. We are designing 124 ponds covering 35 000 m², to be built on two sites in 1979 or 1980. This particular solar pond system is expected to save the Army over 3200 m³ (20 000 bbl) of fuel oil per year.

Teledyne-Brown Engineering of Huntsville, Alabama is also designing a DOE demonstration shallow solar pond facility, with LLL serving as technical advisor. This system will comprise 1600 m² of collectors to supply hot water for a canning plant at Athens, Alabama. Construction is

scheduled to begin this spring, with the system becoming operational this fall.

SOLAR STEAM GENERATION

Although the demand for hot water is significant, the demand for steam in the temperature range be-

Fig. 4. A computer-generated plot of sun-ray paths inside the collector. Some rays strike the tube directly; many more reach it after reflecting from the aluminized lower surface of the inflated cylinder; those at the outer edges miss the receiver tube completely. In this example the outer cylinder is 14 times as large as the receiver tube, and the sun's rays are entering at an angle, α , of 3° to the normal axis (the line through the centers of the outer cylinder and the receiver tube). Because of the noncritical focusing properties of the cylindrical reflector, small changes in α make very little difference in the collected heat. The light circle around the receiver tube represents a thin plastic jacket placed there to minimize heat losses from the receiver.

tween 100 and 175°C is more than 20 times larger. Again, the main obstacle to filling this need with solar energy is the cost of building solar collecting equipment and the associated system components.

To operate in this higher temperature range, a solar collector must gather sunlight over a large area and concentrate it onto a smaller area, such as a pipe containing pressurized water. The typical embodiment of this idea is a highly reflective parabolic concentrator mounted on gimbals and motorized to track the sun. Such systems presently cost \$500 or more per square metre of collector area when installed, a price that makes them impractical for industrial applications.

We are developing a concentrating solar collector whose reflector is a large inflated cylinder of thin-film plastic with a clear upper half and an aluminized lower half. This collector surrounds the receiver tube, a large pipe running horizontally on an east-west line, and can be tilted manually in the north-south plane to intercept as much sunlight as possible. Figure 2 shows an artist's conception of such a collector.

We estimate that this collector and its associated components can be built for a total installed system cost of \$50 to \$70 per square metre of collector area. We expect to achieve this low cost by using inexpensive, weatherable plastics and by eliminating the necessity for tracking the sun. Eliminating tracking also greatly improves the system's reliability.

The ability of the system to function without solar tracking derives directly from the circular cross section of the reflector. Figure 4 is a computer-generated plot tracing the paths of sun rays entering the collector at an angle, α , of 3° off the normal axis (a line joining the centers of the reflector and the collector). In this case the ratio of reflector diameter to receiver tube diameter is 14.

Our optical and heat-transfer codes enable us to show that small variations in α and in the position of the receiver tube have very little effect on the system's performance. Manually adjusting the collector tilt once a week is enough to keep the system gathering over 90% of the energy obtainable by active tracking.

Our computer studies have provided considerable

insight into the effects of geometry, optical and thermal properties, and ambient conditions on collector performance. We can show, for example, that, at an operating temperature of 170°C, winds up to 9 m/s contribute much less than 1% to the heat loss. Basically, this is because there is very little driving force to encourage heat flow; the outer envelope stays within 1°C of ambient temperature at all times.

Another factor that has only a slight effect on the total heat loss is the infrared transmissivity of the plastic jacket around the receiver tube. This jacket is a thin layer of plastic spaced about 10 mm from the surface of the receiver to minimize convective heat transfer from the receiver. The natural convective heat loss is so much larger than the radiative heat loss, provided that the receiver tube has a selective face, that even fairly large changes in the latter have relatively little effect on the total.

The surface properties of the receiver tube, however, present an entirely different situation. We have discovered that, for operation at 170°C, this surface must be a selective radiator: it must be a good absorber of visible and near infrared radiation but a poor radiator of far infrared radiation. This effect is achieved with special plating processes that first give the tube a shiny nickel finish and then cover it with a 2- to 3- μ m layer of black chrome.

In Fig. 5 we plot how the thermal efficiency (defined as the ratio of the energy delivered into the water to the optical energy absorbed by the receiver tube) depends on the receiver tube temperature for surface emittances of 0.1, 0.2, 0.3, and 0.9. For the best selective surface (emittance = 0.1) the thermal efficiency at 170°C is 62%, more than five times better than the 11% efficiency figure for the nonselective surface at the same temperature. Even at the relatively low temperature of 100°C, a tube with a selective surface would deliver 50% more energy to the fluid than an uncoated one.

The stagnation temperature is the maximum temperature the receiver will reach with no fluid flowing. The stagnation temperature depends primarily on two factors: the diameter of the receiver tube and the amount of optical power absorbed by the tube. For a fixed optical power input, expressed in watts

Fig. 5. The thermal efficiency (the percentage of the heat absorbed by the tube that is transmitted to the working fluid) varies with temperature for tubes having different emittances. For efficient heat collection, the receiver must have a selective surface—one that is a good absorber of visible and near infrared radiation but that has a very low emittance for the far infrared. At 170°C the thermal efficiency of the best selective surface (emittance = 0.1) is more than five times better than that of the non-selective surface (emittance = 0.9).

per metre of length, the smaller the receiver tube the higher the stagnation temperature. Figure 6 displays these relationships. The separately plotted point indicates the stagnation temperature (340°C) for a 40-mm receiver (the smallest practical size) and 340 W/m of optical power (the maximum available with our collector in the Southwest at the

Fig. 6. The stagnation temperature (the maximum temperature the receiver tube would reach with no fluid flowing) varies with optical input power and with receiver tube diameter. The higher the input power, the higher the temperature, but the increase is not linear.

summer solstice). Under these conditions the temperature of the plastic jacket will rise to 130°C.

The overall collector efficiency is the product of the thermal efficiency (defined previously) and the optical efficiency. The optical efficiency is the percentage of the total solar energy entering the collector that is absorbed by the receiver tube. Both quantities are integrated over the effective solar day (the 6-hr interval from 9 a.m. to 3 p.m.), and the insolation is summed over the collector's entire length and width. The optical efficiency for our baseline configuration is about 37% regardless of receiver temperature. The thermal efficiency drops gradually from 88% at 40°C to 55% at 170°C. Hence the overall efficiency at 170°C is about 20%.

Variations in receiver tube diameter also affect the efficiency of the collector. Figure 7 plots the thermal, optical, and overall efficiencies for our baseline configuration at 170°C as a function of receiver tube diameter. As the tube diameter increases, the thermal efficiency decreases and the optical efficiency increases. Hence the product, the overall collector efficiency, has a broad, flat maximum between receiver diameters of 50 and 90 mm.

Analogous plots for other operating temperatures all have similar broad, flat maxima, displaced toward larger diameters for lower operating temperatures. This relative insensitivity of the collector efficiency to tube diameter suggests that our choice of tube size for a particular system will be influenced by other considerations, such as stagnation temperature, thermal inertia, adjustment ease, and collector cost.

EXPERIMENTAL SOLAR STEAM GENERATORS

We are currently building two identical experimental collectors so that we can compare various parameters under the same operating conditions. The dual system also allows us to test a greater number of materials and geometries in a given time. Since these units are experimental, they only approximate the features of the envisioned full-scale system. The outer cylinder, for example, has rigid end pieces mounted in a solid frame to permit

Fig. 7. Optical, thermal, and overall collector efficiencies vary with receiver tube diameter at 170°C. The overall collector efficiency is the product of the optical efficiency and the thermal efficiency. Since optical efficiency is rising while thermal efficiency is falling, the variations tend to balance each other, producing the broad, flat maximum in the overall efficiency. Analogous plots at other temperatures all show a similar broad maximum, displaced toward larger receiver tube diameters for lower temperatures. This broad maximum assures us that receiver tube diameter is not a critical consideration and allows us to base our choice of receiver tube size on other criteria.

accurate and reproducible measurement of the tilt angle. The end pieces are slotted to let us vary the position of the receiver tube along the normal axis. Clearly, such refinements will be unnecessary in an operating system.

We are making the first outer envelope of polyester film, which is strong, clear, and easy to join and aluminize but not especially resistant to ultraviolet radiation. Because the plastic jacket and its spacing washers may accidentally be exposed to stagnation temperatures, they are made of teflon. We will be investigating the many other plastic materials that are available as thin films to select those with the best combinations of properties.

Our main purpose in conducting these experiments will be to verify the accuracy of our computer codes; a secondary purpose is to explore the practical difficulties of construction and operation. These units will also allow us to measure quantities that are difficult to calculate, such as wind loading effects and motion of the outer cylinder.

Key Words: process heat; solar collectors; solar energy; solar ponds; solar radiation—applications.

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- Achievements and Developments at LLL (*February 1977*)

NOTICES

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