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A SURVEY OF THE HAZARDS INVOLVED IN
PROCESSING LIQUID METAL BONDED FUELS

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OAK RIDGE NATIONAL LABORATORY

operated by

UNION CARBIDE CORPORATION

for the

U.S. ATOMIC ENERGY COMMISSION

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ABSTRACT

A survey of the character and magnitude of hazards involved in processing liquid metal bonded fuels was made and the scope of a preliminary experimental program outlined. Processing of SRE and CPPD fuels by mechanical decladding followed by controlled reaction of the collected NaK and Na is recommended over chemical dejacketing methods. Similarly, shearing of PRDC fuel and controlled exposure of the Na in the severed portions to water appears more desirable than chemical dissolution of the metallic cladding.

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1.0 SUMMARY AND RECOMMENDATIONS

A survey of the available data of Na and NaK reactions with water and the properties of spent fuels to be processed and consideration of the probability and potentially costly result of a Na or NaK explosion in a radiochemical processing plant led to the following conclusions:

1. The SRE and CPPD fuels should be processed by mechanical decladding with collection of the liquid metal bond in an inert medium and disposal by controlled reaction with water in a separate vessel. Single tubes from both fuel assemblies contain a large (~100 g) reservoir of liquid metal and present an excessive hazard if breached in a reactive atmosphere. This type of element is more suited to mechanical disassembly because of its large diameter (SRE 0.79 in., CPPD 0.65 in.) and large bond thickness (10-30 mils). A facility* now exists for processing SRE elements in this way.
2. The PRDC axial and radial blanket elements should be processed by breaching the clad, most suitably by shearing of single rods or possibly of tubular bundles, and reacting the bond metal in situ with water, followed by leaching of the core from the residue of cladding. These elements contain only a small amount of bond metal (2-10 g Na), in part well-distributed over the length (≤ 0.16 g Na/in.), and may be breached safely in a reactive atmosphere under carefully controlled conditions. This type of element is better adapted to processing by in situ reaction of the Na bond because of its smaller diameter (0.443 in.) and thin bonding (4 mils), which make mechanical disassembly more difficult. The application of mechanical disassembly procedures, however, should not be ruled out. If feasible, only slight modification of present facilities would be required.

If further development of processing methods for the PRDC blanket elements and the CPPD core is contemplated, the following preliminary program is recommended:

1. Make an experimental study of the average damaging event with prototypes of PRDC blanket elements and of the maximum damaging event with small amounts of Na (< 10 g). Adequate equipment design depends on successive handling of the maximum damaging event, which cannot be produced easily by even a large number of routine tests on prototypes but rather must be created intentionally by careful experimentation.
2. Make preliminary tests of mechanical disassembly of prototype PRDC blanket elements to determine the feasibility of processing these with only small modification of the existing SRE facility.
3. Initiate a study of modifications necessary to process the CPPD core in the existing SRE facility. These promise to be extensive and expensive unless a simplified method is evolved. The carrier approach route

* TID-7583; TID-7599, Book 2.

to the cell face, cell length (25 ft), equipment size, and placement will require changes to accommodate these extremely long elements (> 15 ft, 4-3/8 in.). The second core will contain uranium carbide (UC), rather than U-Mo alloy, and processing of this core may require different concepts; this factor should be considered before investing heavily in modifications in building and equipment.

Decladding processes, Darex (aqua regia) and Sulfex (H_2SO_4), do not appear feasible for liquid metal-bonded stainless steel-clad fuel types. Darex processing is prohibitively hazardous; nitrogen oxide-hydrogen explosions appear unavoidable. Direct Sulfex decladding or decladding after in situ reaction of the Na bond of mechanically punctured elements is less hazardous and may be feasible, although sodium reacts violently in boiling 4 M sulfuric acid (and water), since bursts of pressurized hydrogen rather than true explosions are expected. Reaction of limited amounts of sodium on the surface or on a shallow layer of aqueous solutions is permissible since the gaseous hydrogen produced is vented rapidly without a dangerous pressure buildup. However, the dense sodium oxide smoke in hydrogen gas creates a special off-gas handling problem. Reaction at depths of 6 in. to 2 ft and more results in much higher pressure buildup since bodies of liquid cannot vent rapidly. In general, liquid metal-water reactions occur over a millisecond range but may be controlled under some conditions since they are limited by the rate of mixing rather than the chemical reaction rate.

2.0 INTRODUCTION

Consideration of methods applicable for interim processing of stainless steel-clad liquid metal-bonded uranium alloy fuels* has led to a search for safe methods of disposing of the liquid metal that are amenable to use in an aqueous processing plant. This report reviews previous experience with liquid metal-water reactions, and attempts to assess the feasibility of some chemical-mechanical methods that have been proposed and to define the type of experiments that could give an acceptable, reliable measure of the probability and magnitude of hazardous events.

A mechanical process using an inert oil blanket has been developed for disassembling and decladding the SRE Core 1. The NaK bond metal is then reacted separately under an oil blanket with superheated steam under a nitrogen blanket. However, from a cost standpoint, a chemical process that could be operated in existing aqueous processing plants might be advantageous since costly mechanical equipment would not have to be added.

Violent reactions and one explosion occurred during attempts to declad prototype pins directly by the Sulfex and Darex processes.

3.0 CHARACTER AND MAGNITUDE OF HAZARDS ASSOCIATED WITH Na AND NaK

Sodium and sodium-potassium alloys are highly reactive materials. At ordinary temperatures the only common chemicals (except some metals) inert

* SRE, CPPD, PRDC blanket elements.

to them are paraffinic oils, nitrogen, argon, and helium. Liquid ammonia and higher alcohols react relatively slowly. The reactions with other chemicals proceed rapidly and are usually highly exothermic; specifically for water and oxygen, using sodium as an example,¹⁻³

The reaction to form sodium hydroxide occurs in excess water and the reaction of NaOH with Na occurs in excess sodium when NaOH becomes molten. NaH forms from hydrogen in an excess Na system up to its decomposition temperature (~700°F). Na reacts with oxygen in an atmosphere containing > 5% O₂. A peroxide, Na₂O₂, and a superoxide, NaO₂, may also be formed under some conditions. In air atmospheres, hydrogen-oxygen combustion or explosion usually occurs concurrently. Under some conditions⁴ the above liquid metal reactions have been found difficult to initiate (because of protective films, inability to mix the reactants, low temperature, etc.); however, these reactions usually proceed rapidly (fire and/or explosion) as long as the reactants remain and can contact each other or the temperature is permitted to rise. A survey⁵ of reactions of liquid metals with a wide variety of industrial materials and chemicals illustrates the extreme reactivity. In connection with fuel processing applications there are several specific dangerous events to be guarded against during the reaction of Na or NaK with water:

1. Fast (explosive) reaction of dispersed liquid metal with liquid water accompanied by sudden expansion of pressurized hydrogen and steam, particularly in restricted geometry.
2. Explosion of the hydrogen generated with oxidizing chemicals in the system which may include air, nitric acid, nitrogen oxides, chlorine, and possibly others.
3. Combustion of paraffinic blanketing oil or a liquid metal emulsion in oil with air or of liquid metal with air or carbon dioxide when these materials are used in processing.
4. Disastrous temperature excursions when excess liquid metal reacts with a limited amount of water (without an auxiliary heat removal system, i.e., an oil bath as an inert coolant).

Large quantities of Na and NaK have been handled safely in high-temperature heat transfer systems for several years.² Large amounts have been disposed of safely both by reacting underwater and by combustion in air. Reports of underwater disposal methods^{1,6,7} at high rates through nozzles under 5-10 ft of water indicate no fire, detonation, surface burning, or smoke.

One patent⁸ exists on a cyclone-like device to dispose of liquid metals. These methods depend on high shear and good mixing in excess water to ensure immediate reaction, without accumulation of the liquid metal. Methods of disposing of wastes, cleaning equipment, etc., used are combustion in air aided by kerosene, anhydrous ammonia, alcohol, etc., all followed by steam and/or water washing. These are not, in general, applicable to processing of radioactive fuels. The intimate, restrictive configuration required in stainless steel-clad liquid metal-bonded uranium-alloy fuels poses special problems even though only small amounts of Na or NaK are present in any one tube.

A measure of the magnitude of the hazard can be inferred from an abortive attempt to clean a carbonaceous sodium scale (deposited by the heat transfer fluid) from the outer surface of fuel assemblies of the SRE Core 1 at Atomics International. An assembly suspended vertically in a nitrogen-filled cell was sprayed with water from a hose. Violent reactions occurred, resulting finally in a detonation which imploded the thin (10-mil) cladding, rupturing it and releasing the NaK bond metal. Assemblies are now being cleaned successfully by the reaction of NaK with isoamyl alcohol diluted with about 10 parts of kerosene.⁹

Allowing direct reaction of the bond metal (of from one tube to a complete bundle) to occur in a dissolver during decladding by either the Darex or Sulfex process would give the result desired most simply in a single vessel without transfer problems.¹⁰ A small number of trial runs of this type were made on prototype pins. One of five Darex decladding runs terminated abruptly in an explosion;¹¹ no explosion occurred in three Sulfex runs but some violent reactions were observed.¹² The explosion was tentatively attributed to ignition of a hydrogen-nitrogen oxide mixture in a gas bubble by an incandescent particle of NaK.

However, the small number of tests hardly excludes the possibility that a more violent reaction could occur during a Sulfex decladding. Direct decladding in this manner appears to be an extremely dangerous process.¹³ This type of test, seeking the occurrence of what may be a relatively rare event, gives inconclusive results unless an explosion is produced. However, the capability for a damaging reaction exists and depends on the amount of reactants present and the probability of these being brought together and reacted in a short enough time.

With careful experimental methods and a large number of tests it might also be possible to assign values to the probability of the occurrence of damaging events of given magnitudes for some applicable configurations of element, vessel, and method of introduction of water. However, the consequences of an explosive event are potentially so costly as to render the above experimental methods useless to produce a sufficiently reliable estimate of the probability of the event. A simpler experimental approach would be to produce intentionally the maximum credible damaging event with the amount of sodium involved under experimental configurations simulating expected methods of operation. If equipment can be designed to meet this condition, safe operation may be shown to be possible.

Other evidence on the reaction of Na and NaK with water, summarized in a review¹ of the subject, indicates that an explosively fast reaction is quite possible in water. After attempts to determine the actual reaction rate of NaK with water, experimenters¹⁴ concluded that the reaction rate was essentially instantaneous (< 2.5 msec), with the total reaction time of a given mixture being controlled by the rate of mixing. In tests^{1,6,7} introducing NaK into water through different sized nozzles, it was found that, if the contact area was constant (either laminar or turbulent flow), the reaction was smooth and continuous, but if the area varied (as in transition flow) the reaction became spasmodic. Both a high initial pressure and high temperature of reactants were found to result in smoother reactions under given experimental conditions.^{15,16} A high initial temperature ensures prompt initiation, and high initial pressure reduces spasmodic mixing events from bursts of hydrogen and promotes a smooth, continuous reaction. Spasmodic reactions are accompanied by a higher rate of pressure rise and higher pressures.

Most processing methods that do not involve complete mechanical decladding in or under an inert medium* fall in the class likely to result in spasmodic reactions, depending somewhat on the method in which protective cladding is breached for removal of the bond. Elements with a sodium bond are particularly subject to this danger since sodium is a plastic solid below 97°C .

An attempt to reduce the reaction rate of NaK with liquid water by using a concentrated aqueous solution, 10 M KOH, was not effective.¹⁴ In rapid mixing events a rate of pressure rise of the order of 6 atm/msec was observed. The rise was limited by the maximum rate of mixing possible in the apparatus and not by the chemical reaction rate. With larger amounts of sodium, exceedingly violent reactions are expected from secondary reactions following an initial burst in which an appreciable amount of the reactants is dispersed into very fine particulates with a high surface area.

Limited reports of some work done at Hanford on underwater sawing of Na- and NaK-containing capsules, using both a friction saw and a hacksaw, have been published.¹⁷⁻²⁰ It was concluded¹⁷ that 5-6 g of Na could be safely released underwater by shearing using an inert gas blanket to prevent burning or explosion in the gas space over the water. Later results¹⁸ of sawing NaK-containing capsules under 6-24 in. of water showed that the amount of NaK reaching the water surface decreased but explosive pressure increased as the depth of water increased. Finally, tests¹⁹ showed that instantaneous exposure of up to 24 g Na under a maximum of 2 in. of water submergence could be handled safely if the gas space was purged with helium to reduce the oxygen concentration to less than 5%. It was concluded that PRDC core pins could be handled successfully in this manner.²¹ Dense white oxide fumes were formed in the gas space and must be scrubbed out of the off-gas.²²

In further studies at Hanford²³⁻²⁶ simulating conditions in direct decladding, Na-containing copper capsules were allowed to react under 8 ft

* Suitable inert materials are: among liquids, paraffinic oils; among gases, N_2 , Ar, He; CO , CO_2 and H_2 can react with liquid metals.

of 60% nitric acid at room temperature in an 8-in.-dia 15-ft-high pipe (dissolver). The pressure transient was followed with a ballistic transducer recording an oscilloscope trace. With 11 and 22 g of Na, peak pressures of 375 psia and 850 psia, respectively, were recorded. In the latter test, some acid was splashed out of the dissolver and the unit (weighing ~700 lb) was displaced sideways 1 ft. In additional tests with 11- and 20-g batches of Na, pressures of 700 psig and 850 psig, respectively, were measured. The most reliable values of peak pressure were reported²⁵ as 4 g of Na, 250 psig; 11 g of Na, 700 psig; and 22 g of Na, 850 psig. Although the events were violent it is not certain from the reports that the maximum damaging event per gram of liquid metal was obtained since a series of explosions occurred in each test.

A complete and excellent review³ of sodium-water reactions was made by the staff at Nuclear Development Corporation of America (NDA) in connection with design of the sodium-cooled D₂O-moderated reactor (SDR). This review includes articles referred to here and others. The main interest of NDA is in Na-H₂O heat transfer systems. However, some information in their report applies to the static Na-H₂O reaction system found in fuel processing applications:

1. Only a small expansion volume is required to eliminate shock wave formation.
2. Pressure buildup can be decreased by providing expansion volume and venting facilities.
3. Attempts to limit Na-H₂O rates by chemical additives have not succeeded.
4. The Na-H₂O reaction occurs in the millisecond range and its kinetics are limited by diffusional processes (rate of mixing of reactants).
5. The major energy release (underwater) is in heat and in pressurizing the hydrogen produced. Very little occurs as shock wave energy.
6. Detonations occur in the microsecond range, resulting in intense shock waves; the Na-H₂O reaction is usually a milder event, usually called a "deflagration," which produces shocks of limited intensity, if any.

In recent experimental studies at NDA^{27,28} techniques were developed for measuring the rate of reaction of droplets of sodium on the surface of liquid water and time-temperature traces of reacting Na in flowing streams of argon containing water vapor. Reactions of pieces of Na (50-200 mg, at 27°C) on the surface of water (at 27°C) were 50% complete in ~10 sec, a relatively slow reaction, even though particles glowed at times. When the Na was preheated to 300°C, reactions were completed in ~2 sec. Reaction rates could be retarded by foaming agents or oils which tend to coat Na, and accelerated by addition of acetone or glycerine to the water or by using Na in Pb or Hg alloys.

Reaction of small pieces of Na in a flowing stream of moist argon (partial pressure of water ~14 mm)²⁸ demonstrated the drastic temperature excursions possible in water-deficient systems where no effective coolant

is available. Depending on the initial droplet temperature, average rates of temperature rise of $\sim 200^{\circ}\text{F}$ per second were measured over periods of ~ 5 sec, resulting in peak temperatures of 1300 - 1750°F .

Experimental work at NDA showed that in processing Na-bonded fuel elements the reaction of Na with water carried in a gas phase (or steam), without supplying a liquid heat transfer agent, will result in a dangerously high temperature within a short time, and that reaction of Na particles on the surface of liquid water is relatively slow if the reactants are at room temperature.

Comments on safety in handling liquid metals and in cleaning Na- and NaK-contaminated equipment, applicable in part to the present problem, are found in the current literature.^{1,2,29-33}

4.0 REVIEW OF SOME PROPOSED PROCESSING METHODS

A number of procedures for the removal and disposal of the liquid metal bond have been suggested.^{10,34} Problems of breaching of the fuel jacket, required in all processing methods, and several possible operational methods are discussed in light of the specific type of hazards pointed out in Sect. 3.0. Shearing and/or mechanical disassembly appears to offer the safest and surest, if not the most convenient, method of processing spent reactor fuels.

4.1 Breaching Fuel Elements

Breaching of fuel jackets by various methods, e.g. puncturing, slitting, breaking, or shearing of either single tubes or assemblies, is required for removal of the bond metal and subsequent reaction with water. The initial breaching action must be handled with care. At 98°C or above it will quite possibly be accompanied by spurting of the liquid sodium* because of seal gas and fission gas pressure. Even at temperatures below 98°C some spurting may occur since sodium is a plastic solid that will flow under pressure. If liquid water is available, either as a medium or in a pool beneath the element, rapid mixing with an appreciable amount of liquid metal will result in a pressure rise if the vessel is blanketed with nitrogen. A hydrogen-oxygen explosion is possible if the vessel is filled with air. Under conditions where water can enter the tubes through puncture holes (while a temporary protective NaOH scale covers the sodium surface inside), the resultant spasmodic pressure surges from the Na- H_2O reaction after dissolution of the scale in water again could be explosive. Sudden underwater exposure of Na with a large surface area would result in high pressures.

The outstanding characteristic required of a breaching method is certainty; shearing or mechanical disassembly supply this to a degree not possible by other procedures.

* NaK (78% K) will flow readily above room temperature.

4.2 Operation in a Canal

The breeching operation is not considered feasible underwater in an open storage canal unless the following contingencies can be coped with:

1. Gaseous activity release
2. Particulate activity release
3. Combustion of hydrogen above the water surface
4. Na_2O fume release
5. Caustic formation in the canal water

Similarly, a smaller restricted chamber in an open canal suffers to a lesser extent from the same difficulties. The reaction chamber could be (a) hooded and inert-gas-blanketed to prevent hydrogen combustion and to collect activity and fumes, and (b) stainless steel-lined to reduce corrosion and with a separate water supply and disposal system to handle contaminated water. Facilities for removal of particulate fissionable material from the bottom of the canal may also be required. A resealable water lock between this chamber and the main storage canal is also needed since it is not feasible to handle fuel elements or to operate at any time without a full depth of water for shielding purposes. This factor also precludes breeching elements in inert gas (in the canal area) followed by reaction with water. In addition, long, heavy elements with a thin cladding, characteristic of liquid metal-bonded fuels, are relatively weak and require careful handling. Extensive handling and transfer are not advisable, particularly after breeching. One advantage inherent in operating in a canal, rather than an in-cell vessel, is that breeching elements would be easier and much more certain because of greater accessibility. However, the description of facilities required for a canal suggests that an in-cell vessel, i.e., a dissolver, may have the same features with fewer disadvantages.

4.3 Operation in In-Cell Vessels

The characteristics of the disposal operation seem to require an in-cell vessel, rather than a canal, i.e., a dissolver or other reaction vessel designed specifically for this purpose. The combined use of a dissolver for bond removal, dejacketing, and core dissolution offers the most desirable attributes from several viewpoints:

1. There is no mechanical transfer of elements that have been breeched, exposing highly radioactive core material.
2. There is single vessel accountability for fissionable material, fission products, and sodium.
3. The use of only a single vessel and auxiliaries is cheaper and requires less space.

The solution to this problem, however, rests on conceiving operating procedures that meet the following stringent criteria:

1. It must be ensured that no detonation is possible.
2. The vessel must be designed to resist the maximum damaging pressure, and the vent system must be designed to handle the gas and/or liquid flow associated with this event if such limiting magnitudes can be determined.
3. The device used for breeching must operate without failure on all tubes inside a closed dissolver. (An assembly or several tubes must be charged each time to maintain a high overall usage of a dissolver.)
4. Safe and complete reaction of the liquid metal must be ensured. This entails limiting one of the reactants to control the maximum rate of energy release to within the limits of the equipment design for temperature, pressure, and pressure gradients per unit time.
5. It must be demonstrated, by an accountability procedure that recovers both NaOH formed and Na₂O that has gone off in the gas phase, that the reaction is complete.

Several formidable development and design problems have to be solved to meet these criteria.

Direct dejacketing in acid and underwater breeching in a dissolver are eliminated under these criteria as not being controllable. For the SRE (NaK) and CPPD (Na) elements, having ~100 g of bond metal per tube, a steam-nitrogen mixture is the only method of supplying water that meets the criterion that one of the reactants be limited and have no way to accumulate in the system. In a dissolver or a separate reaction vessel, extremely high temperatures could result unless an inert heat transfer medium was provided.

The alternative procedure is to limit the amount of liquid metal. This method appears to be applicable to single tubes from the PRDC (Na) blanket elements which contain only ~0.16 g of Na per inch of length for a total of 2-10 g per tube. Exposure of this small amount of Na, distributed in this way, may prove feasible under water in an enclosed system.

4.4 Shearing Methods

The use of the mechanical shear, designed for shearing bundles of stainless steel-clad ceramic fuels into short lengths,²⁵ has been suggested as a means of ensuring removal and reaction of a sodium metal bond from stainless steel-clad uranium-alloy fuels. This may be a feasible method of limiting the amount of sodium metal allowed to react at any given time. The blade assemblies are enclosed and could be blanketed with nitrogen. The sheared pieces fall from the nitrogen-blanketed area into a water-containing vessel to complete the reaction. Control of temperature near the shear head could be difficult if a steam-Na reaction occurred there. On the other hand, this type of blade shear may not be suitable for stainless steel cladding as thin as 10 mils. Blade wear by galling in close clearances is greatly increased for clad thicknesses less than 20 mils.

An alternative, and much cheaper, shear is a rod cutter for single tubes. Chopping in an inert atmosphere and allowing the pieces to drop into water appears to offer a reasonably safe technique pending experimental measurements of the consequences of the maximum damaging event. This method appears to satisfy all the criteria for a safe reaction: (1) The amount of sodium present is limited by controlling the rate of chopping; (2) sodium does not accumulate since it is readily accessible to water in short chopped lengths; and (3) the pool of water is an effective heat transfer agent.

4.5 Mechanical Disassembly

The SRE-1 core will be processed by complete mechanical decladding under an inert oil blanket and reaction of the NaK (bond metal) under an oil blanket with superheated steam by bubbling the gas phase into the liquid metal phase. Uranium slugs are to be steam cleaned of residual NaK and oil and subsequently dissolved in nitric acid.³⁵

Other types of liquid metal-bonded fuels are less amenable, but perhaps adaptable, to being processed in the existing facility. The CPPD core is excessively long (166 in.); the slugs in the PRDC blanket elements are of small diameter (0.415 in. vs 0.75 in. for the SRE) and removal from the clad may be more difficult. Both have a sodium bond (melting point, 97°C), requiring a higher operating temperature than for elements with a NaK bond (melting point is 16°F for NaK of 78% K). However, these elements could probably be completely disassembled mechanically in properly designed equipment. At present, mechanical disassembly appears the safest method of handling stainless steel-clad liquid metal-bonded fuels. Details of the method of operation will require considerable development before use in the available facility.

5.0 APPLICATION TO CURRENTLY AVAILABLE FUELS

Each of the liquid metal-bonded fuels (first cores) now available and subsequent cores constitute separate problems because of the type of cladding, length, diameter, construction of the assembly, amount and type of bond metal, and core material composition. A consideration of the general properties of the four kinds of fuels to be available soon allows a division into two types (Table 1):

Type I: SRE and CPPD cores: ~100 g of liquid metal per tube; large diameter, 0.75 and 0.65 in.; thick bond, 10-25 mils; length 90-180 in. (Designer, Atomics International).

Type II: PRDC axial and radial blankets: 2-10 g of Na per tube; small diameter, 0.423 in.; thin bond, 4 mils; length 17-72 in. (Designer, Atomic Power Development Associates).

The large diameter and thick bond make it easy to remove the slugs of Type I elements mechanically. These fuels would be dangerous to handle near water because of the large amount of liquid metal per tube. Processing of them by other than mechanical disassembly probably should not be considered at this time. Arrangements to mechanically process the SRE have already been made. The CPPD Core 1 may not be cleaned of Na residue on the outside,

Table 1. Na and NaK-bonded Fuels to be Available

Fuel	Composition	Core Properties			No. of Tubular Elements per Assembly	Clad		Bond Type	Bond Thickness, in.	Total Wt of Liquid Metal per Subassembly, g	Total Wt of Liquid Metal per Assembly, g
		Length, in.	Slug Dia, in.	No. of Slugs per Tube		Length, in.	OD, in.				
Type I											
SRE Core 1	U	72	0.75	12	7	92-5/16	0.79	NaK (+ He)	0.010	110	770
CPPD Core 1	U-10% Mo	162	0.59	1	18	182	0.66	Na (+ He)	0.025	111	2000
Type II											
PRDC Axial Blanket	U-2.7% Mo	14	0.415	2	16	17.5	0.443	Na	0.004	~2.25	~36
PRDC Radial Blanket	U-2.7% Mo	65	0.415	8	25	71.5	0.443	Na	0.004	~9.7	~242

making it difficult to handle by any process. The CPPD Core 2 will have a uranium carbide core, creating an additional extremely hazardous situation associated with hydrogen and acetylene gases which evolve from UC in contact with water or steam. The unique properties of Na-bonded UC elements will probably require the development of special techniques for processing subsequent CPPD cores, and it is suggested that preliminary development studies be undertaken on a mechanical disassembly process capable of handling Na-bonded elements as long as the CPPD cores 1 and 2 before the first core becomes available (September 1963).

Type II (PRDC) elements are much more amenable to direct reaction of the bond with water since they have a very small amount of sodium in a thin bond layer, ~0.16 g Na per inch of length. Direct reaction with water or steam may be feasible since the maximum damaging reaction is limited by the small amounts of sodium present. Experimental work with prototype pins and small amounts of Na may be fruitful in showing that processing of this type of core is feasible.

6.0 PRELIMINARY EXPERIMENTAL PROGRAM

Consistent with the conclusion that initial experimental work should first be done on fuel elements like the PRDC (Type II), containing only a small amount of sodium, it is suggested that the experimental program be limited initially to the following types of studies:

1. Determination of the maximum credible damaging event (exclusive of an H_2-O_2 explosion) in the reaction of ~10 g of sodium, solid and liquid, with H_2O , as water and steam, under suitable experimental conditions.
2. Observation of the reaction of prototype pins breeched in environments simulating proposed methods of process, e.g., underwater, in superheated steam, in a steam-nitrogen mixture, or in steam over boiling water, and chopping of single pins into water.

Instrumentation adequate to measure the temperature and pressure transients is required for some types of experiments. Closer review of the work at Hanford, however, may indicate some of their results to be adequate to define certain experimental procedures sufficiently well. In particular, sawing and shearing results in water show that shearing elements in an inert atmosphere (< 5% oxygen) into shallow water (~2 in. deep) has promise as a safe processing method. Work is also progressing on methods of handling the dense oxide fumes.

7.0 REFERENCES

1. C. B. Jackson et al., "Liquid Metals Handbook, Sodium-NaK Supplement," USAEC Dept. of the Navy, Washington, D. C., TID-5277 (July 1, 1955).
2. M. Sittig, "Sodium: Its Manufacture, Properties and Uses," Reinhold Publishing Corp., New York, 1956.

3. "Chemical Considerations on the Sodium-Cooled, D₂O-Moderated Reactor (SDR)," NDA-84-6 (April 30, 1958).
4. G. A. West, "NaK-Water Reaction Experiment," ORNL CF-60-2-32 (Feb. 10, 1960).
5. C. O. Nelson, "Summary Report of Reaction Tests on Various Materials Tested with Sodium and Sodium-Potassium," KAPL-557 (June 1, 1951).
6. W. Milich and E. C. King, "Underwater Disposal of Sodium with EB Nozzle," Mine Safety Appliances Company, NP-5670 (June 14, 1955).
7. R. C. Anderson and E. C. King, "Underwater Disposal of Molten Sodium," Mine Safety Appliances Company, NP-5261 (June 30, 1954).
8. British Patent No. 574360, "Improved Apparatus for Reacting Sodium and Water," Jan. 2, 1946.
9. C. D. Watson and W. F. Schaffer, Jr., "Trip Report: Visits to Atomics International, Canoga Park, California and Vallecitos Atomic Laboratory, Pleasanton, California," ORNL CF-60-4-63 (March 28, 1960).
10. A. R. Irvine, "Power Reactor Fuel Processing: Required Unit Operations Development," ORNL CF-60-1-98 (Jan. 21, 1960).
11. J. C. Bresee et al., "Chemical Technology Division, Unit Operations Section Monthly Progress Report, August 1959," ORNL CF-59-8-76.
12. F. G. Kitts, unpublished data, ORNL Notebook No. A-1322, August 1959.
13. D. E. Davenport, Stanford Research Institute, Consultant to ORNL, Meeting on Feb. 18, 1960.
14. M. Kilpatrick et al., "Studies of Fast Reactions Which Evolve Gases. The Reaction of Sodium-Potassium Alloy with Water in the Presence and Absence of Oxygen," J. Phys. Chem., 57: 385-390 (1953).
15. E. C. King, "The Reaction of NaK and H₂O," Mine Safety Appliances Company, NP-3334 (Sept. 7, 1951).
16. E. C. King, "The Reaction of NaK and H₂O," Mine Safety Appliances Company, NP-3646 (Confidential)(Jan. 30, 1952).
17. L. P. Bupp, "Chemical Research and Development Monthly Report," Hanford Atomic Products Operation, HW-59717C (Secret)(March 1959).
18. L. P. Bupp, "Chemical Research and Development Monthly Report," Hanford Atomic Products Operation, HW-60233C (Secret)(April 1959).
19. L. P. Bupp, "Chemical Research and Development Monthly Report," Hanford Atomic Products Operation, HW-60505C (Secret)(May 1959).

20. V. P. Kelly, "High-Speed Saw Tests, Power Reactor Fuels Reprocessing Development," HW-62490 (Nov. 18, 1959).
21. V. P. Kelly, Hanford Atomic Products Operation, personal communication, June 17, 1960, at ORNL.
22. G. Rey, Hanford Atomic Products Operation, personal communication, February 1960, at ORNL.
23. L. P. Bupp, "Chemical Research and Development Monthly Report," Hanford Atomic Products Operation, HW-61374C (Secret)(July 1959).
24. L. P. Bupp, "Chemical Research and Development Monthly Report," Hanford Atomic Products Operation, HW-61702C (Secret)(August 1959).
25. J. J. Shefcik and C. E. Huck, "Transient Pressures Developed by Sodium-Nitric Acid Reactions," HW-62450 (Oct. 21, 1959).
26. J. J. Shefcik, Hanford Products Operation, personal communication, June 17, 1960, at ORNL.
27. L. Corrsin et al., "Sodium-Water Reaction Rate Status," NDA-84-19 (May 15, 1959).
28. E. Bernsohn, "SDR Project Quarterly Technical Progress Report No. 6," NDA-84-13 (Aug. 1 1958 to Oct. 31, 1958).
29. Reactor Handbook, 1st Edition, Vol. II, page 328ff, "Recommendations for Handling the Reaction between Alkali Type Liquid Metals and Water," from a memo by E. C. King, Mine Safety Appliances Company (May 1955).
30. "Procedure for Handling Sodium and Other Alkali Metals. Special Chemical Technology Division Regulations in the Chemical Technology Division Safety Manual," ORNL CF-60-4-32.
31. Bulletin from National Distillers Chemical Corp., "Handling Metallic Sodium," 1951.
32. C. G. Banister et al., "A Report on the Proceedings of the Liquid Metal Utilisation Conference," AERE X/R 1381 (1954).
33. "Handling and Uses of the Alkali Metals," Advances in Chemistry Series No. 19, American Chemical Society, Washington, D. C., May 1957, paper entitled, "Recleaning Sodium Heat Transfer Systems," H. W. Braggeman et al., General Electric Company (1957).
34. C. D. Watson, ORNL, personal communication.
35. "Proceedings of the AEC Symposium for Chemical Processing of Irradiated Fuels from Power, Test, and Research Reactors, Richland, Washington, October 20-21, 1959," paper entitled, "Mechanical Processing of Spent Power Reactor Fuel at Oak Ridge National Laboratory," C. D. Watson et al., TID-7583, p. 306.

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