UK FAST REACTOR COMPONENTS – SODIUM REMOVAL DECONTAMINATION AND REQUALIFICATION

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PART 1 - APPROACH

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INTRODUCTION

Over the past two decades extensive experience on sodium removal techniques has been gained at the UKAEA's Dounreay Nuclear Establishment from both the Dounreay Fast Reactor (DFR) and the Prototype Fast Reactor (PFR). This experience has created confidence that complex components can be cleaned of sodium, maintenance or repair operations carried out, and the components successfully re-used. Part 2 of the paper, which describes recent operations associated with the PFR, demonstrates the background to these views. This past and continuing experience is being used in forming the basis of the plant to be provided for sodium removal, decontamination and requalification of components in the UK's future commercial fast reactors.

Further improvements in techniques and in component designs can be expected in the course of the next few years. Consequently UK philosophy and approach with respect to maintenance and repair operations is sufficiently flexible to enable relevant improvements to be incorporated into the next scheduled fast reactor - the Commercial Demonstration Fast Reactor (CDFR). This paper summarises the factors which are being taken into consideration in this continuously advancing field.

There are two options on component maintenance/repair philosophy - recovery for re-use (including part recovery) or throwaway. Plant provisions to meet these alternatives can cover a wide spectrum. At one extreme lies provision of a comprehensive facility which can cope with all the operations required in maintaining or repairing the range of components installed in the primary and secondary circuits of fast reactors. At the other extreme lies the limited facility required to meet only a throwaway philosophy: for example, in this case the sodium removal/decontamination process installed need not be designed to avoid component damage but only to meet the conditioning requirements for subsequent disposal. The UK approach lies between these extremes, ie some components will warrant recovery for re-use, and is the basis for determining the extent of facilities to be installed.

Classification of components into recovery and throwaway categories is the subject of on-going assessments. Concomitantly details of plant facilities to be provided will be defined: decisions of this nature are not required urgently and will be taken when all necessary data on reactor and component design, development programmes, and associated assessments are completed. For example, as data continue to be amassed from PFR and as CDFR component designs become more detailed, re-assessments against the headings below will be carried out.

i   Component: Further analyses of each component will be carried out covering ease of disassembly, estimated frequency of discharge (involving information on reliability and maintenance frequency), expected level of contamination, decontamination factor required for hands-on maintenance. The cost of the component (or its parts) will also be necessary to assess against the cost of recovery.

ii Radioactive Waste: The extent of solid waste arisings and of effluent treatment requirements and their associated costs will be included in assessments.

iii Sodium Removal/Decontamination Procedures: Selection of sodium removal/decontamination and maintenance procedures (not processes) raises a number of issues, viz:

a. Separate facilities or not for primary and secondary components. Considerations of minimising active effluent arisings and ensuring no contamination of secondary circuit components are relevant in this context.

b. Remote maintenance or decontamination and hands-on maintenance. The extent of active nuclide penetration into the substrate of component materials is relevant in that decontamination factors which permit hands-on maintenance may not always be achievable. (Note that this factor emphasises minimising activity release in fast reactors and interacts therefore with tribology restrictions, operating philosophy with fuel pin failures, and the need for development of activity traps.)

c. Component breakdown before or after decontamination. In this context the detailed component design will be relevant.

d. Immediate or delayed sodium removal (and decontamination). This issue will arise if, for example, component maintenance operations have to be delayed for any reason.

iv Plant Provisions: Selection of the sodium removal/decontamination process and decisions on procedures arising from (iii) above will influence factors such as maintenance plant type and layout.

v Radiation Dose to Operators: The activity dose and dose rate limitations placed on plant operators will be an important factor affecting decisions on both philosophy and procedures.

SODIUM REMOVAL/DECONTAMINATION PROCESS CHOICE

One major sodium removal process plant will be provided for CDFR. Whilst both butyl cellosolve and water-based processes have been successfully applied at Dounreay, options on three processes are being kept open at present: these are water-based, alcohol (low boiling-point) and evaporative (vacuum distillation). Current plans ensure that sufficient experience or information will be available to permit a choice to be made.

Plans are in hand to modify the existing Decontamination Pit on PFR at Dounreay to take a controlled water vapour/(nitrogen and/or carbon dioxide) process. Consequently experience will be acquired over the next few years on controlled water vapour processes applied to PFR components to supplement the already-extensive experience on water droplet processes. A water-based sodium removal stage presents an easy extension into a decontamination stage, and a combined process is to be installed at Dounreay. The main arguments against a water-based process are the uncertainty surrounding the subsequent freedom from hydroxide stress corrosion cracking damage and the possibility of a violent reaction should a pool of sodium exist within the component. These issues, particularly the former, tend to place undesirable constraints on the designers in that either stressed, creviced zones and bolted assemblies would have to be prohibited or the component capable of complete strip-down following cleaning. It must be added that no evidence of hydroxide stress corrosion cracking has yet been observed in the UK following use of a water-based process for sodium removal.

An alcohol (ethanol) process is currently being installed at Dounreay - construction is virtually complete. Experience will be gained from the cleaning of PFR components in this facility, but only on components uncontaminated with activity. The advantage of this process in reducing the problem of hydroxide stress corrosion cracking is recognised and is the reason for its installation. However, the process uses a highly flammable fluid (which will become radioactively-contaminated if used to clean primary circuit components), requires a solvent recovery stage, and will still require an aqueous decontamination stage.

The UK is also considering vacuum distillation as offering a promising alternative to fluid-based processes. In Holland the process has already been applied successfully to large components in situ. However, in situ application is not feasible for pool-type fast reactor primary circuit components, and therefore a separate cleaning plant will be required. It is appreciated that component transfer will bring in its train a number of additional problems, such as those arising from part oxidation of the contaminating sodium: such issues are being

investigated. The advantages to the component designer in relaxation of design constraints is seen as the major benefit which could arise from successful development of a vacuum distillation process for sodium removal - although drainability of sodium still remains one essential requirement which cannot be avoided.

Attention must be drawn to the advantages of a small, versatile sodium removal/decontamination facility in addition to a major cleaning plant. PFR experience has shown such a facility to be invaluable: to date it has been used for a number of components with the application of sodium removal techniques ranging from steam lancing to washing with organic fluids.

No significant problems are envisaged in the development of a decontamination process. There is little doubt that a suitable decontaminant fluid can be found, and Part 3 of the paper reports some preliminary sorting data in a decontamination fluid test programme. The key issues on decontamination relate to the extent of metal removal required to achieve an acceptable decontamination factor and the concomitant damage to the component and its surfaces.

CURRENT UK VIEWS

Current views favour discharge of a component and immediate replacement with a spare. It is appreciated that this policy requires availability of an adequate stock of spares, and the spares philosophy for both a single large commercial-type reactor (CDFR) and for a series of reactors is under continuous review. Nevertheless, studies show that this is the most economic approach.

Following removal of a sodium-contaminated component, current preference is to remove sodium as soon as possible rather than store for any significant period of time. This procedure minimises opportunity for substantial oxidation of the contaminating sodium, particularly important if the evaporative or alcohol sodium removal process is chosen. One further advantage of immediate removal of sodium is that inert atmospheres are no longer necessary: subsequent operations on the component can be carried out in an air atmosphere.

In general a remote maintenance approach for activity-contaminated components is not favoured. It is accepted that remote dismantling of some components may prove necessary because of difficulties of ensuring thorough decontamination of the assembled unit. However, remote maintenance and, more particularly, remote assembly and requalification is not believed to be a feasible approach for routine operations, especially for large components.

Fortunately those components which have moving parts, and therefore require most frequent attention, are readily strippable. UK experience has demonstrated the ease with which such components can be maintained. Consequently, the main attention in the future is expected to be focused largely on large components such as the intermediate heat exchangers and the steam generator units. The primary pumps may also require further consideration as more extensive data on extent of activity deposition and its depth of penetration become available.

The views expressed above, including those on selection of sodium removal process, pre-suppose a component recovery philosophy. They may be reinforced or modified as experience from PFR continues to accumulate, both during the course of normal operational events and as part of the development/endorsement policy of the

on-going commercial fast reactor programme. Continuing dialogue between reactor designers, system developers and plant operators will ensure that the interactions between component performance specification, component design and maintenance/repair philosophy is appreciated by all concerned.

PART 2 - SODIUM REMOVAL

BY

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INTRODUCTION

There is no single preferred method of sodium removal used in the various UK liquid metal installations. The techniques include steam, water droplet dispersion in nitrogen, alcohols and (to a limited extent on small components) vacuum distillation or hot oil cleaning. Work on full-scale reactor components is centred at the Dounreay Nuclear Establishment in Scotland where DFR and PFR are situated.

Major sodium removal and decontamination facilities have been provided at PFR based on aqueous processes and these have been augmented recently by an alcohol plant capable of dealing with large items. A number of components from PFR have been cleaned and returned to service.

PFR FACILITIES

The main sodium removal and decontamination facilities are situated within the reactor hall where direct access from the large overhead travelling crane is possible. An underground pit 18.75 m deep and 6.4 m square has been excavated in the rock strata below the reactor floor. This is used to house a cylindrical stainless steel vessel 18 m in length capable of accommodating each of the main removable reactor components. The pit also contains access platforms at several levels together with stairways and a hoist (Fig 1).

The vessel is 2.74 m dia for the upper 12 m of its length and 1.9 m for its remaining 6 m. It is fitted with an adjustable internal support structure and has three pairs of diametrically opposed access doors at each of the working platform levels. The vessel is equipped with nitrogen, steam, water, air, vent, sample and drain systems and the gas can be recirculated through an impingement plate scrubber. The top of the vessel can be sealed with a 2.1 m dia gate valve or by one of several adaptor plates which can also act as component supports.

To deal with a particular component it is placed in the vessel using an appropriate support structure (Fig 1 shows the Charge Machine in position) and purged out with nitrogen. Steam and nitrogen are then admitted to a mixing chamber at the base of the vessel and venting/circulation rates are adjusted to maintain the required conditions. The vessel then slowly fills with a water droplet dispersion in nitrogen which is the cleaning medium. The control of the process is achieved by:

a. measurement of oxygen concentrations which are not permitted to exceed 200 ppm by volume.

b. measurement of hydrogen levels which are not permitted to exceed 4% in the vent lines.

c. monitoring temperatures of the vessel and the component which should not exceed 60°C.

d. visual checking of the component and the vessel atmosphere.

e. sampling of the condensate water which collects in the lower part of the vessel for sodium hydroxide concentration measurements.

f. acoustic monitoring to detect any vigorous sodium/water reaction noises.

The end point of the reaction is determined from items b, d and e.

At this stage water washing can be carried out using a series of spray rings within the vessel. The final stage is air drying and the component is then ready for:

a. removal for further inspection or maintenance work.

b. direct work within the vessel via the access doors.

c. decontamination procedures if required.

The lower part of the vessel has been designed to hold decontamination liquids into which a component can be immersed whilst the solution is circulated. A separate inner vessel 7 m high by 1 m dia can be fitted to reduce the volume of liquid needed for smaller components.

In addition to the pit and its vessel system PFR also houses a simple steam cabinet (Fig 2) which is a stainless steel room 3.2 m wide by 2.13 m deep x 2.6 m high. This can deal with the sodium removal and low level decontamination of small items by steam lancing, water washing and/or hand scrubbing. It has a hinged and counter-balanced roof to enable heavy items to be handled by the overhead travelling crane. A 2 tonne capacity turntable is available in the floor of the cabinet to facilitate access to a component placed on it. Steam and water lances can be inserted through rubber flaps in slots situated below the viewing windows in the front face of the cabinet. Appropriate ventilation and drainage systems are provided. This facility deals with the steady day to day flow of small items requiring cleaning for maintenance or disposal. Other small wash tanks, cabinets and an abrasive slurry washing machine can deal with the decontamination of small items.

The irradiated fuel handling cave complex at PFR contains a steam cleaning vessel for removing sodium from fuel sub-assemblies before either in-situ examination (if desired) or sending the fuel for reprocessing or further examination. The item can be heated with circulating nitrogen before admitting steam. As the steam reacts with the sodium further nitrogen purging and venting is used to keep hydrogen levels to less than 4% before venting. The end point is determined using

a sensitive hydrogen detector. Once the optimum exposure time has been established a routine procedure will be adopted for standard items. The unit is equipped with suitable control gear and instrumentation for use with lower water concentrations at lower temperatures, ie a water vapour nitrogen process if required.

Alcohol immersion processes have been used successfully on some PFR components. For non-radioactive items where there are many crevices and bolted structures (such as the steam generator tube bundles) an alcohol cleaning facility has been constructed on a site near the reactor. This comprises a stock tank, component vessel, pumped loop, disentrainment tank, venting and drain systems (Fig 3). The component vessel is 7 m long by 1.8 m dia and the free capacity of the system is approximately 12,000 litres. The liquid chosen for this process is de-natured anhydrous ethyl alcohol mixture. Reaction end point in this plant will be measured by hydrogen and alcohol analysis.

SOME EXAMPLES OF PFR COMPONENT CLEANING

Two primary pumps and one secondary pump have been removed for inspection and maintenance. They were bagged out using the "engineered" silicone elastomer impregnated fibreglass bagging systems provided for that purpose. One of the primary pumps and the secondary one were required to be cleaned before the sodium removal and decontamination facility was fully commissioned.

The aqueous based water droplet dispersion in nitrogen for which the plant was designed could not be used and it was therefore decided to clean them by immersion in an alcohol solvent. The material preferred for this purpose was ethylene glycol n-butyl ether ($CH_3-(CH_2)_3-O-CH_2-CH_2-OH$) which is usually known under its trade name of butyl cellosolve (1). At this time the shortage of oil-based feedstock meant that the solvent (which is mainly used as paint thinner) was in short supply. It was therefore decided to use a 60:40 mixture of cellosolve with the more readily available n-butyl alcohol (C_4H_9OH). In all some 1500 litres of mixed solvent were needed for each pump. The cleaning and decontamination facility was fitted with the inner tank which is capable of accepting the pump. It was equipped with shaped blocks to reduce the 'dead volume' inside the bellmouth of the pump. The system was purged out with nitrogen and the solvent mixture slowly added to the tank in stepwise additions over a period of ten to twelve hours until it was filled to the pump seal joint level. Hydrogen levels and temperatures were monitored to control the process together with solvent samples which were analysed for sodium content. The tank contents were sparged with nitrogen to promote mixing after some twenty-four hours. The samples indicated that ~2 kg of sodium had been removed from the primary pump and ~4 kg sodium from the secondary pump.

When the pumps were drained, dried and examined in air it was found that all the free surfaces had been adequately cleaned but that crevices, particularly those which had trapped pockets of sodium behind them, had not been reached by the solvent. It was necessary to carry out further local cleaning as dismantling proceeded.

When the next primary pump (Fig 4) was cleaned the sodium cleaning and decontamination facility was available for use in its design mode and water droplet dispersion in nitrogen method was employed (known at Dounreay as 'Scotch Mist'). The temperature was controlled using the admission rate of steam to about $40^{\circ}C$ and hydrogen monitoring determined the end point of the reaction. The condensed water

from the process was collected in a vessel below the pump which could be sampled for sodium concentration.

At the end of the process the pump was washed down with water sprays and then air dried. Examination of the pump afterwards showed that the cleaning pattern was very similar to that found with the alcohol immersion, ie clean surfaces but little or no penetration of crevices with sodium hold up beyond. A particular problem was found with the large (1 m dia) piston ring seals used to locate the primary pump body into its flow duct. These have a number of small clearances (less than 0.075 mm) in the complex vertical and horizontal sealing faces but the space behind and within the rings allows a hold up of up to 500 g of sodium. In order to remove the rings it is necessary to pull them off centre sufficiently to extract a locking pin. The bonding effect of the residual sodium prevented this being done and moderately drastic cleaning methods such as immersion in demineralised water at $60^{\circ}C$ for 24 hours had to be employed. The difficulty with this kind of geometry is that the cleaning fluid enters the crevice and reaches the bulk sodium when hydrogen is evolved which pressurises the spaces and blows the reactant back through the crevice. Any further reaction is not possible until the hydrogen has dispersed when a similar sequence is repeated.

Charge Machine

During the early stages of commissioning the Charge Machine was cleaned several times for adjustment and maintenance using the mist technique. This assembly contains a recirculatory ball screw mechanism and number of roller bearings. When cleaning operations were first carried out the sodium removal was adequate except for the ball screw channels and the bearing internals. It was found that immediate application of a commercial dewatering fluid to the roller bearings enabled them to be re-used without further treatment as the problem was one of aqueous corrosion rather than reaction product build up. The ball screw assembly required complete strip-down each time to achieve an acceptable degree of cleaning.

Primary and Secondary Cold Trap Baskets

So far there has been one basket change in the primary cold trap and four changes in the secondary cold trap. The primary basket change took place before there had been any substantial running at power and radioactivity levels were low. It was therefore possible to transfer the basket using a bagging system (fibreglass/silicone), and to break down the basket for examination and disposal using an inert gas filled PVC bag for access. The secondary baskets were dealt with in a similar fashion. One difficulty with breaking down the baskets in air is that the contents which are mainly sodium monoxide (Na_2O) and sodium hydride (NaH) tend to be in a crystalline matrix which contains at least 50% sodium metal distributed throughout. If this is disturbed mechanically the combined effect of an extended surface and the presence of less stable oxides such as sodium peroxide (Na_2O_2) can lead to rapid oxidation and ignition of the free sodium.

This effect also occurs when steam lancing or water mist methods are applied in air. There is however no possibility of recovery of the used knitmesh basket packs and this technique has been used in the DFR decontamination booth which is designed to deal with sodium disposal by burning. The exhaust system incorporates a water scrubbing plant together with radioactivity and smoke monitoring instru-

ments. Future disposal of primary baskets will be carried out by water mist cleaning in the inert atmosphere of the PFR decontamination plant.

Steam Generator Units

It has been necessary to remove several of the steam generator unit tube bundles in order to inspect, modify or repair them. The design and construction of these items is such that there are some regions of sodium hold up and large numbers of crevices and screwed joints. So far a complete clean-up of a tube bundle has not been carried out but in order to facilitate in situ machining of a stainless steel tube plate (2) extensive part cleaning was undertaken. A sketch of a superheater bundle is shown in Fig 5 and the area cleaned with the outer annulus between the tube plate and the upper grid plate. The method used was to withdraw the tube bundle into a triple layer flexible bag consisting of an inner wrapper of polyethylene, a polyethylene inner bag and an outer PVC welded bag. Polyethylene has good permeability properties but PVC is superior in mechanical strength etc.

When the bundle was fitted into its support frame the upper portion of the bag was peeled back to the first grid plate level where a second steel securing band had been fitted. This exposed the outer rings of tubes to the atmosphere and the moist air reacted with the residual sodium film to form a layer of sodium hydroxide solution on the surfaces. This was reduced to as thin a layer as possible by wiping with lint-free cloths to prevent caustic liquor running down through the grid plates (0.17 mm radial gap). The region was then flooded with carbon dioxide to convert the caustic soda to sodium carbonate which formed a reasonable temporary seal between the tubes and grid plates.

The bundle in its support frame was then laid in a horizontal position (Fig 6) and tilted further to ensure that drainage was towards the tube plate end. The whole region except for the grid plate was then drenched in butyl cellosolve using a spray gun until all visible reaction had ceased. Further washes with progressive dilutions of the alcohol with demineralised water were then carried out until at the fifth stage 100% water washing was employed. After hot air drying the bundle was returned to the vertical and the segment to be worked on was left exposed whilst the remainder was sealed off using polyethylene sheet, and acrylic plastic divider panels. The grid-plate/tube gaps in the open portion were sealed off with shaped silicone rubber mats inserted between the rows of tubes. Using this technique of part cleaning it was possible to machine cracks out of the tube plate whilst avoiding the necessity of complete bundle cleaning. Although a thin layer of oxide was found on the surface of the sodium within the storage bag it was possible to return the bundle to service without further treatment.

A different type of cleaning operation was carried out on an evaporator tube bundle, fabricated from $2\frac{1}{4}$ Cr 1 Mo ferritic steel. In this case sodium had entered the water side due to a small leak in a tube to tube sheet weld (3). A sodium-water reaction had then taken place within the bundle. Although no significant damage was apparent the reaction products had effectively blocked 80 tubes out of the 498 in the bundle. The plugs extended over much of the tube length and were formed of mixtures of sodium oxide and sodium hydroxide with free elemental sodium distributed throughout. In some tubes there were 'slugs' of pure sodium present. Attempts to drill out the reaction product and sodium plugs were made but were unsuccessful. The bundle was therefore removed from its shell using a bagging technique, fitted into a metal container and the sodium-side blanketed with argon. The bundle was then laid down in a horizontal position and with the closure head removed the ends of the tubes were accessible from a platform provided at the working level. (Fig 7).

The actual tube cleaning process was carried out using a system of high pressure water jets. A flexible stainless steel lance fitted with fine cutting jet nozzles was fed from a water pump developing 65 MPa. The lance was fed into the tube entries. The cutting action broke up the deposits and the copious flow of water dissolved and flushed out the sodium oxide and hydroxide without notable temperature excursions. When the jet reached the pure sodium slugs a more vigorous reaction was detectable but these were of very short duration and the flow of water was sufficient to prevent any significant temperature rises.

After this treatment the bores of the tubes were examined using intrascopes and a television camera capable of being drawn round the bends of the U-tubes. Although some pitting was present the tubes were in reasonable condition but much of the protective magnetite layer had been removed. A chemical clean of the tubes using citric acid was then applied followed by a sodium nitrite passivation using impregnated absorbent 'moles' fired round the tubes with nitrogen gas. The bundle was returned to service following a hydraulic pressure test and has since accumulated many thousands of hours of operating time. During this period a small crop of six leaks occurred, necessitating three separate removals of the closure head: on each occasion repair was accomplished successfully.

Control Rod Magnet Faces

Another example of the development of a special technique to deal with a particular problem is the method used to remove the build up of sodium deposited from aerosol in the primary circuit blanket gas on to the mating surfaces of the hold-on magnets. This can reach a sufficient thickness to reduce the efficiency of the electromagnets and it is necessary to clean the faces from time to time. The upper component can be withdrawn and is readily cleaned in a portable glove box available for that purpose. The lower face has to be cleaned in situ by remote tools which can extend to the 4 m depth required.

A number of designs of scraper were used with limited effectiveness only and abrasive pads were tried. One of the abrasives tested was a nylon fibre mat impregnated with silicon carbide particles bonded on to the fibres (a material used for domestic cleaning purposes). This tended to clog easily and was not successful in the dry state. It was found however that if it was slightly wetted with butyl cellosolve its ability to remove and retain the sodium and sodium/oxygen/iron compounds which form the deposit was greatly enhanced. The retention capability of the mat for cellosolve was measured and an optimum loading of 50% of this was adopted to prevent spillage of excess alcohol. A hand operated tool incorporating a circular pad of abrasive and an alcohol dispenser was made up to deal with the in situ face cleaning and proved to be effective, non-damaging and rapid. The amount of alcohol available is insufficient to react with the sodium present and the effect is one of combined abrasion, lubrication, reaction and retention of the sodium and reaction products.

The DFR facilities are somewhat similar to those at PFR but on a smaller scale (4). The pit vessel is 0.6 m dia by 4 m deep to suit the size of the components for this reactor. The increased chemical reactivity of NaK alloy and the risk of vigorous reactions between potassium superoxide and organic materials means that alcohol cleaning methods are seldom used and then only for small items.

The normal cleaning procedure used is steam lancing in air but a number of plant items have been dealt with by nitrogen/water vapour or mist methods using spray nozzles or bubbling techniques. As the DFR decommissioning proceeds the whole of the liquid metal circuitry will be cleaned with one or other of the aqueous processes.

The DFR cleaning and decontamination booth was designed to deal with the disposal of the alkali metals by burning. This is especially useful for dealing with items with sizeable metal hold up which are not to be re-used as in the case of the cold trap baskets described previously.

TEST LOOP AND LABORATORY FACILITIES

The UK test loop and laboratory installations which carry out sodium removal employ a variety of methods including alcohol, steam, water mist and vacuum distillation (5, 6, 7). These methods appear to be adequate for the tasks in hand and demonstrate the need to tailor cleaning procedures to suit the particular circumstances prevailing.

Where alcohol cleaning is used the tendency now is to use de-natured ethyl alcohol almost exclusively. The use of methyl, butyl and higher alcohols including glycols has declined. The slightly higher notional flammability risks are offset by the limited temperature excursions due to the lower boiling point. There is also a cost and availability benefit when using ethanol.

FUTURE DEVELOPMENTS

The main interest here is in the development of large-scale facilities to deal with full-scale reactor components. No single method can be applied to every circumstance and there is a need to have a repertoire of cleaning methods available to deal with the range of requirements. The facilities provided at PFR are extremely versatile and with relatively simple modifications can be adapted to give better control of reaction by using an all gas phase aqueous system as an alternative to the mist method. The provision of a simple alcohol washing plant has given a further variant for suitable components. Evaporative methods although requiring a costly initial plant installation would appear to have many advantages in dealing with complex items. This approach is being investigated.

BY

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INTRODUCTION

As part of the fast reactor support programme at Harwell a study of the transport and deposition by flowing sodium of radioactive corrosion and fission products has been mounted. A by-product of this work is the availability of test section specimens contaminated with deposited radioactive nuclides. These specimens are being used to provide test data on chemical reagents which may be selected as potential decontaminant fluids. Currently, the programme uses pipework specimens cut from the Mass Transfer Loop (MTL). This loop is being used in studies of the deposition of activated stainless steel corrosion products. Through time the programme will be extended to include specimens from a projected fission product loop (which will be studying caesium transport and deposition in the first instance), specimens from fuel capsule experiments in materials testing reactors, and specimens from both the decommissioned Dounreay Fast Reactor and the PFR. This report presents the results of the first screening tests of various chemical decontaminating reagents. A short description of the Harwell MTL is also given to define the conditions under which the contaminated pipework is produced.

HARWELL MASS TRANSFER LOOP (MTL)

A schematic diagram of the loop is shown on Fig 8: a few relevant features are reported. During the last run, following which the first specimens of pipework for decontamination were taken, sodium was circulated at temperatures between $585^{\circ}C$ and $370^{\circ}C$. At the high temperature it flowed through a test section containing several corrosion specimens of different materials. In particular, the central specimen was type 316 stainless steel which had been previously irradiated in a fast neutron flux so that it contained the radioactive nuclides $Cr51$, $Mn54$, $Co60$, $Fe59$. During the run the hot flowing sodium leached off significant quantities of these nuclides and deposited them on colder parts of the loop. After traversing the test section the sodium flowed through a 7 m long tube of 17.1 mm bore which was finned on its outer surface and cooled by an air blast to reduce the sodium temperature gradually to $370^{\circ}C$. Following this air-cooled exchanger was an uninterrupted length of pipe also of type 316 stainless steel of 18 mm bore. Several 50 mm lengths were cut from it at the end of the 2280 hr run and used for decontamination tests. The sodium velocity in this pipe during the run was 0.75 m/s giving a Reynolds Number of 3.3×10^4.

DECONTAMINATION METHODS

Each of the pipe specimens taken was first subjected to two methanol washes in succession to remove the sodium. The specimen was then washed with the chosen chemical reagent to be tested as a decontaminant. Finally it was subjected to a 24 hr wash with distilled water. The overall decontaminating procedure therefore consisted of four steps, viz:

1. First methanol wash.

2. Second methanol wash - 2 hr duration.

3. Decontaminating reagent wash - 6 hr duration: two separate tests were carried out at 25°C and 50°C respectively.

4. Final distilled water wash - 24 hr duration.

The radioactivity on the specimen was gamma-counted in a fixed geometry at the start of the procedure and also in between each of the steps listed above. The decontamination achieved over each step, and over the entire procedure, was thus calculated. In this first series of tests only Mn54 and Co60 were identified. The two gamma emissions for Co60 are labelled Co60A and Co60B in the Figures. The decontaminating reagent washing from step 3 were retained and analysed chemically for Fe, Cr and Ni. This gave some data on the relative aggressiveness of each of the reagents towards stainless steel.

In this screening test eight different chemical reagents were tested as decontaminants under step 3. These were:

Citric Acid (10%)
NaOH/K MnO4 mixture (18%/2%)
Ammonium Citrate (10%)
Acetic Acid (90%)
Glycollic Acid (20%)
NaOH (40%)
KOH (40%)
Oxalic Acid (20%)

RESULTS OF DECONTAMINATION TESTS

Figs 9 to 12 show the progress of removal of Co60A and Mn54 during each step of the decontaminating procedure for each of the eight chemical reagents. It can be seen that, generally, the acids appear to be the more effective decontaminating reagents, and that better decontamination is achieved when the reagent wash is used at the higher temperature. Table 1 summarises the overall decontamination factors obtained with the five best reagents. The table shows that Co60 is more easily removed than Mn54: in some cases no significant Co60 count remains after the washing procedure. The comparative difficulty of Mn54 removal could be a consequence of its known capacity to diffuse into stainless steel.

Figs 13 and 14 show the other important aspect of decontaminating procedures, viz the amount of base metal removed: the data are for Fe, Ni and Cr and relate to the amount removed during the decontaminating reagent wash only, ie during step 3 of the decontamination procedure. As expected Fe shows the greatest removal during washing. Table 2 summarises the element removal by the five best reagents, the upper half of the Table showing the amount removed from the contaminated specimens and the lower half the amount removed by the reagent wash from similar pipe specimens but which had never been exposed to sodium. It can be seen that the quantities removed from the contaminated specimens are much greater and the question arises whether metal deposited during the course of the experiment and which has not been shifted by the previous methanol washes, is contributing to the total.

DISCUSSION

The results presented are the first from an expanding programme of decontamination reagent tests. The high decontamination factors in Table 1 show that, although there are clearly other chemicals, duration, and temperature of washings which might prove more effective than those tested so far, Co60 removal should not present too great a problem. Removal of Mn54 is clearly going to prove a more difficult task and may require a more aggressive reagent which will result in removal of larger amounts of the surface metal. The amounts of metal removed by the current treatments (Table 2) are not particularly high. These conclusions must, however, be qualified by the observation that all the work was carried out on specimens with a low level of contamination. Higher levels of contamination may well modify these conclusions.

One interesting aspect of decontamination emerges from Figs 9 - 12. These show a significant drop in count rate due to the two successive methanol washes. These washes can only remove sodium from the surface of the tubular specimens and since radioactive species were also removed, it must be concluded that these must have existed in significant concentration in the sodium (either as soluble species or in the particulate form) or that they were very loosely bound to the specimen surface. This aspect will be further examined in the Harwell programme.

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TABLE 1

SUMMARY OF OVERALL DECONTAMINATION ACHIEVED BY THE FIVE BEST PROCEDURES

| Radioactive Species	Decontaminating Reagent	Citric Acid (10%)		NaOH/KMnO$_4$ (18%/2%)		10% Ammonium Citrate		90% Acetic Acid		20% Glycolic Acid	
		25°C	50°C	25°C	50°C	25°C	50°C	25°C	50°C	25°C	50°C
Co60A	Overall decontamination factors obtained during complete washing procedure, ie Steps 1 to 4	5.82	Total Removal	1.69	2.65	2.30	25.1	11.5	Total Removal	160.6	Total Removal
Co60B		6.82	Total Removal	1.75	2.70	2.28	42.1	14.5	Total Removal	Total Removal	Total Removal
Mn54		2.06	10.07	1.66	2.63	1.44	3.38	2.30	4.83	2.57	4.54

TABLE 2

SUMMARY OF Fe, Ni AND Cr REMOVAL BY DECONTAMINATING REAGENTS DURING STEP 3

Element Removed	Decontaminating Reagent	Citric Acid (10%)		NaOH/KMnO (18%/2%)		10% Ammonium Citrate		90% Acetic Acid		20% Glycolic Acid	
		25°C	50°C	25°C	50°C	25°C	50°C	25°C	50°C	25°C	50°C
Fe (μg/cm^2)	Note: These values quoted for removal from test section specimens	45.17	52.12	16.68	23.63	38.22	38.22	90.34	38.20	97.29	34.40
Ni (μg/cm^2)		10.77	20.50	3.48	3.48	3.82	2.26	17.72	16.68	17.72	3.48
Cr (μg/cm^2)		1.74	2.75	90.34	69.49	1.39	1.81	2.26	0.97	2.81	3.27
Fe (μg/cm^2)	Note: These values quoted for removal from unexposed control specimens	1.49	1.74	6.60	8.34	1.56	2.19	0.94	0.90	2.43	1.81
Ni (μg/cm^2)		<.07	<.10	3.46	3.13	0.17	0.10	0.07	0.07	<.07	0.14
Cr (μg/cm^2)		0.21	0.28	1.04	1.70	0.17	0.28	0.10	0.17	0.28	0.28
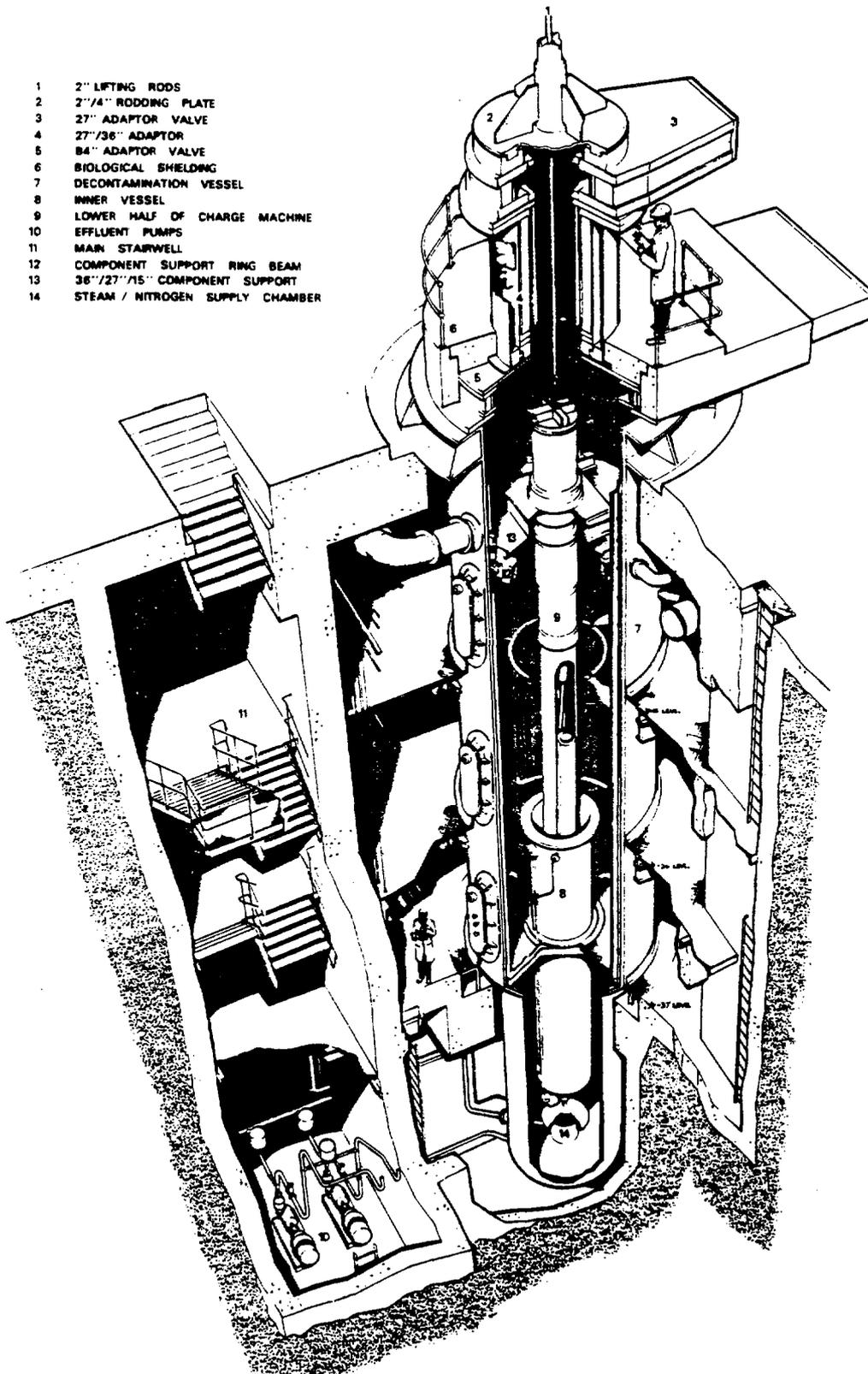
1   2" LIFTING RODS
2   2"/4" RODDING PLATE
3   27" ADAPTOR VALVE
4   27"/36" ADAPTOR
5   84" ADAPTOR VALVE
6   BIOLOGICAL SHIELDING
7   DECONTAMINATION VESSEL
8   INNER VESSEL
9   LOWER HALF OF CHARGE MACHINE
10 EFFLUENT PUMPS
11 MAIN STAIRWELL
12 COMPONENT SUPPORT RING BEAM
13 36"/27"/15" COMPONENT SUPPORT
14 STEAM / NITROGEN SUPPLY CHAMBER

FIG. 1.   PFR Sodium Removal and Decontamination Pit
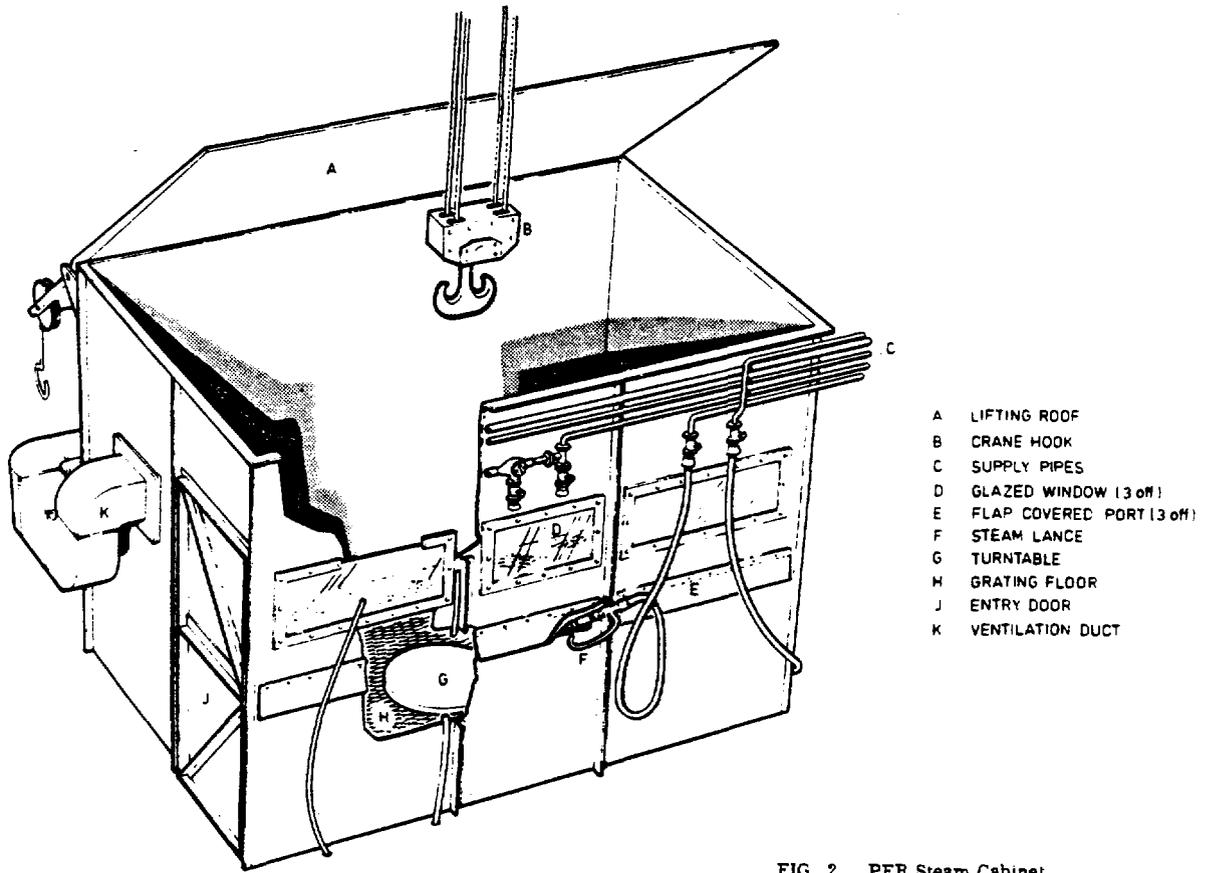
A   LIFTING ROOF
B   CRANE HOOK
C   SUPPLY PIPES
D   GLAZED WINDOW (3 off)
E   FLAP COVERED PORT (3 off)
F   STEAM LANCE
G   TURNTABLE
H   GRATING FLOOR
J   ENTRY DOOR
K   VENTILATION DUCT

FIG. 2.   PFR Steam Cabinet
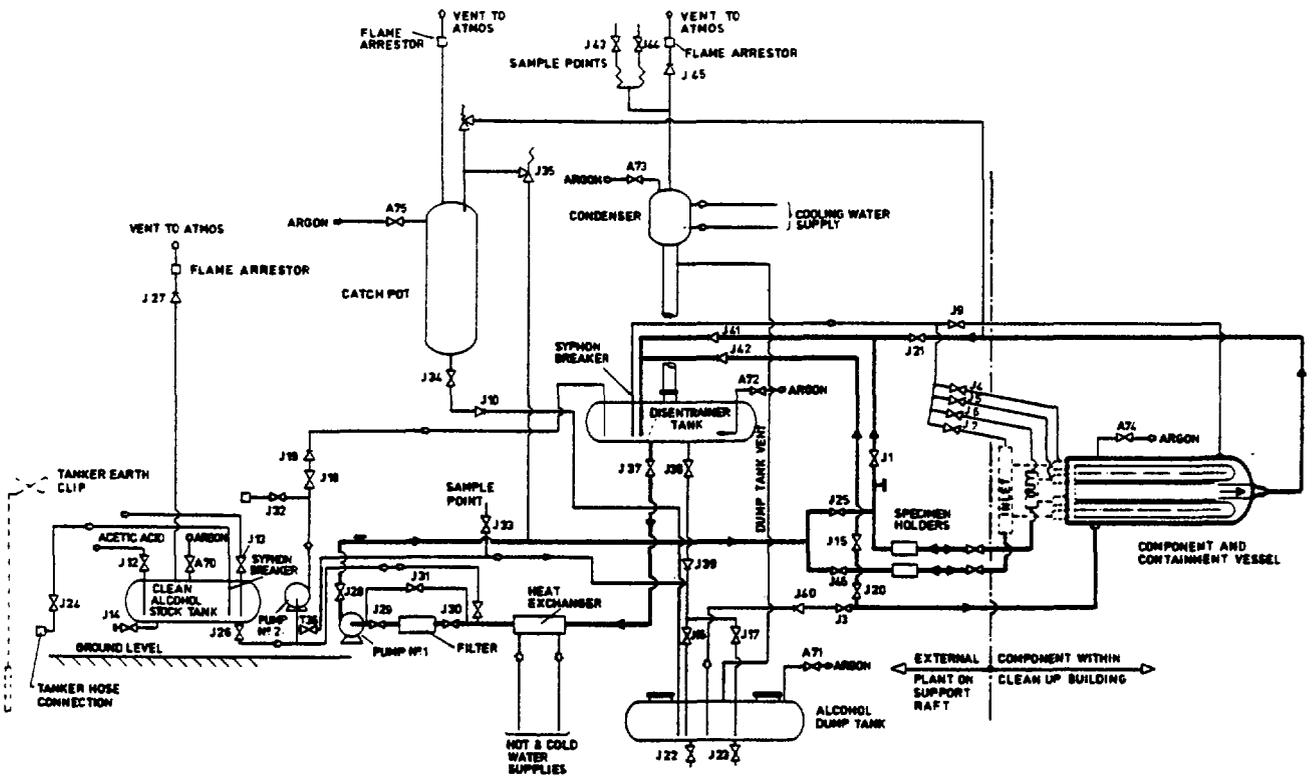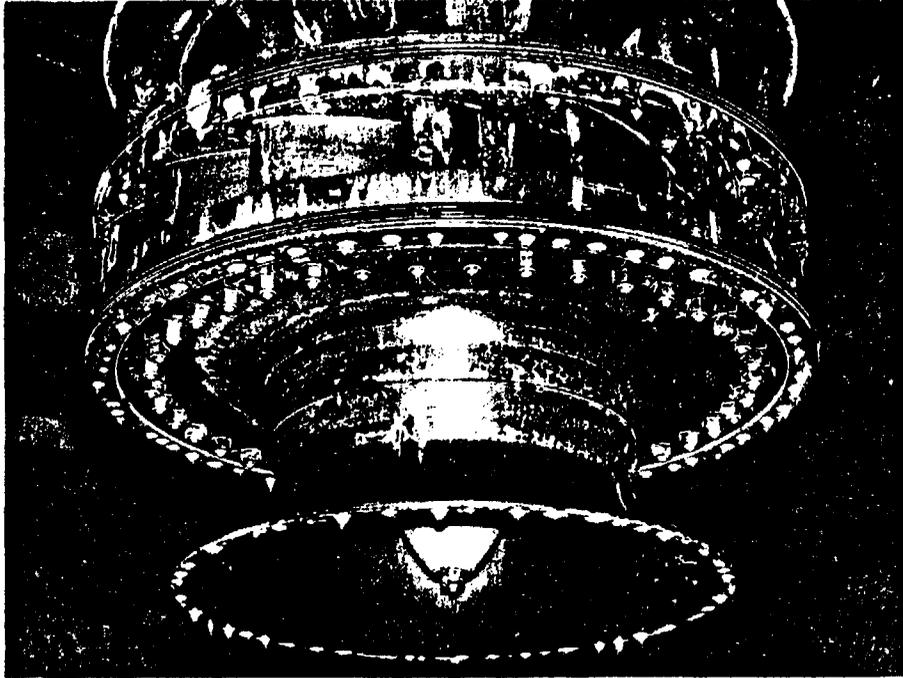
FIG. 3.   Flow Diagram PFR Alcohol Clean Up System

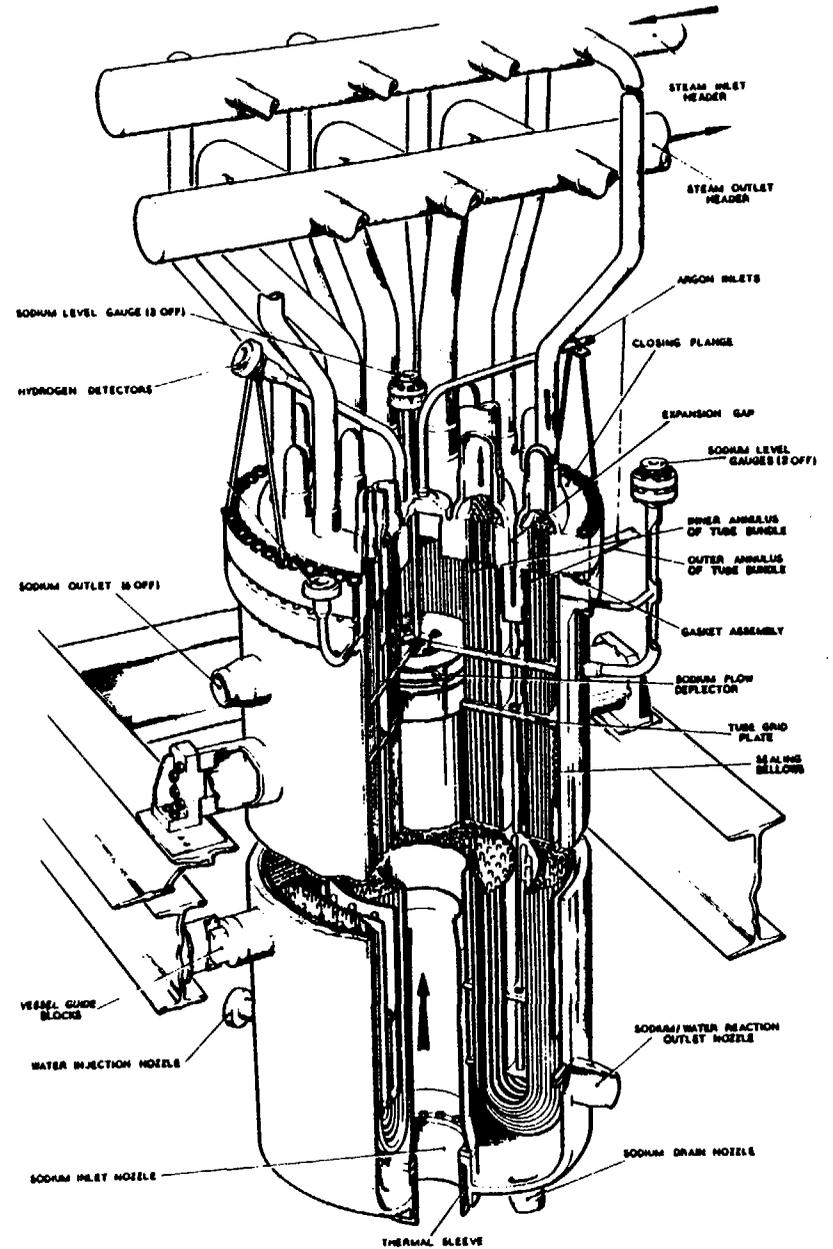FIG. 4. Lower End of PFR Primary Sodium Pump Showing Piston Rings (before cleaning)

FIG 5. PFR Superheater Arrangement

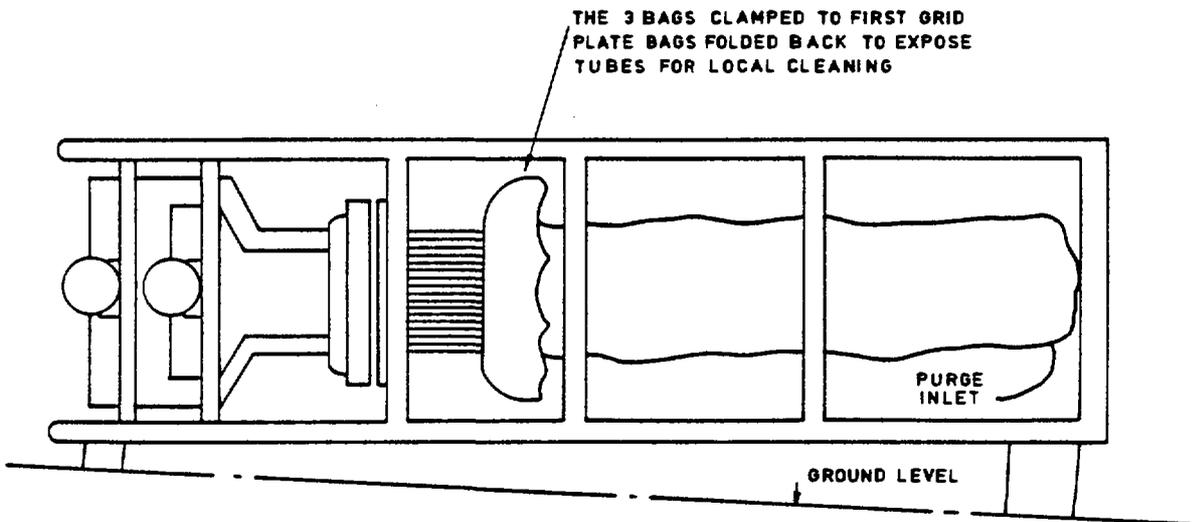FIG. 7.   Water Jet Cleaning of Sodium and Sodium/Water Reaction Products from Tubes in PFR Evaporator

THE 3 BAGS CLAMPED TO FIRST GRID
PLATE BAGS FOLDED BACK TO EXPOSE
TUBES FOR LOCAL CLEANING

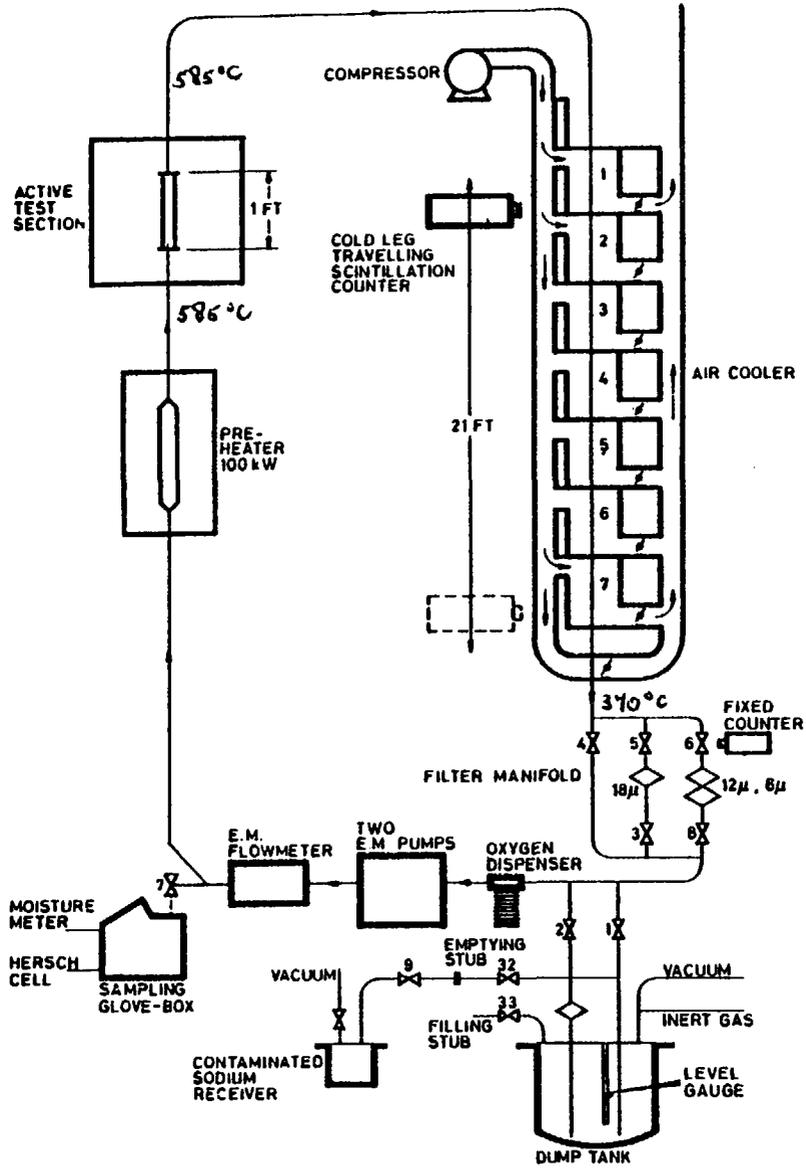PURGE
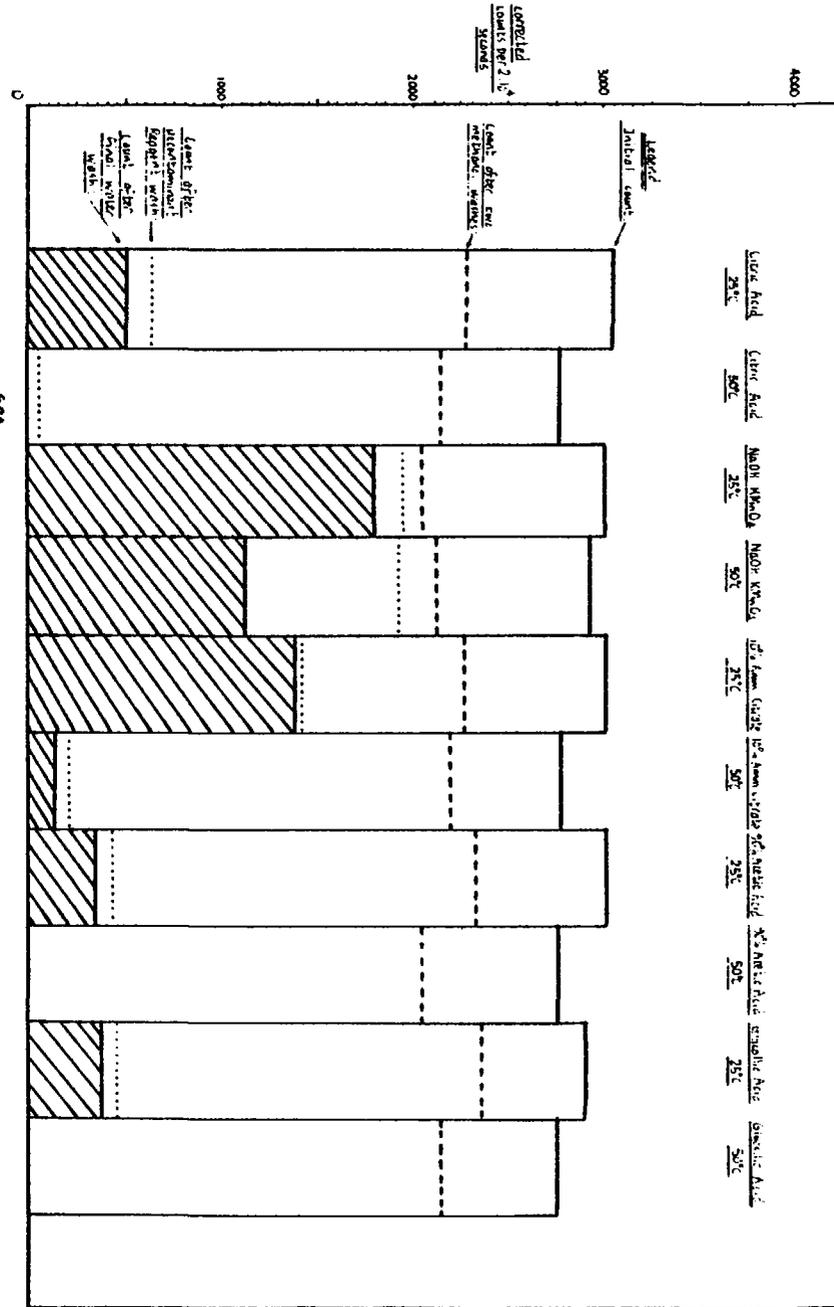INLET

GROUND LEVEL

FIG. 6.   PFR Superheater Tube Bundle Tilted for Cleaning

MASS DEPOSITION RIG
FIGURE 8

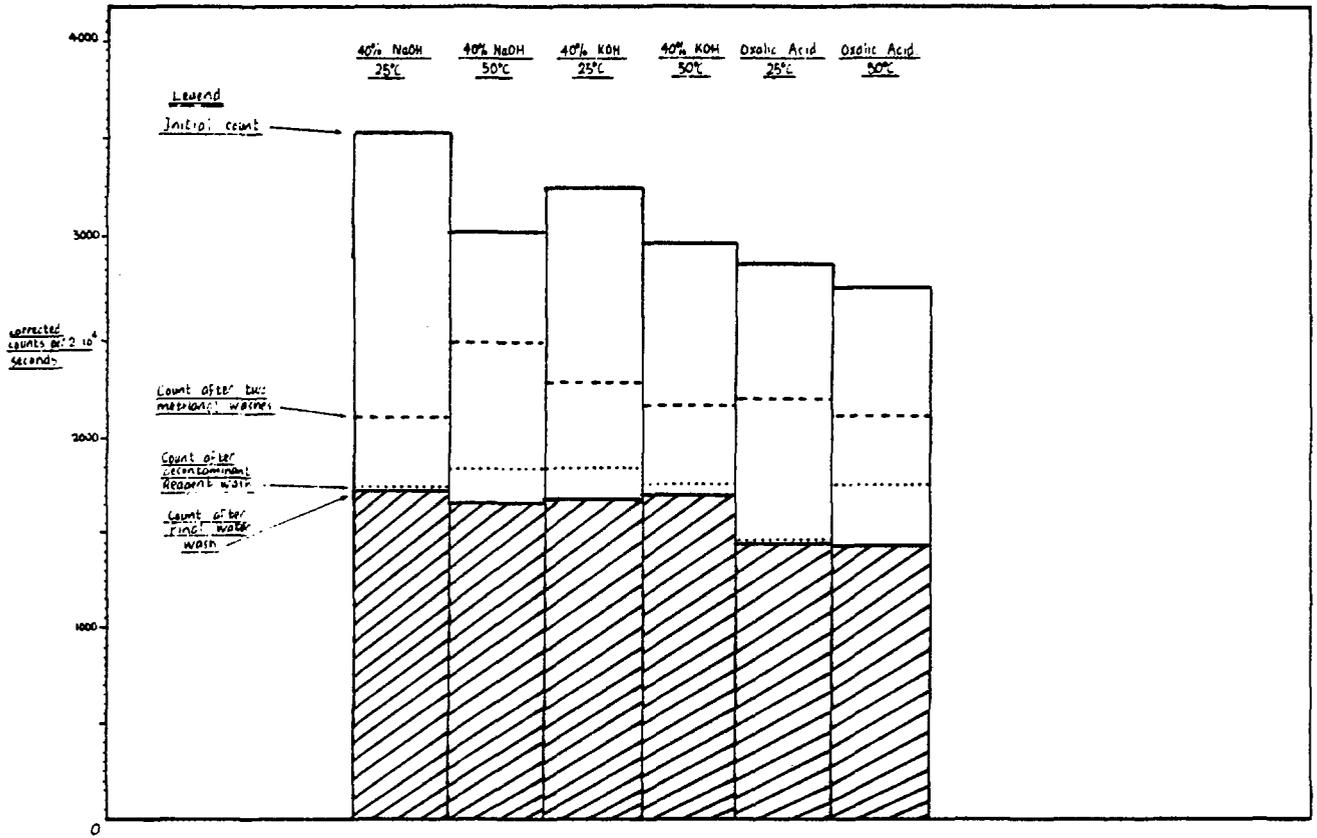FIG. 9. REMOVAL OF ⁶⁰Co FROM TUBULAR SPECIMENS BY WASHING PROCEDURE

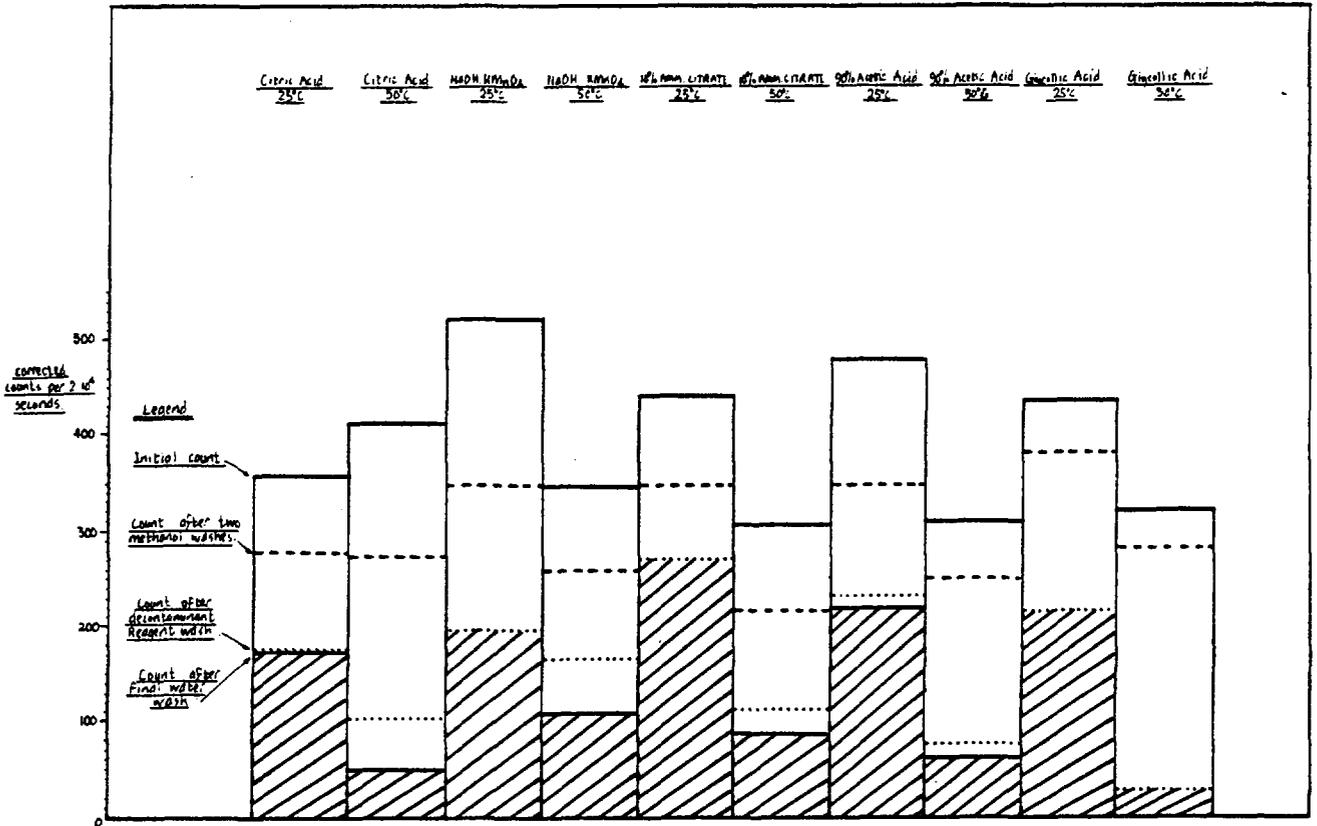FIG 10   REMOVAL OF $^{60A}C_o$ FROM TUBULAR SPECIMENS BY WASHING PROCEDURE

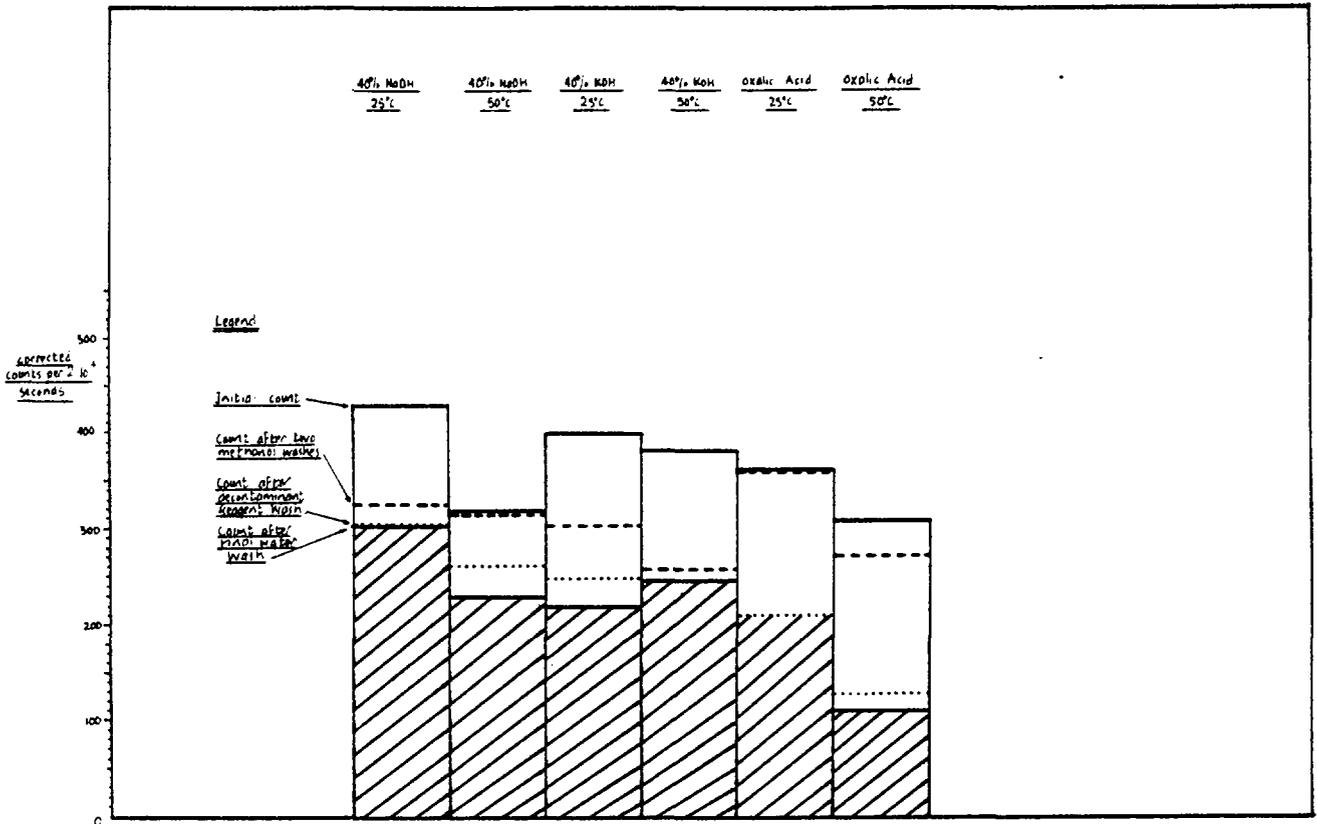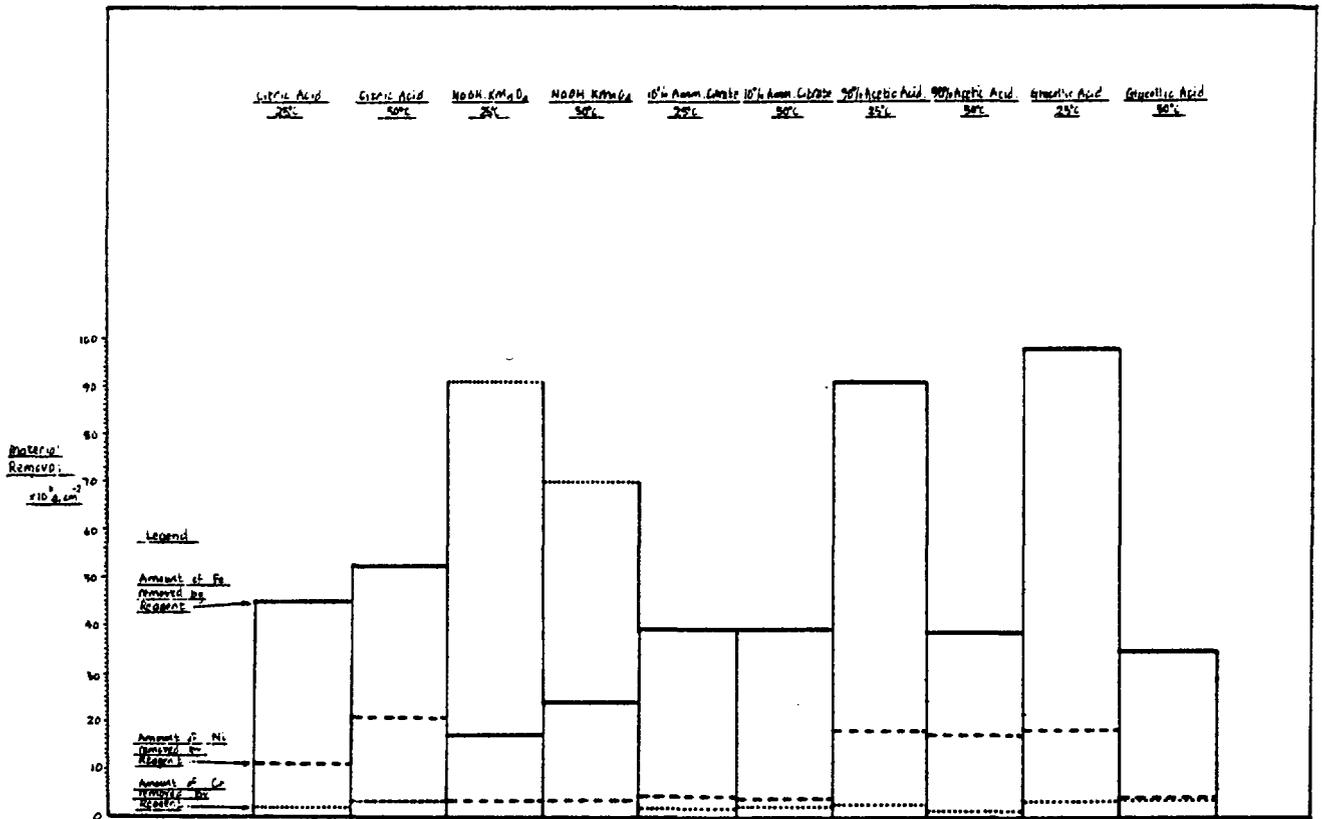FIG 11   REMOVAL OF $^{54}M_n$ FROM TUBULAR SPECIMENS BY WASHING PROCEDURE

Legend

Initial count
Count after two methanol washes
Count after decontaminant reagent wash
Count after final water wash

corrected counts per 2 10 seconds

| 40% NaOH 25°c | 40% NaOH 50°c | 40% KOH 25°c | 40% KOH 50°c | Oxalic Acid 25°c | Oxalic Acid 50°c |

FIG 12   REMOVAL OF ^{54}Mn FROM TUBULAR SPECIMENS BY WASHING PROCEDURE

Material Removed x10^{-2} g. cm^{-2}

Legend

Amount of Fe removed by reagent
Amount of Ni removed by reagent
Amount of Cr removed by reagent

| Citric Acid 25°c | Citric Acid 50°c | NaOH KMnO$_4$ 25°c | NaOH KMnO$_4$ 50°c | 10% Amm. Carb 25°c | 10% Amm. Carb 50°c | 50% Acetic Acid 25°c | 50% Acetic Acid 50°c | Glycollic Acid 25°c | Glycollic Acid 50°c |

FIG 13   REMOVAL OF Fe Cr AND Ni FROM TUBULAR SPECIMENS DURING STEP 3 OF WASHING PROCEDURE

Material Removal $\times 10^{-6}$ g cm^{-2}

100
90
80
70
60
50
40
30
20
10
0

40% NaOH 25°C   40% NaOH 50°C   40% KOH 25°C   40% KOH 50°C   Oxalic Acid 25°C   Oxalic Acid 50°C

Legend

Amount of Fe removed by Reagent

Amount of Ni removed by Reagent

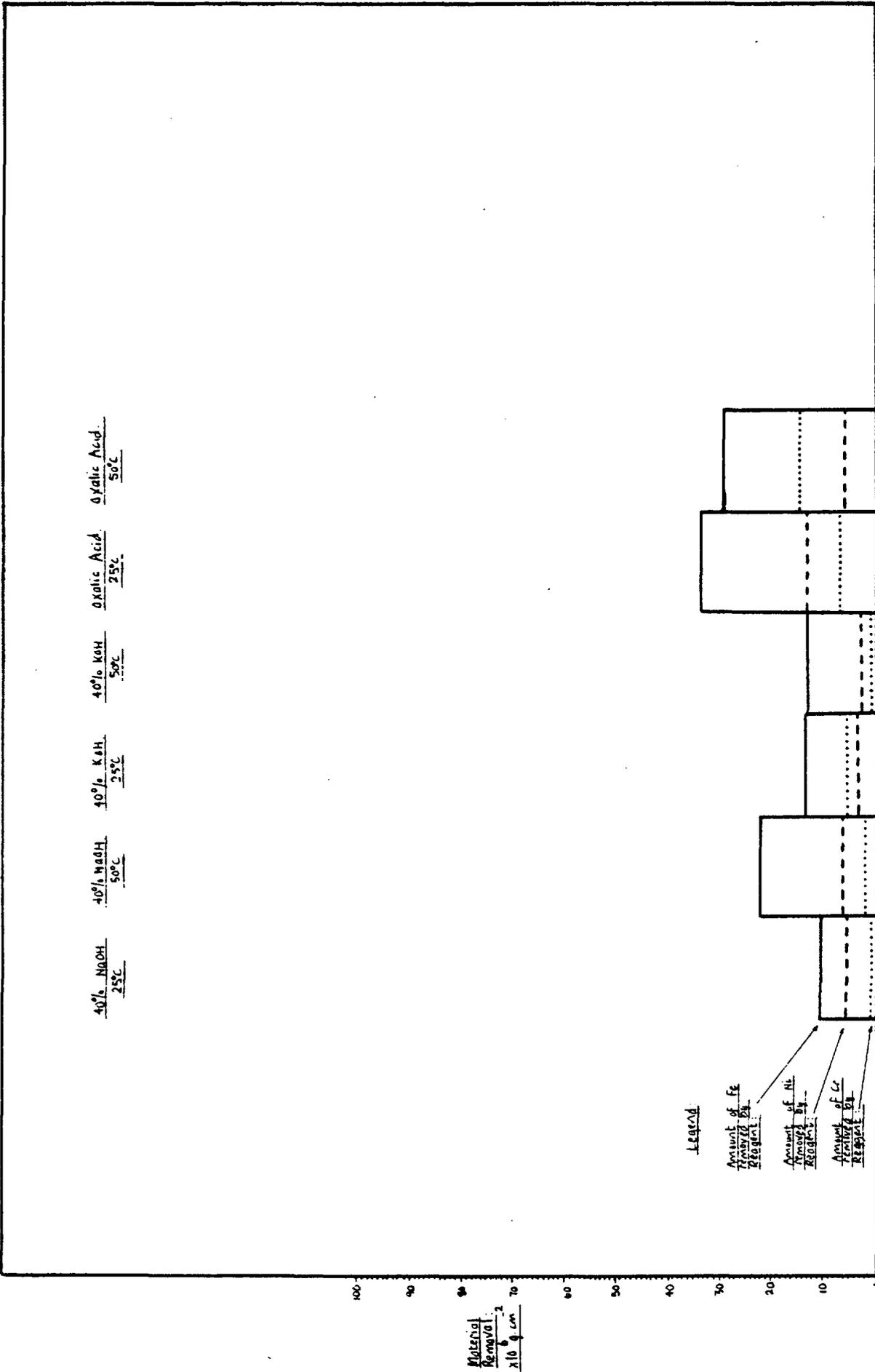Amount of Cr removed by Reagent

FIG 14   REMOVAL OF Fe Cr AND Ni FROM TUBULAR SPECIMENS DURING STEP 3 OF WASHING PROCEDURE