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THE VARIOUS SODIUM PURIFICATION TECHNIQUES

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Abstract

In the framework of sodium waste treatment, the sodium purification phase plays an essential role in the chain of operations leading to the transformation of the active sodium, considered as waste, into a stable sodium salt.

The objectives of the purification operations are :

- To keep a low impurity level, particularly a low concentration in oxygen and hydrogen, in order to allow its transfer to a processing plant, and in order to avoid risks of plugging and/or corrosion in sodium facilities.
- To reduce the sodium activity in order to limit the dose rate close to the facilities, and in order to reduce the activity of the liquid and gaseous effluents.

After a recall of the different kind of impurities that can be present in sodium, and of the different purification methods that could be associated with, the following points are highlighted :

- Oxygen and hydrogen purification needs, and presentation of some selection criteria for a purification unit adapted to a sodium processing plant, as well as 2 cold trap concepts that are in accordance with these criteria : PSICHOS and PIRAMIDE.
- Tritium reduction in a bulk of liquid sodium by swamping, isotopic exchange, or permeation throughout a membrane.
- Caesium trapping on carbonaceous matrix. The main matrices used at present are R.V.C. (Reticulated Vitreous Carbon) and Actitex/Pica products. Tests in the laboratory and on an experimental device have demonstrated the performances of these materials, which are able to reduce sodium activity in Cs^{134} and Cs^{137} to very low values.

The sodium purification processes as regards to the hydrogen, oxygen and caesium, that are aimed at facilitating the subsequent treatment of sodium, are therefore mastered operations. Regarding the operations associated with the reduction of the tritium activity, the methods are in the process of being qualified, or to be qualified.

1. INTRODUCTION

During the shut down and dismantling phase of Liquid Metal Fast Reactors (LMFR), it is necessary to finalise several sodium purification techniques which are essential to its subsequent treatment.

The aims of sodium purification operations are multiple. These techniques are all aimed at promoting the subsequent processing of sodium in adequate facilities; that is to say, to adapt the specifications required by the sodium to the acceptability criteria of the processing plants. Thus, so the sodium purification phase plays an essential role in the chain of operations leading to the transformation of the active sodium, considered as a waste, into a stable sodium salt.

The aims of sodium purification operations are therefore :

- To keep a low impurity level, particularly a low concentration in oxygen and hydrogen, in order to allow its transfer to a processing plant, and in order to avoid risks of plugging and/or corrosion in sodium facilities.
- To reduce the sodium activity in order to limit the dose rate close to the installations, and in order to reduce the activity of the liquid and gaseous effluents.

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After a recalling of the different kind of impurities that can be present in sodium, and of the different purification methods that could be associated with, some of these methods will be highlighted in more details :

- Oxygen and hydrogen purification needs, and presentation of some selection criteria for a purification unit adapted to a sodium processing plant, as well as some cold trap concepts that are in accordance with these criteria.
- Tritium reduction in a bulk of liquid sodium.
- Caesium trapping on carbonaceous materials.

Sodium purification operations can play an important role in the domain of sodium waste processing as they permit adapting or submitting these wastes to the criteria and specifications required by the processing plant. These processes must be designed keeping in mind that, at the end of their service, the purification plants themselves will also be considered as sodium wastes that have to be treated.

2. SODIUM IMPURITIES

Various impurities of different kind : soluble, or insoluble, active, or inactive, are present in the sodium of any LMFR :

- hydrogen and oxygen
- tritium,
- caesium (^{137}Cs),
- metallic impurities as iron, chromium, and some of their active isotopes, coming from their activation under the neutron flux : ^{54}Mn, ^{51}Cr, ^{60}Co, etc.,
- carbonated particles, calcium oxide, etc..

2.1. Hydrogen and oxygen

The hydrogen and oxygen pollution sources, for which sodium is having a large chemical affinity, are of various nature for a sodium to be treated as a waste :

- gas desorption from new structures,
- pollution by inerting gas renewal, or due to air/humidity inlets occurring during operations on the sodium circuits.

The sodium purification as regard to these impurities is based on a crystallisation process in specific unit : cold trap. This process is operating continuously during the whole operating life of LMFR.
In the general case of sodium waste processing, the need for a purification unit will be exposed, and in this case, choice criteria for that unit will be highlighted. Waste processing must be based as far as possible on processes that are existing, well tested and proven, in order to limit the needs for research and development. In this point of view, two cold traps concepts that are answering partly to these selection criteria will be presented.

2.2. Tritium

Several tritium purification processes can be considered, depending on the tritium activity in the sodium :

- For low tritium activity, the basic principle of co-crystallisation of tritium along with hydrogen in tritiated sodium hydride can be considered, with hydrogen injection in the sodium of the circuit (H_2 gas, or NaH crystals). This principle is called « swamping ». Another principle is based on the isotopic exchange of solute tritium with hydrogen from the sodium hydride of a cold trap already loaded.
- For high tritium activity, going along with high hydrogen concentration and a permanent pollution source of these 2 elements, permeation through a membrane in a specific unit is conceivable. This is the particular case of the processing of used cold traps by dissolving its impurities.

These purification methods will be highlighted in the following.

2.3. Caesium

Caesium (^{137}Cs) is a fission product, γ emitter, and is responsible to a large extent of the sodium overall activity, that has for consequence a significant dose rate when in close contact with the sodium facilities.

2.4. Others impurities

The others impurities that can be found in sodium are mainly in the form of particles :

- Metallic impurities as filings, or cuttings, issued from operations on the sodium circuits (cutting, welding, components change, or replacement, etc.). Particles size of more than 100 µm is possible.
- Metallic impurities issued from the corrosion of the structure, and in particular the fuel claddings, iron, chromium, nickel, etc., as well as some of their isotopes generated by the activation under the neutron flux, ^{54}Mn, ^{51}Cr, ^{60}Co, etc. The particles form of these impurities is not clearly defined. According to literature review, the form of these impurities in sodium may be either colloidal, particles, or solute component. The typical size of these metallic components is in any case lower than 10 µm. These components tend to deposit on cold surfaces, as for instance the cold surface of the intermediate heat exchanger.
- Carbonated particles. This impurity is issued from the incidental ingress of oil in the sodium, that is cracked at high temperature. Only graphite must remain after a sufficient stay in sodium. It presents a particle size around 1 µm, with the tendency to stick to the surfaces, and to agglomerate. Such cluster formed in the sodium, or on the surface, shows a particles size much larger.
- Different oxides as calcium oxide, initially present in sodium. These particles, which size of more than 100 µm is not uncommon, are put in suspension by stirring.

The purification processes for particles impurities are based on filtering, and/or eventually settling, with the help of decreasing mesh filters down to the order of the micrometer (poral filter). The filtration process has been successfully achieved for the disposal of the sodium from the RAPSODIE reactor : it is not highlighted in this paper.

Moreover, there is another activation product that can be found in the sodium, which is issued from the sodium itself : ^{22}Na, showing a relatively short half life of 2.6 years. The only purification method is to wait for its natural decay.

3. SODIUM PURITY IN RELATION TO OXYGEN AND HYDROGEN

3.1. Sodium hydride and oxide crystallisation mechanisms

Sodium hydride and oxide (Na_2O, NaH) are crystallising in liquid sodium when cooling, that produces the necessary sodium sur-saturation against hydrogen and oxygen to let the nucleation mechanism happen, and then to allow for crystal growth. Sur-saturation represent the gap between the real impurity concentration in sodium, either oxygen or hydrogen, and the equilibrium concentration corresponding to the solubility of that impurity at the sodium temperature. The nucleation mechanism is occurring either on a support, as metallic surface provided by a steel-wool packing, or as crystal particles, or either in the bulk of liquid sodium. These two mechanisms are referred to as, respectively, heterogeneous and homogeneous nucleation mechanisms.

In the case of sodium oxide, homogeneous nucleation is difficult to perform : the sur-saturation needed can be counted in tens of degrees, due to nucleation and growth kinetics that are relatively slow. The crystal growth kinetic limiting step would appear to be the diffusion of the oxygen atom in the diffusion boundary layer. In the lack of efficient cooling, turbulence, compact metallic surface in the flow (packing), a high sodium sur-saturation is consequently maintained.

In the case of sodium hydride, homogeneous nucleation is easier to perform : the sur-saturation needed is in general lower than 20°C, because of nucleation and growth kinetics that are faster. Concerning the crystal growth kinetic, the limiting step would appear to be the integration of the hydrogen atom in the crystalline network mesh, which is itself relatively rapid. Diffusion coefficient is much larger for hydrogen than for oxygen.

Consequently, in the case of the cooling of a sodium tank, oxygen sur-saturation will be maintained, when hydrogen will not create any sur-saturation. This oxygen sur-saturation presents a real risk of sodium oxide deposits, leading to plugging, in the circuits located between the sodium tank and sodium processing unit, if the following conditions are fulfilled : local turbulence, filling of a facility insufficiently pre-heated, narrow and cooled section such as filters, flow-meters, valves, etc. Filters, that in addition offer a large metallic surface for nucleation, are then particularly exposed to the risk of plugging by sodium oxide. This has a direct consequence on the availability of the sodium processing facilities. A careful control of the circuits temperatures must be performed to lower that risk of plugging.

In order to offer a permanent availability of the facilities, the most adapted solution is the use of a purification unit, that allows for lowering the oxygen sur-saturation, and control the liquid sodium quality in accordance with the subsequent sodium processing units [5].
In the particular point of view of the sodium disposal facilities, this purification unit must be selected thanks to some main guidelines.

3.2. Guidelines for the selection of a purification unit

In the general framework of treatment of nuclear waste, the practice is to limit to the maximum the volume of effluents and waste produced by treatment operations. The sodium purification unit, which constitutes one step in the processing chain leading from active sodium to a stable sodium salt, becomes itself a waste which must be treated in turn.
In order to proceed to the design choices for a purification unit adapted for this practice, and compatible with waste limitation, the following guidelines can be emphasised.

1- A large capacity, in order to limit the number of traps to be used, as well as the number of replacement operation.
The optimal solution consists in adapting the trap's capacity to the plant's service life, which limits the number of wastes to one cold trap, and limits the costs associated with the operation of this purification function (manufacture of traps, replacements, handling and processing as waste).

2- A device whose operation can be adapted to the type of pollution encountered, from low concentrations to strong pollutions.
A cooler with variable performances which enables this condition to be fulfilled. In addition, a cold trap equipped with such a cooler, and operated with adapted operating rules, makes it possible to guarantee a high retention capacity, while reducing the risks of early clogging. It also guarantees a high purification efficiency, when associated with an isothermal zone, and therefore a maximum purification speed for a given output.

3- A design that facilitate subsequent processing operations.
A system of cooling by air is, in this context, simpler than a system of cooling by organic liquid and liquid metal thermal joint as the sodium-potassium alloy (NaK), to which are added the difficulties linked to draining and processing of NaK. Moreover, the operation of the trap is more complex, particularly due to the risk of cracking of the organic liquid beyond a certain temperature (280°C for the oil used at SUPERPHENIX). The construction costs are also very different.

4- Specific trapping zones, in order to separate the impurities, oxide and hydride, into two distinct zones.

For subsequent processing of the trap, a physical separation of these two zones can be conceived, in order to separately apply a destruction treatment specific to oxide, which is not very active, and a treatment for hydride, which is greatly tritiated.

It must also be considered that for a waste processing plant, the choice is directed initially towards proven processing operations.

Following the experimental feedback obtained from the operation of the PHENIX and SUPERPHENIX reactor cold traps, and also following the improvement in knowledge of the basic mechanisms of the crystallisation of sodium oxide and hydride, two cold trap concepts of large capacity, and with modular cooling, were developed and qualified at the end of the 1980's, with the initial aim of fitting fast reactor secondary cooling system purification circuits :

- PSICHOS, sodium hydride trapping in cooling zone, and sodium oxide trapping in a packed isothermal zone.
- PIRAMIDE, trapping on a packing of modular design.

The PSICHOS concept was recently brought up to date by the replacement of 4 SUPERPHENIX secondary cold traps by traps constructed on this principle in 1997.

The PIRAMIDE concept was developed for application to the secondary cooling systems of the European Fast Reactor (EFR) project-reactor.

3.3. The PSICHOS concept

The initials of the name of this cold trap concept stands for : « Piège Séparant les Impuretés par Cristallisation de l'Hydrure et de l'Oxyde de Sodium » (trap separating impurities by the crystallisation of hydride and sodium oxide). This concept, developed with the aim of replacing the SUPERPHENIX secondary cold traps with optimised traps, as regards operating and capacity, benefits from a certain number of known facts coming from crystallisation mechanisms knowledge and operating feedback (Figure 1) :

1- A 3 modules cooling zone, using an organic liquid circuit and a sodium-potassium alloy (NaK) thermal joint, which is liquid under normal operating temperatures, in order to obtain a sufficient thermal flux on the wall for hydrogen trapping. The hydrogen capacity is not limited by a passage through packing, therefore the risk of clogging is very low.

2- An isothermal zone, located in series with the cooling zone. It supports two concentric packing pads, presenting two different densities which increase in the direction of the sodium flow, in order to promote a homogeneous distribution of sodium oxide, and a maximum retention capacity. This capacity is, indeed, directly proportional to the packing inlet section.

This type of trap has recently been brought up to date by the replacement of 4 SUPERPHENIX secondary cold traps, by units constructed on this principle. The capacity of these traps has been estimated, per trap, at a minimum of 340 L of NaH on the cooling zone wall, and 110 L of Na_2O in the packing. They therefore present, at the minimum, a double capacity compared to the traps formerly used. Indeed, a capacity of approximately 150 L of sodium oxide and hydride after 320 Equivalent Full Power Day was estimated by pollution mass balances for each of the 4 secondary cold traps at the time of their replacement (31st December, 1996).

3.4. The PIRAMIDE concept

This large capacity trap concept was initially developed within the scope of the European Fast Reactor (EFR) project, for which, taking into account ferritic steel, which is more permeable to hydrogen than austenitic steel, used for steam generator tubes, the question of a large capacity was primordial (Figure 2).

The principle of this trap rests on a modular packing design, distribution by plates and arrangement in baffles, and a modular cooler with 3 air cooling zones distributed over the whole trap surface area. This concept enables a large sodium oxide and hydride retention capacity to be provided, without the risk of total plugging of the trap, due to the arrangement in baffles of the packing plates : the sodium always flows in the direction of the minimum pressure drop, and when a packing module clogs, the sodium bypasses it and fills the next module.

The system of cooling by air allows to avoid the constraints associated with the operation of an organic liquid cooling system, and the handling of NaK. Moreover, it presents a much simpler

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manufacturing design, due to the absence of coiling in a jacket, and by its modular design packing. This has a significant effect on the cost of its manufacture and dismantling.

3.5. Conclusion

These two cold trap concepts fulfil the needs of the operator of a sodium destruction facility in relation to the significant retention capacity and the flexibility of operating. PSICHOS has the advantage of separating the oxide from the hydride into two distinct trapping zones, whilst PIRAMIDE is of a simpler design.

Each of these traps can be implemented on a mobile purification unit, of which the principle is largely used on sodium test loops.

4. REDUCTION IN TRITIUM ACTIVITY

Primary and secondary sodium from LMFR show relatively low levels of tritium activity : hundreds to thousands of becquerels per gram of sodium. To reduce this activity to a lower level, 2 methods are conceivable :

- Swamping, by injection of hydrogen, and crystallisation of tritiated hydrogen in a cold trap.
- Isotopic exchange by circulating in a cold trap loaded in sodium hydride.

Indeed, tritium level never reach the sur-saturation level at the operating temperature of the liquid sodium, so that it could form sodium tritide (NaT) by itself. These two purification methods are then based on the known tritium trapping mechanisms : co-crystallisation with the hydrogen, and isotopic exchange of solute tritium with sodium hydride.

For higher level of both hydrogen and tritium in the sodium, and if that level is kept constant, as it is the case for the processing of a cold trap with the PRIAM process [4], where the pollution source is the dissolution of sodium hydride, it is conceivable to permeate the hydrogen along with the tritium through a diffusion membrane by vacuum pumping.

4.1. Swamping

A pre-dimensioning study of the purification of the tritium content of the 3500 tons of primary sodium of the SUPERPHENIX reactor shows that it takes 16 days to reduce the activity by half, with a cold trap having a purification efficiency close to unity, and having a sodium flow rate of 10 m^3/h. The hydrogen flux injected is in the order of 8 L/hour, corresponding to 3 m^3 in the standard temperature and pressure. Sodium hydride volume generated in the cold trap by this operation is 4.7 L. The purification law is an exponential decreasing. So to reduce again the tritium level by half, the same period and the same quantity of hydrogen are needed. The hydrogen pollution could be either gaseous (H_2), or solid (NaH).

4.2. isotopic exchange

Circulating in a cold trap loaded with sodium hydride could reduce the solute tritium activity in the liquid sodium by isotopic exchange with the hydrogen present in a crystalline form. That isotopic effect is not accurately known and measured. According to the literature, it may represent as much as 10 to 20% of the trapping of tritium by co-crystallisation with hydrogen in normal operating conditions. An experiment is actually in progress in a sodium loop of Cadarache to measure this isotopic effect. The experiment consists of measuring the evolution with time of the hydrogen and deuterium, used to simulate tritium behaviour, concentrations when circulating in a cold trap loaded in pure sodium hydride. The cold trap temperature is set to equalise the combined hydrogen and deuterium saturation, so as to cancel the trapping by the co-crystallisation effect. Results are expected by the end of 1997.

4.3. Permeation

The permeation phenomenon is widely used for the sodium-water detection in LMFR. It is make up of a permeation membrane in Nickel, followed with high vacuum pump and a mass spectrometer. The permeation unit is based on the same principle, but, in order to maximise the hydrogen flux throughout the membrane, it tends to favour :

- larger surface of the membrane,
- lower thickness of the membrane,
- highest hydrogen partial pressure gap between liquid sodium and vacuum,

An operating temperature of that unit as high as possible contribute to maximising the flux.

A modular permeator composed of n cells can be envisaged, each cells developing an established surface. Thus, in order to act on the permeator surface, it is sufficient to act on the number of cells.

A pre-dimensioning study of a prototype permeator showing an area of 0.53 m^2, and a thickness of 50 μm, operating at 550°C, with a level of vacuum of 100 Pa, and an hydrogen concentration around 50 ppm, has enabled the flux to be assessed at 2.3 10^{-4} gram(H)/s, corresponding to 9 STPL/hour. This flux can be increased by a factor 2 by the use of twice as many cells.

Manufacture of one prototype permeator has been achieved. 3 others prototypes showing different surface and thickness are under way. It should allow qualification tests of the permeator on the ECRIN sodium loop in Cadarache to be carried out, as well as the correct functioning of the permeator-pumping unit chain to be verified, and the dimensioning rules to be qualified.

4.4. Conclusion

Tritium activity reduction methods remain essentially to be qualified.
However, in all cases, the use of one method for the reduction of tritium activity depends essentially of the authorisations of tritium release delivered to the sodium processing facilities.

5. REDUCTION OF CAESIUM ACTIVITY

5.1. Caesium

In LMFR, one of the major radio-isotope in the coolant (liquid sodium) is the caesium. Indeed, concerning the corrosion products, which are responsible for the contamination of main components, their solubility remains low in sodium, thus their participation to the level of activity in the coolant. Caesium is a fission product, released in the sodium mainly when fuel cladding failure occurs. However, even in absence of fuel cladding failure, it is present at low concentration due to external pollution of the cladding by fissile material (for instance in SUPERPHENIX sodium). Moreover, it has the particularity of being very soluble in sodium.

Fuel cladding failure can be accidental in reactors, or voluntary, for specific studies as it is the case in the SURA loop from the Nuclear Safety and Protection Institute (IPSN) of CEA. One of the objective of the tests performed in this loop is to simulate the plugging of a fuel assembly, which cause a strong overheating, and the melting of fuel cladding.

The main isotopes of caesium are ^{133}Cs, stable, ^{134}Cs (half life = 2.06 year), ^{136}Cs (half life = 13.16 days), and above all ^{137}Cs whose half life is 30 years. Even at tiny concentration in sodium (10^{-8}g Cs /g of Na), they are responsible of significant level of activity during operation, and even more during dismantling phases.

Being very volatile, caesium can also be found in the cover gas, with a high enrichment factor (100 to 400 in sodium aerosols).

5.2. Trapping on carbonaceous material

Experience has proven that carbon or carbonaceous material (e.g. grease) have the particularity of trapping caesium. This property was used to purify the sodium of some fast reactors, as RAPSODIE, or other experimental facilities. For instance, the sodium from the SURA loop was purified this way, with technical support from STML, in 1988, 1994 and 1997.

5.2.1. Fundamental studies

In order to get a better knowledge of the mechanism involved in the process (adsorption, intercalation...) and to develop a calculation tool for designing new facilities, a PhD. thesis is in progress in CEA.

The trapping process is based on the affinity of alkaline metal (with the exception of sodium) for carbonaceous material, and more specifically graphite. Various type of carbon and conditioning have been tested in reactor or in laboratories, essentially abroad (USA, Germany, former USSR).

5.2.1.1. RVC

The choice of R.V.C. (Reticulated Vitreous Carbon) is mainly due to its physico-chemical characteristics. It is a rigid, nongraphitizable carbon foam with a honeycomb type alveolated structure, of very low apparent density.
R.V.C. has a wide open macroporosity, which enables highly satisfactory load losses to be obtained, as regards sodium flow. Its mechanical resistance to liquid sodium is excellent and its implementation as a trapping device entails no difficulties.
The optimal operating conditions are a trapping temperature between 200°C and 300°C, and a liquid sodium output within the trap of several mm/s.
The effectiveness of this type of trap allows a reduction in ^{137}Cs activity by a factor of 10.

5.2.1.2. Recent developments

The recent developments are linked to the PhD. thesis works by B. REDON at the Liquid Metal Technology Service (STML) of Cadarache.
This PhD. thesis includes a study of the characterisation of adsorbent carbonaceous materials with the Laboratoire d'Electrochimie et de Physico-Chimie des Matériaux et des Interfaces (laboratory of electro-chemistry and physico-chemistry of materials and interfaces) of the National Polytechnic Institute of Grenoble in France, and a study on trapping modelling.
The aim of this work is to optimise the purification operations and to arrive at a dimensioning model which is a function of the properties of the carbonaceous adsorbent (specific surface, porosity), of the fluid flow mode in the column (trap), of the adsorbent/adsorbate equilibrium and of the parameters linked to purification (volume, activity, open or closed circuits...).

- Characterisation studies of carbonaceous material

First, the laboratory work allowed the determination of the texture and structure of the carbonaceous adsorbent (microporosity and specific surface), in order to establish the ^{137}Cs trapping mode. Various complementary techniques, such as helium pycnometry, BET adsorption, transmission electronic microscopy or small angles X-rays scattering, enabled the intimate microstructure of the R.V.C. to be revealed and its theoretical sorption capacity to be estimated.
The study case continued with identification of the carbonaceous adsorbent/alkali metal interactions, thanks to surface analyses (ESCA/XPS) and measurements of sorption heats by differential scanning calorimetry, which allowed quantification of the interaction energies which come into play (comparison with graphite literature data).
A hydrodynamics approach within the trap was obtained using measurement of load losses (validation of the COMITI and RENAUD flow model [6]) and a study by distribution of the residence time (influence of the dispersion phenomena).

- Trapping experiments in dynamic mode

The experimental section at CEA consists in a series of tests on the experimental loop, ELCESNA (CaESium ELimination in relation to NA). This loop is dedicated to the study of ^{137}Cs trapping, in dynamic mode and in active mode; it is operating with primary sodium from RAPSODIE, of which the residual activity in ^{137}Cs allows the activity on the trap and at the trap output to be followed by direct gamma spectrometry on various points of the unit.
The results (breakthrough curve, trapping profile, mass balances) are compared with simulations carried out by the *MODELI* code :
Sodium samplings allow a calibration of these measurements to be performed.

- Trapping experiments in static mode

Complementary tests are carried out with the same sodium, but in static mode in a glove box on small devices. This allows parametric tests but in extreme conditions, in order to define, in particular, the adsorption isotherm (nature of equilibrium) and the saturation capacity of this type of material, by adding inactive caesium to the sodium, until the R.V.C. sample is saturated.

- Other carbonaceous materials

The experience feedback having been acquired solely with R.V.C., a significant part of the tests were performed with this material. Nonetheless, there are other carbonaceous material manufacturers, particularly the SEP (European company for Propulsion) and the PICA company.
After mechanical resistance to liquid sodium tests, experiments were carried out on the ELCESNA loop with carbon in the form of active carbon granulates, provided by the PICA company. These tests have very promising results, regarding the effectiveness of the purification. Special manufacturing is under way in order to further reduce the load losses of this type of material.
These granulates could therefore be implemented as a replacement for R.V.C. on future sodium purification plants. The complementary tests envisaged, particularly with active sodium in a glove box, in addition to the characterisations of these materials, should confirm the similarity between the two products.

5.2.1.3. Modelling

The other aspect of the study is aimed at modelling ^{137}Cs trapping on carbonaceous material : the *MODELI* code. The trapping procedure can be compared to a fixed bed isotherm adsorption operation. Using a pellet/grain model, a mass balance is established on each volume element of the adsorbent column. The adsorption kinetics takes into account all adsorption resistance's (external diffusion, axial dispersion and internal diffusion). A sensitivity study enables evaluation of the limiting process according to the operating conditions selected.
The resolution of the system of differential equations obtained is carried out by rendering discrete the variables, according to the method known as Runge-Kutta-Merson. The results, essentially a function of the data arising from characterisation of the adsorbent and of the adsorption isotherm (concentrations at equilibrium), tell us about the ^{137}Cs concentration at column output and that trapped on the carbon, depending on the time.

5.2.2. Practical application

The French IPSN is studying the behaviour of fuel test devices in accidental conditions. Thus, numerous fuel cladding failure, with fission products release occur in the SURA facility. After several tests, the caesium activity in the coolant, in the storage tank and on the pipes induces health physics problems for the people who are to intervene on the circuits. Purification campaign of sodium are achieved, in order to strongly reduce the dose rate close to the facility.

The latest purification campaign of SURA's sodium was run in July 1997. It was performed by an IPSN team, using IPSN material, in collaboration with STML. STML was more specifically involved in the development of the process, the design of the trap and the analyse of the feedback. In the facility, the weight of sodium is about 15 tons. The trap was designed to be introduced in the circuit at the location of an experimental device. Final treatment of the trap was taken into account from the very beginning of the design phase. Initial activity was around 1.9 10^5 Bq of ^{137}Cs per cm^3 of sodium. After 26 hours of operation with the trap , the activity went down 1.4 10^4 Bq/cm^3, that is a purification factor greater than 13. The total activity trapped is roughly 2.6 10^{12} Bq. Volumetric activity was permanently measured on line, using a gamma spectroscopy device. The evolution of activity decrease is presented on figure 4. This operation resulted in a strong decrease of the global dose rate of the circuit (minimum factor of 3). After the purification, the dose rate on the storage tank was below 100 mRad/h.

The feedback from previous campaign lead us to modify some parameters, such as increasing flow rate in the primary circuit in order to get a better homogenisation of the sodium in the loop, or increasing the pre-heating temperature of an auxiliary circuit to put back in the coolant the caesium deposited on the wall.

People in charge of this facility are satisfied, as for previous operations, of the efficiency of the purification campaign.

The final objective is to propose a continuous trapping set up in an auxiliary circuit, in order to avoid the accumulation of caesium in the sodium and in the circuits.

5.3. Conclusion.

Efficiency of the trapping of caesium present in the primary sodium of LMFR or test facilities on carbonaceous material (RVC or PICA granulates) was demonstrated.

In spite of satisfactory first results, basic studies are driven either in test loop or at the laboratory scale in order to get a better knowledge of material characteristics and of the mechanism involved in the process.

Results from these studies are the bases for the undergoing development of a model. The objective is to have a calculation tool for designing of purification units of caesium from sodium.

6. CONCLUSION

The sodium purification processes as regards to the hydrogen, oxygen and caesium, that are aimed at facilitating the subsequent treatment of sodium in other dismantling units, are therefore mastered operations. Guidelines for the selection of a purification unit of sodium dismantling facilities has been given as well as 2 cold traps concepts that partly fulfil theses guidelines.

Regarding the operations associated with the reduction of the tritium activity, several methods are conceivable depending on the level of activity. However these methods are still in the process of being qualified, or to be qualified. In all cases, the use of one method for the reduction of tritium activity depends essentially on the authorisations of tritium release delivered to the sodium processing facilities.

To limit the biological protections around the sodium disposal facilities, and to limit the activity of the soda produced during the destruction of sodium by reaction with water, purification of the sodium prior to destruction can be achieved on carbonaceous material specific trapping units.

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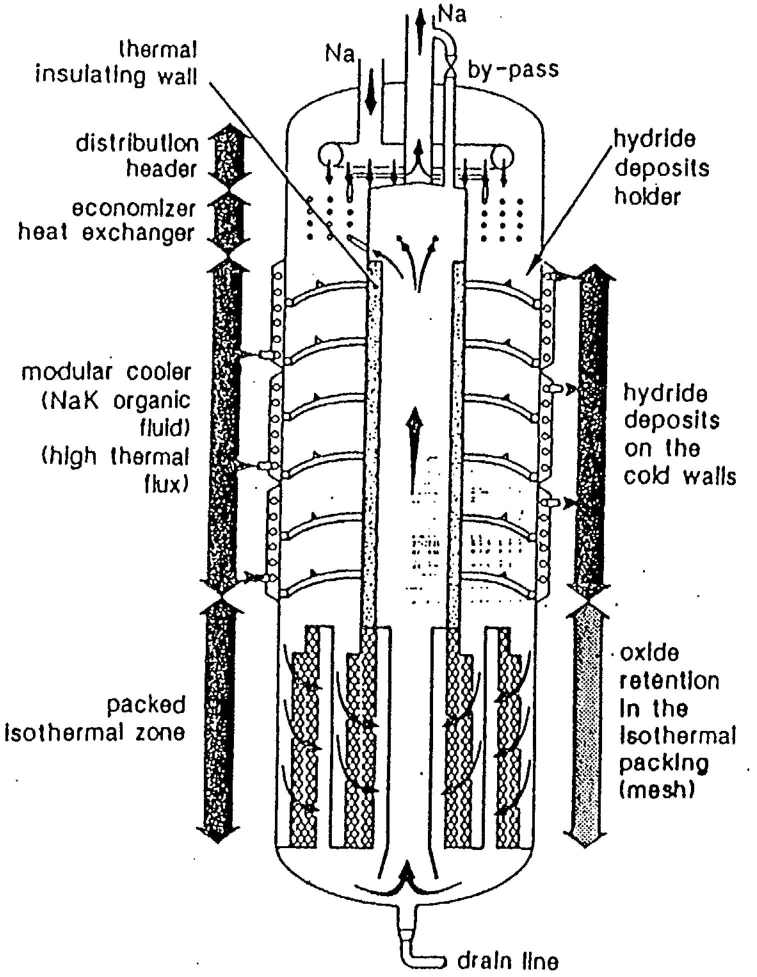

Figure 1 : Scheme of a PSICHOS cold trap

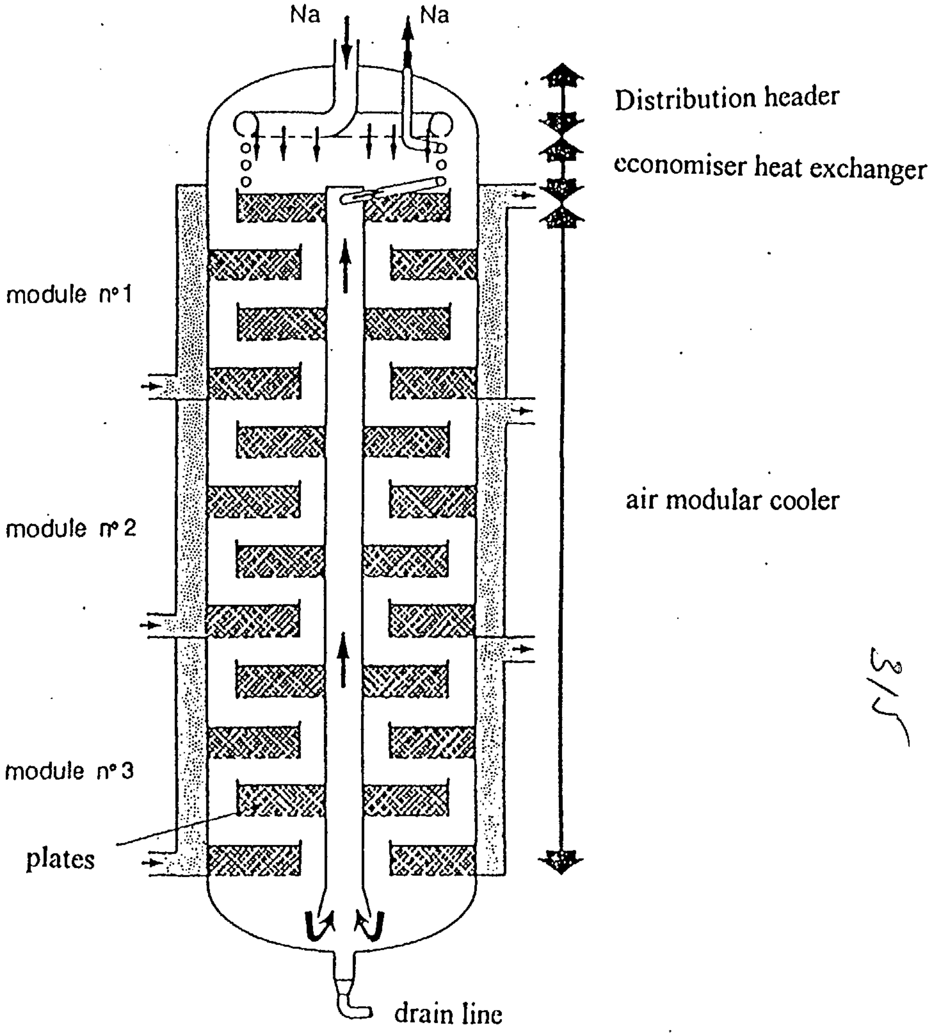

Figure 2 : Scheme of a PIRAMIDE cold trap

Figure 3 : Reticulated Vitreous Carbon (RVC)

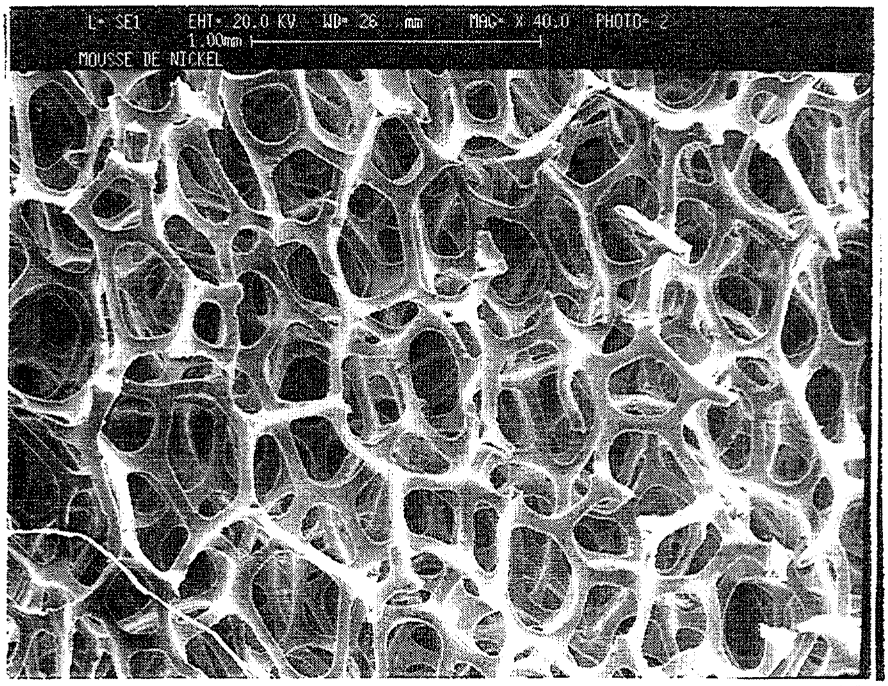

Figure 4 : Evolution with time of the caesium activity during purification campaign at SURA