STATUS OF THE FRENCH AVM VITRIFICATION FACILITY

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HISTORICAL BACKGROUND

The research and development of the solidification of high level radioactive wastes has been under investigation in France for over 20 years.

First radioactive glasses and synthetic micas were made in 1958 [1]. Glass blocks weighing 5 to 15 Kg and having a specific activity of about 1000 Ci/l were achieved in 1963 by a gel technique in graphite crucible for examination purpose. In the meanwhile, processes of industrial interest were under development : a batch and a continuous one [2] [3] [4] [5] [6] [7] [8]. The first in operation was the batch one, a pot vitrification technique which performs drying, calcination and vitrification in the same vessel, a metallic pot heated by induction. In 1969, a pilot plant called PIVER, using this process was operated under fully active conditions at Marcoule Nuclear Center. Until 1973 it has processed solutions issued from spent natural uranium fuel yielding 12 tons of glass with a maximum activity of 3000 Ci/l.

Because of the successful operation of PIVER, it was decided in 1972 to build an industrial vitrification plant at Marcoule. The continuous process was chosen in preference to the pot technique, as it was found to be more flexible, cheaper and easier to scale up for higher throughputs . A plant called AVM was built and started up in active operation in June 1978 [9].

BASIC PROCESS

The process lies in performing the vitrification in two stages (Fig 1). The first one is a calcination. The feed solution is

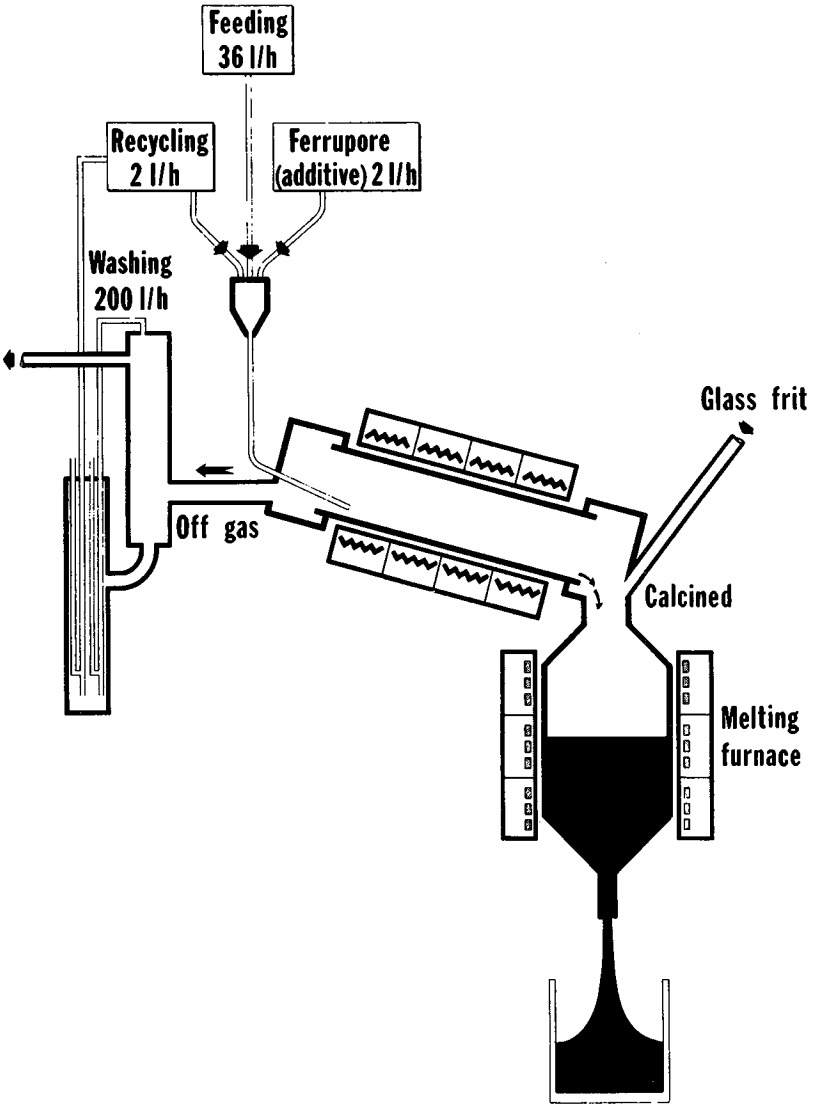

FIG. I CONTINUOUS VITRIFICATION PROCESS

introduced in a rotary kiln to be thermally decomposed into a solid form. The calcined products are then mixed with suitable raw materials in an electric furnace to make the wanted glass.

The glass is poured at stated intervals in containers which are transfered to a disposal facility. The off-gas released during the fabrication are processed in a specific off-gas treatment equipment.

GLASSES INVOLVED IN THE MARCOULE PROCESS

The main liquids to be vitrified at Marcoule fall in 3 types related to the reprocessing of MTR spent fuels, military and commercial natural uranium spent fuels (graphite gas reactor system).

Table I gives the characteristics of these wastes. As it is impossible to determine any single glass dealing with the various composition of the wastes, 3 different groups of glass compositions, all of borosilicate type, have been selected after investigations carried out in cold and hot laboratories [3] [4] [6] [10] [11] [12] [13].

The characteristics of the glasses are plotted in Table II. The examination of the glasses was carried out to determine the behaviour of the structure under irradiation and thermal conditions and their physical and chemical properties connected to the technology of fabrication and radioactive hazards in a final disposal.

THE PLANT

The vitrification plant, designed jointly by C.E.A and S.G.N was built for COGEMA (a) in order to solidify all the solutions of fission products generated by the local reprocessing plant, including those already in storage for over 20 years.

A.V.M is located close to the storage tanks. The construction which started up in the summer of 1974 ended in January 1977 with the first non-radioactive run and gave rise to radioactive operation in June 1978.

(a) Compagnie Générale des Matières Nucléaires

Table I : CHARACTERISTICS OF MARCOULE HLW

		Commercial GG spent fuels	Military GG spent fuels	M.T.R. spent fuels
Burn up		4000 to 5000 MWd/t	1000 to 1200 MWd/t	500 MWd/Kg
Chemical composition (g/l)	F.P.	35 to 40	20 to 25	very low
	U	2.5 to 3.5	1.5 to 2.5	very low
	Np	≃ 0.2	≃ 0.1	very low
	Pu	≃ 0.02	≃ 0.02	very low
	Al	8 to 12	30 to 35	81
	Cr	≃ 0.4	≃ 1.5	< 0.5
	Fe	6 to 10	15 to 17	0.5 to 2
	Ni	≃ 0.3	≃ 1	< 0.5
	Na	4 to 6	19 to 23	1 to 3
	F	--	≃ 8	≃ 12
	Mg	2 to 3	4 to 5	--
Acidity		1.0 M	2.0 M	- 1.8 M (depleted)
Specific activity after 2 years cooling time (Ci/l)		1600 to 1700	500 to 1000	low
Specific power after 2 years cooling time (W/l)		4 to 5	2 to 3	low
Concentration (l/t)		≃ 100	30 to 40	12 000

Table II: CHARACTERISTICS OF MARCOULE GLASSES

		Commercial GG spent fuels	Military GG spent fuels	M.T.R. spent fuels
Example of composition (Weight %)	SiO_2	42.5	40.0	37.0
	Al_2O_3	8.5	13.0	23.5
	B_2O_3	17.5	16.0	16.5
	Na_2O	14.0	17.0	19.5
	MgO	1.0	2.0	-
	Fe_2O_3	1.6	5.0	0.2
	F	1.4	2.0	1.8
	$NiO+Cr_2O_3$	0.5	1.0	0.2
	FP oxides	13.0	4.0	1.3
Current volume reduction (related to the concentrated liquid)		7.0	5.4	3.5
Volume of glass per ton of fuel		13 to 15 litres	5 to 6 litres	3.4 m^3
Specific gravity of the glass		2.50	2.61	2.43
Viscosity at 1100° (Po)		150	100	430
Thermal conductivity at 100° (W/m.°)		1.25	1.25	1.25
Range of the leaching rate (1) ($g/cm^2/d$)		$10^{-7}/10^{-6}$	$10^{-7}/10^{-6}$	$10^{-7}/10^{-6}$

(1) Leaching operated at room temperature with tap water

Description

The plant is made up of two different parts : the so-called plant and a nearby storage facility.
The facility has been previously described [14], so the following descriptions is succinct.

Figs. 2 and 3 show a vertical and a horizontal section.
The main part is the vitrification cell which is totally lined with stainless steel. This cell encloses the chief equipment of the process, the device ensuring the welding of the lids to the cannisters and the head part of the gas off treatment line (deduster and condensor).

Every part of the equipment located inside the cell was designed to be removed for taking off or refitting (as an example see Fig. 4 picturing the dismantling of the calciner).

The vitrification cell is connected to :

- a solution storage cell which comprises two 10 m^3 buffer tanks.
- a decontamination cell which moreover plays a part in the transfer of the glass to the storage area.
- a maintenance cell which is used in addition to drain the equipment in and out and as a garage for the bridge-crane.
- an off gas treatment cell (showed on the cross section of the Fig. 3) which contains a column for the recombination of nitrogen stream and another one as a washing column which also collects the gas issuing from various breather tubes.

The storage facility is close to the vitrification building. It is made up of three engineered underground vaults made of concrete. The canisters (1 m high, 50 cm diameter) are piled up in the 10 m high vertical pits fitted in the vaults. There are 220 pits which meet the need until 1990. A further extension is available.

The ventilation has been designed in order that the maximum temperature of the forced air does not exceed 100°C when the maximum specific power (50 W/l) is applied to the glass. The reason is to limit the temperature of the concrete to 60°C and those of the glass to 500°C (forced air cooling) and 600°C (natural convection) at the center line. (See Figs. 5 and 6).

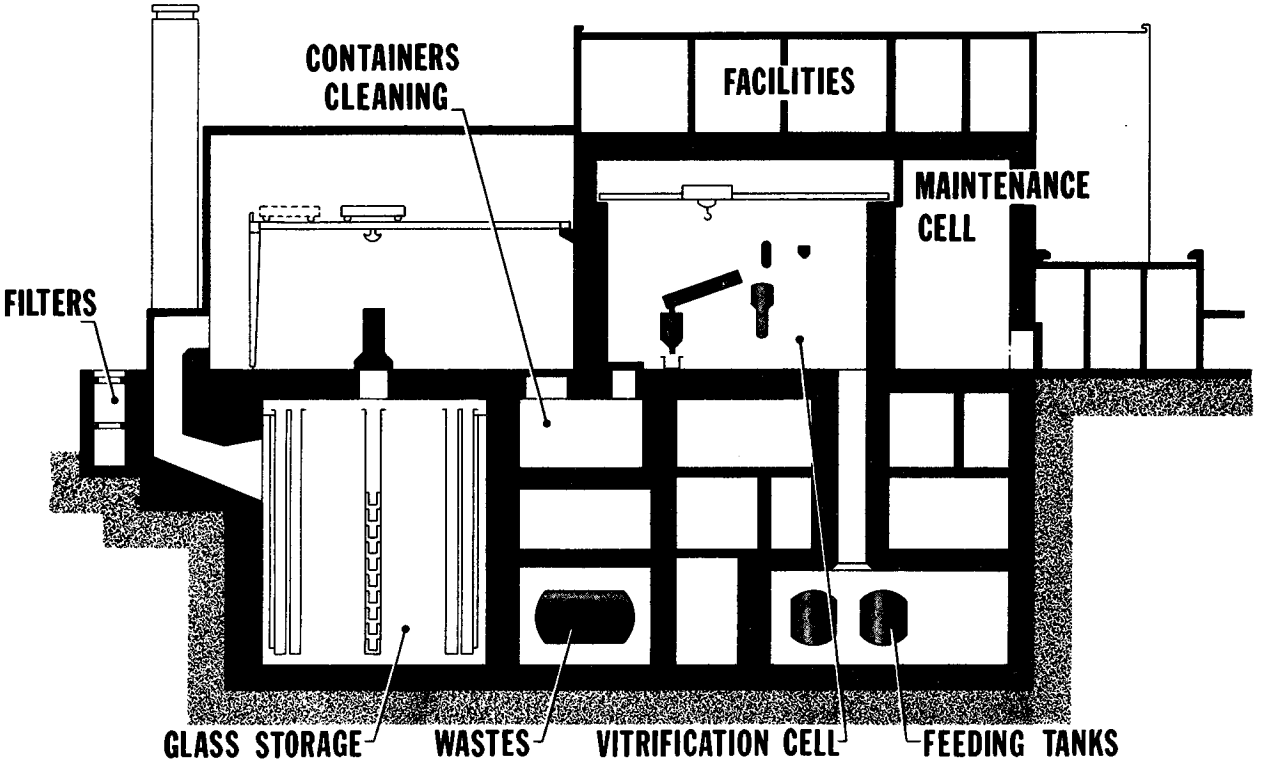

FIG. 2 AVM VERTICAL SECTION

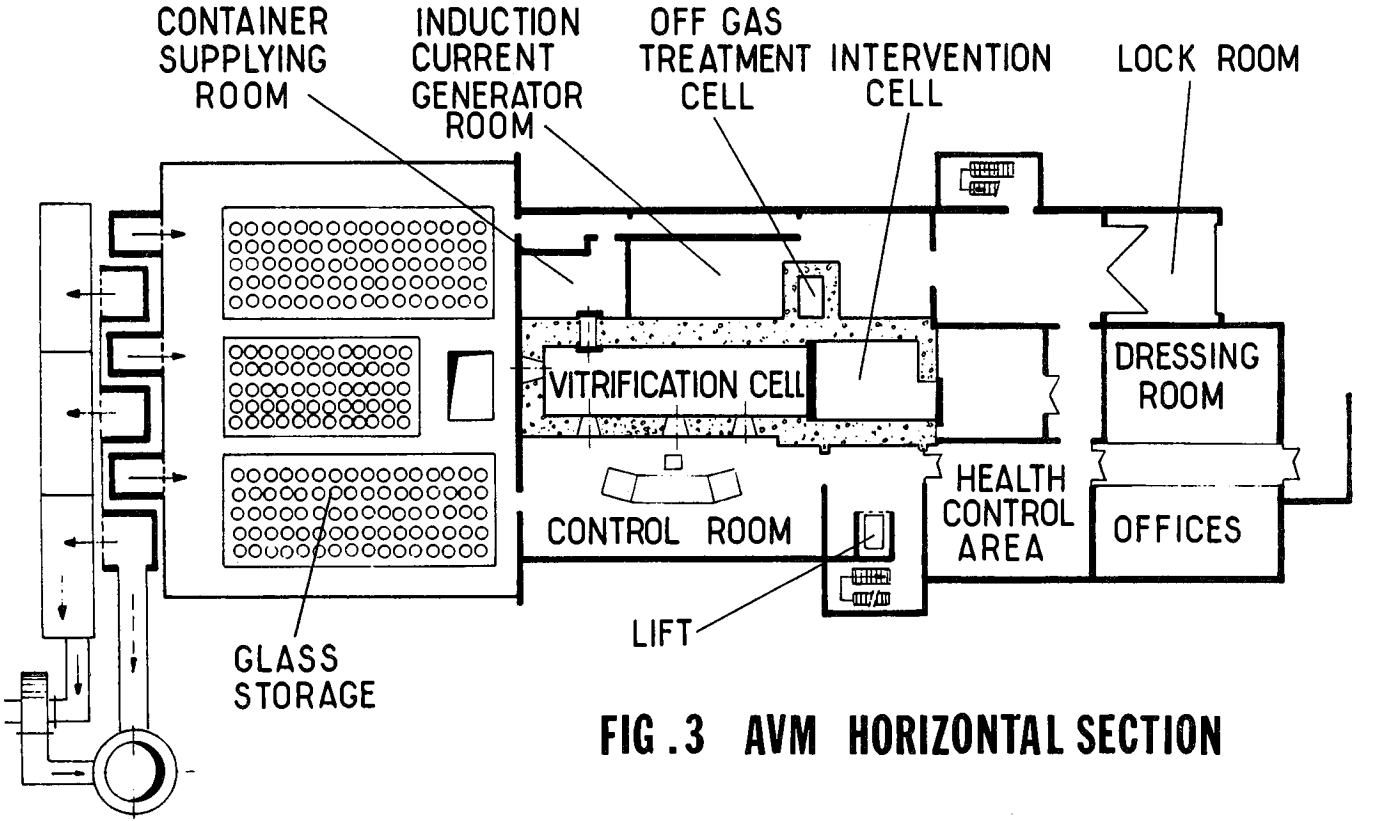

FIG .3 AVM HORIZONTAL SECTION

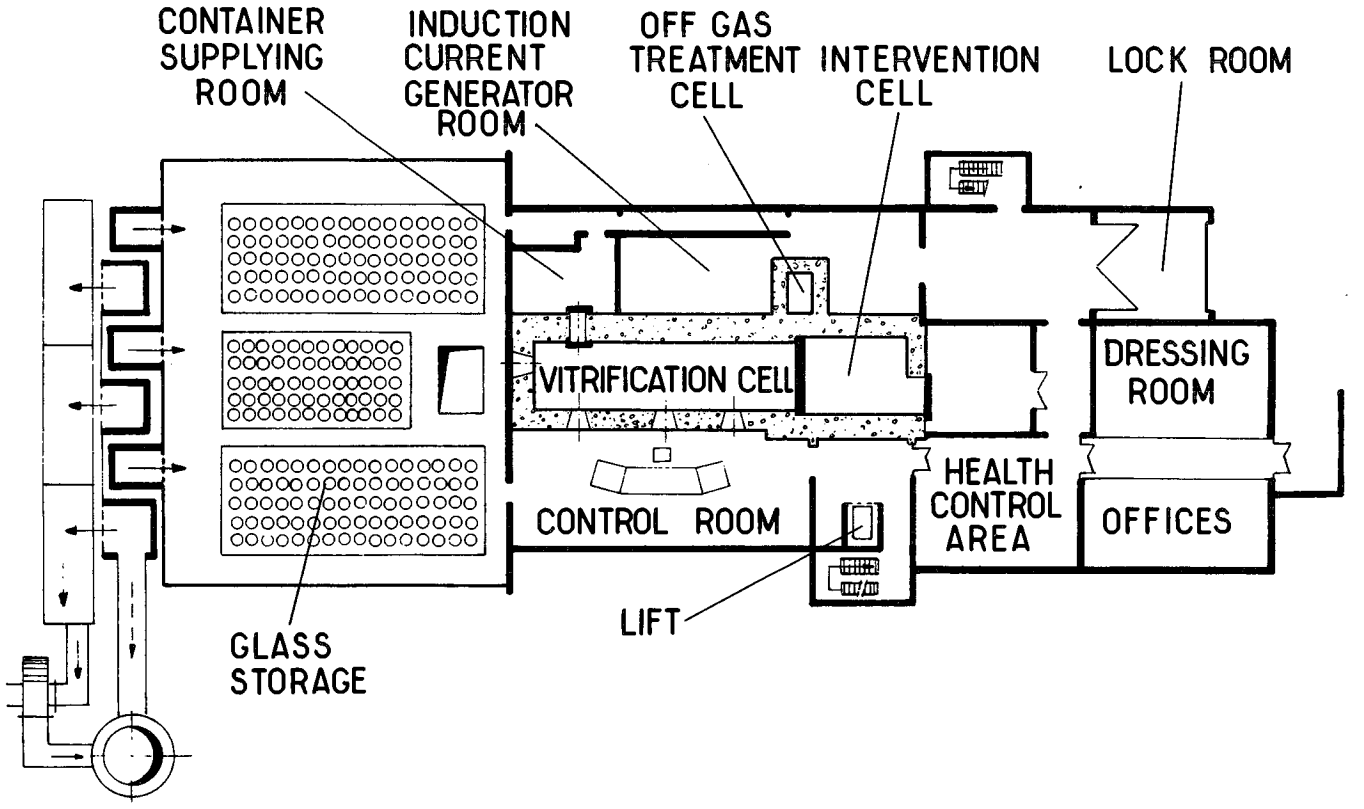

FIG. 3 AVM HORIZONTAL SECTION

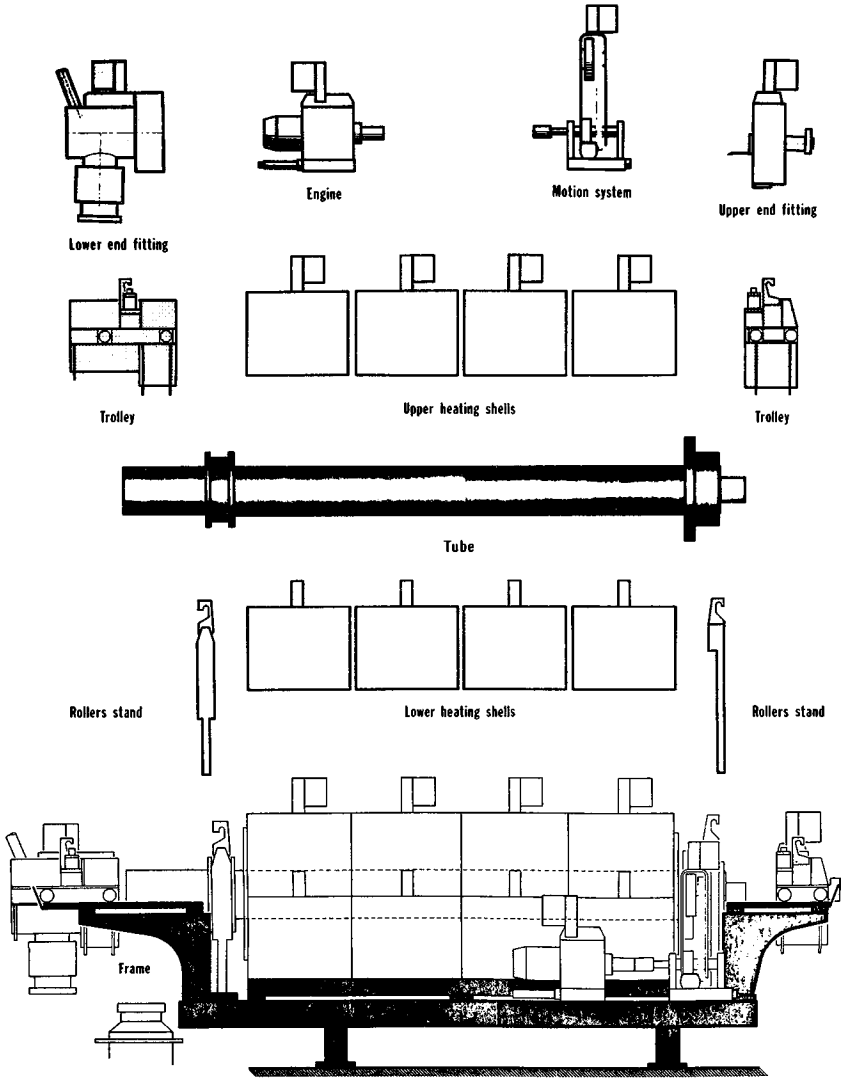

FIG:4 . CALCINER DISMANTLING PRINCIPLE

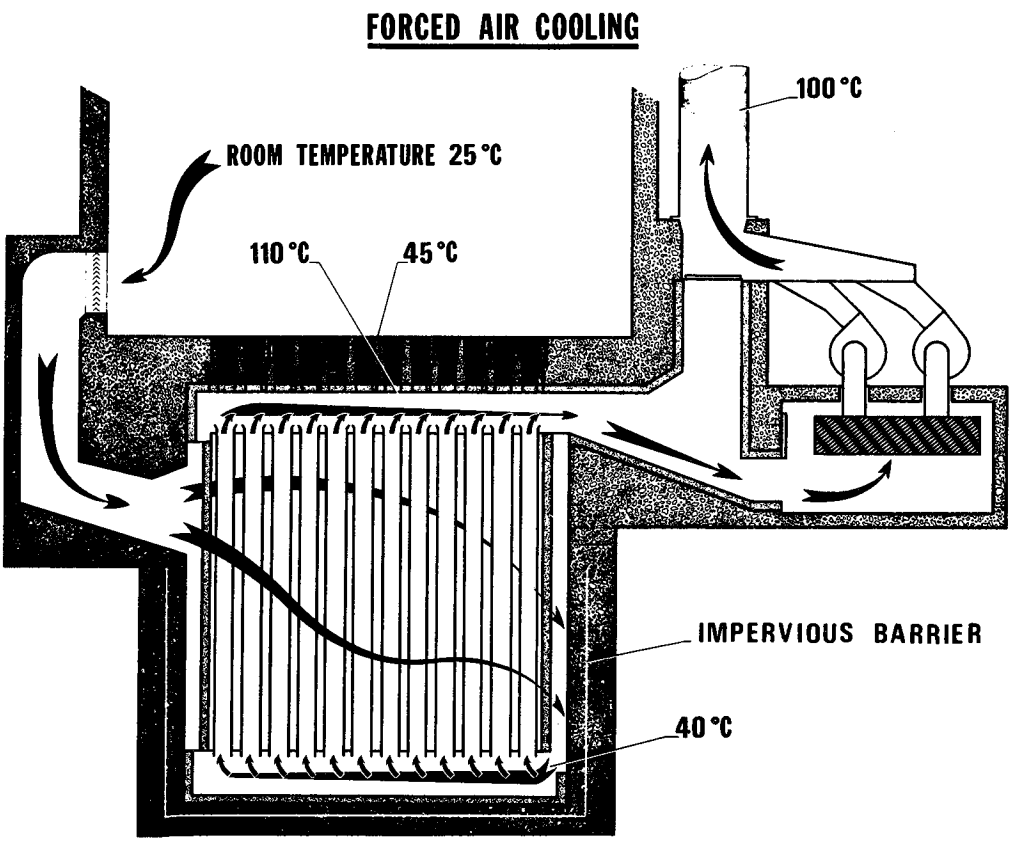

FIG. 5 AVM GLASS STORAGE

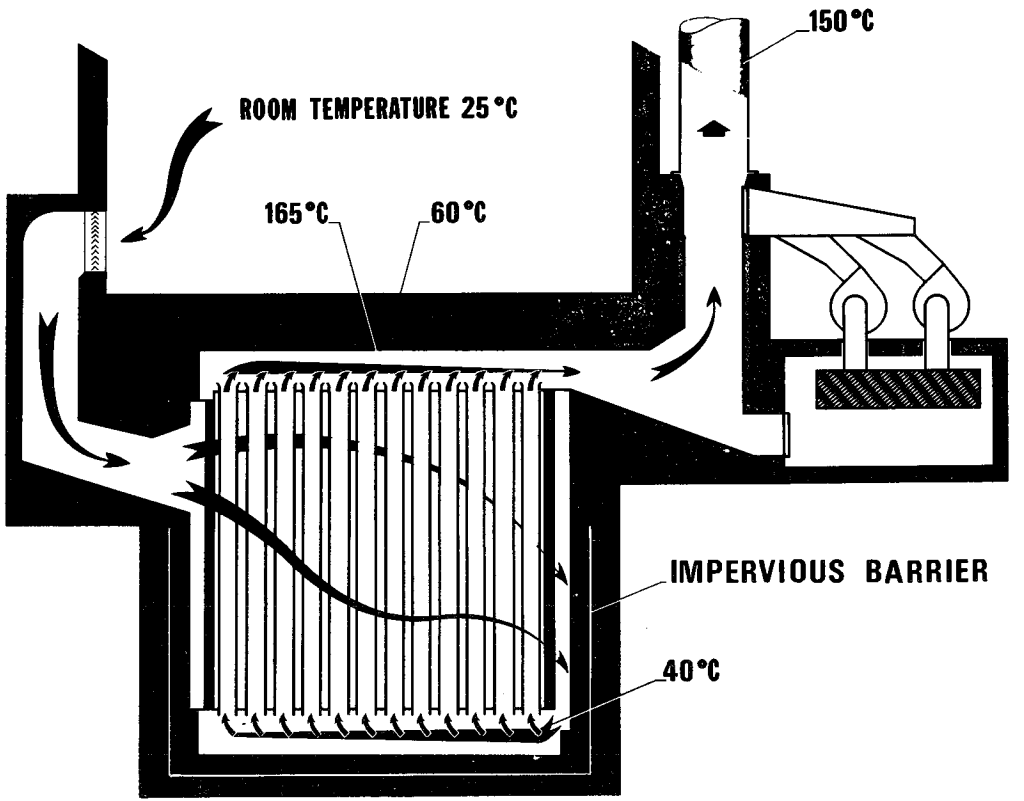

FIG. 6 AVM GLASS STORAGE

The canisters are transfered from the vitrification facility to the pits by means of shielded casks.

Operation :

The solutions coming from the liquid storage tanks are transfered in 10 m^3 batches in the A.V.M tanks. These tanks are cooled and mechanically stirred. The solutions are sampled for analysis and eventually chemically adjusted to cope with the composition of the raw materials.

A double air-lift exhausts them to the vitrification cell into a metering device which is able to feed the calciner with a flow-rate in the range of 30 to 36 l/h according to the concentration of the liquid.

At this step, a chemical additive is introduced at a rate of 2 l/h. This additive is an organic product (azodicarbonamide) the aim of which is to prevent caking on the wall of the calciner and to improve the size distribution of the calcined product. A solution issued from the first cleaning gas equipment is also fed back at a flow-rate of 4 l/h in the metering device. Consequently the total liquid flow-rate can vary from 36 to 42 l/h.

The calcination is operated in a tube heated by a 4 zone furnace running at 30 rpm. The tube which is set with a 3 % tilt is fitted with 2 end-fittings ensuring tightness.

The feed solution is injected continuously through the upper end-fitting. The resulting calcined product (8 to 9 Kg corresponding to 30 l of solution) falls straight into the melting furnace through the lower end-fitting. A loose rod, located inside the tube makes the calcination easier and prevents caking.

The lower end-fitting is also used for the connection to the melting furnace and to feed the raw material in the form of a primary glass (frit). The frit is fed at a mean rate of 9 to 12 Kg/h at stated intervals in 400 to 600 g batches through a lock chamber.

The melting pot is a metallic cylinder made of Inconel 601. A drain tube is fitted in the bottom.

The heating is supplied by a medium frequency induction furnace (10KHz) which is made up of 4 main induction coils.

The throughput is about 15 Kg/h of glass melted at 1100 - 1150°C. The pouring is initiated every 8 hours by heating the cold glass plug located inside the drain tube. Every eight hours, 120 Kg of glass are cast.

The gas evolved in the melting furnace and the calciner exit through the upper end-fitting. They are composed of steam, nitrogen compounds generated by the denitration and an amount of dusts, the main part of which are soluble in nitric acid. For this reason the first step of the off-gas treatment is a counter current washing in a "deduster". The wash solution is continuously recycled into the calciner (Fig. 7).

The stream flows through a condensor and into standard equipment composed of : an absorption column, a washing column, an absolute filter and a fan supplying the necessary negative pressure.

The glass is cast in metallic canisters made of refractory stainless steel. They receive three casts. The weight of the glass is measured during pouring.
One canister loaded with 360 Kg of glass (150 l) is thus produced each day. This is equivalent to about 800 l of fission products solution.

Some hours after filling, the canister is shifted to the welding area where it is sealed with a lid welded with a plasma torch. The cleaning is done one day later by washing the surface with high pressure water (200 bars).

Liquid wastes :

The processing of the liquid wastes depends on their specific activity.

- the condensate and used liquids issued from the first column are considered to be highly radioactive liquids and are recylced to the fission products solutions concentration unit in order to be finally fed back and vitrified.

- the used liquids of the washing column and of the canister decontamination (500 l/d) have an activity below 1 mCi/m^3 and are therefore deemed only as dubious wastes and transfered to the low-level waste treatment plant. They are, in a way, the only liquid wastes released by A.V.M.

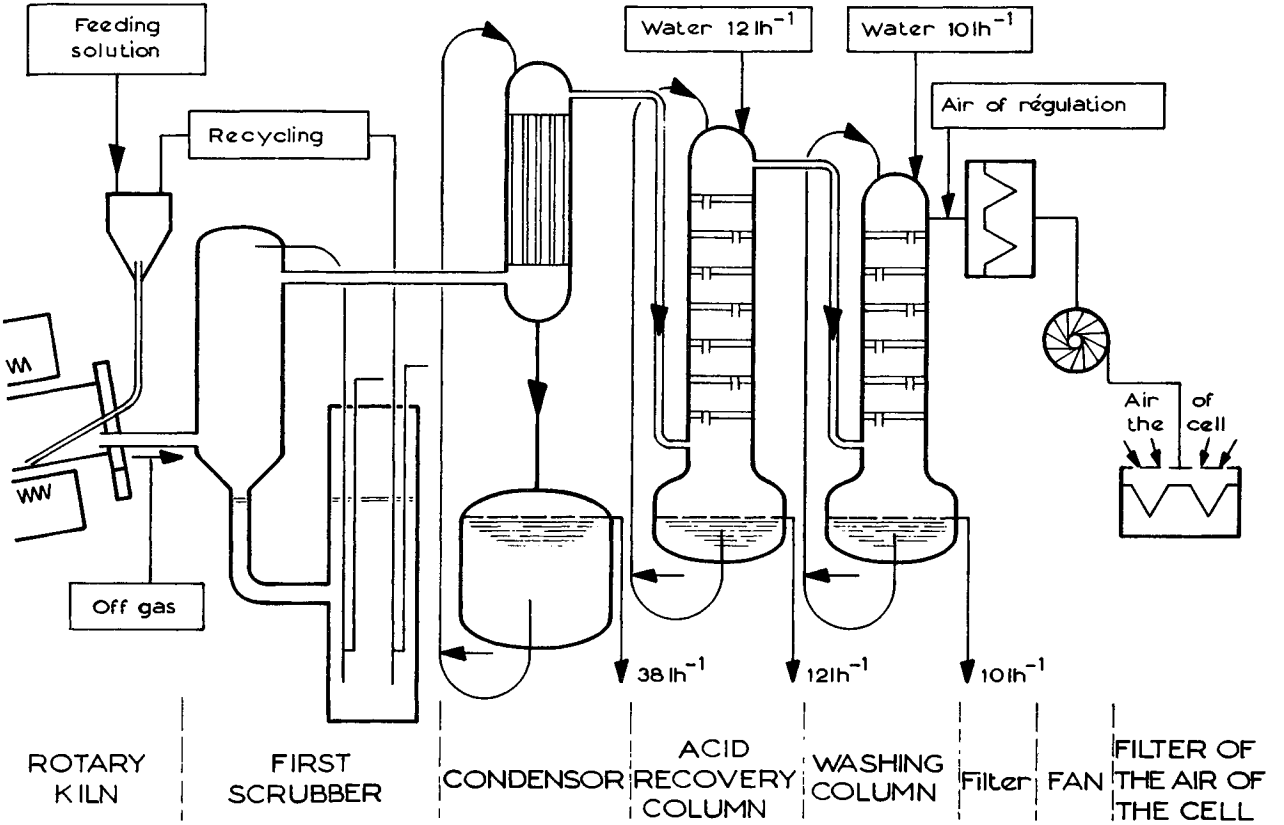

Fig: 7 - OFF GAS TREATMENT OF THE A.V.M.

Solid wastes :

Solid wastes are possibly generated by any failure or by the removing of equipment or vessels at predeterminated life-time. The latter specifically includes the vitrification pot which has to be removed every 2000 hours.

These wastes are able to be submitted to a previous decontamination in the vitrification cell by soaking in a special tank. It is also possible to cut them partially to shorten their overall size, they are then either transfered to the maintenance cell to be conditioned in concrete in drums or put in canisters similar to the glass ones.

The latter is the case of the pot vitrification which is, when used,put in a canister. This one is welded and disposed in a particular storage pit prior to be conditioned by melting as the major part of the metallic wastes will be later on.

FINDINGS AND RESULTS

Operational experience :

From January 1977 to April 1978, running tests have been achieved. Their target was :

- To vitrify simulated solutions similar to the three types stored at Marcoule. During this period of time, 18.1 m^3 of solution were vitrified to yield 8.8 tons of glass (see Table III).

- To perfect automatized devices and to look for better safety conditions in case of failure of various feed (raw material, electric current, air, steam etc...).

- To carry out every remote operation in order to verify each operation. These tests required over 10,000 hours of working time.

The plant was gradually put in active condition by increasing the radioactivity of the feeding solution. The first campaign using concentrated fission products solutions started up on June 28, 1978 and ended on July 24, 1978 for staff vacations. The processed solution was an old one of 30 Ci/l of specific activity generated by the reprocessing of SICRAL type spent fuel (concentration 35 l/ton) and fed at a rate of 30 l/h.

TEST WITH SIMULATED SOLUTION	WORKING TIME	VOLUME OF TREATED SOLUTION	WEIGHT OF GLASS	NUMBER OF GLASS CONTAINERS FILLED
Military GG Spent fuels (low burn up)	365 h	10.0 m^3	5.0 t	15
M.T.R Spent fuels	231 h	6.6 m^3	3.25 t	10
Commercial GG Spent fuels (high burn up)	52 h	1.5 m^3	0.55 t	2
TOTAL	648 h	18.1 m^3	8.8 t	27

TABLE III : COLD TEST OF THE AVM PLANT

Another campaign related to the reprocessing of the same type of solution was initiated at the end of September 1978 (flow-rate 33 l/h) and stopped 1000 hours later, to remove the pot vitrification. This was an operation scheduled due to the age of the pot (over 2000 hours). On the other hand the interruption was extended to remove a valve from the raw materials feeding line which failed unexpectedly.

A third campaign began on 23 January 1979 involving solution 6 to 8 times more active than the previous one. It is presently still running with a feeding flow rate of 33 lh^{-1}.
Apart from the above mentioned valve, all equipment has been running satisfactorily. The liquid feed was ensured very accurately by the metering wheel. The linear expansion of the tube has kept the same value and the inside pressure (- 15 cm H_2O) was also very stable.

The melting of the glass in the pot was operated, keeping the temperatures of the wall below 1130°C. The casts initiated every 8 hours drained out 110 to 120 Kg of glass during 15 to 20 minutes feeding time viz.at a mean flow-rate of 350 Kg/h.

Results :

During the 2 first campaigns, 210 casts were performed in 70 canisters (3 casts in each).
A total volume of 50 200 liters were processed during a 1644 hours feeding time. This corresponds to 1400 tons of spent fuels giving rise to 23.4 tons of glass disposed in 70 canisters lodging 330 Kg to 360 Kg of glass each. The factor of volume reduction is 5.4 as it was calculated.

At the date of 9 February 1979, 23 more canisters were filled and an additional volume of 17 300 liters of fission products solution was vitrified (see Table IV). The welding of the lid on the canister, operated some hours after the last cast, did not suffer any difficulty except two times when an extra welding had to be done.

Each canister was washed with 450 liters of pure water under a 200 bars pressure for three minutes. The radioactivity of the wash water was found to be below 2.10^{-4} µCi/ml which is the limit of detection.

		WORKING TIME	VOLUME OF TREATED SOLUTION	WEIGHT OF GLASS	NUMBER OF GLASS CANNISTERS FILLED
FIRST CAMPAIGN	Cold campaign	97 h	2.4 m^3	1. 15 t	3
	Hot campaign	649 h	19.2 m^3	8. 96 t	28
SECOND CAMPAIGN		994 h	31.0 m^3	14. 44 t	42
THIRD CAMPAIGN (a)		520 h	17.3 m^3	8. 2 t	23
TOTAL		2260 h	69.9 m^3	32.8 t	96

TABLE IV : AVM HOT RUNS DATA

(a)still in progress - Assessment on 9 February 1979

There was no activity in the air stream injected around the canister.

- The efficiency of the recombination of nitric acid and nitrates reached over 90 % in the condensor and over 82 % in the first column. This confirms the data from the non-radioactive campaign.

- About the non radioactive cations, no insoluble dust was noticed in the liquid of the deduster. Chemical analysis of this liquid and of the condensate pointed out an average draining out of 3 to 4 % and a trapping efficiency of 96 to 98%. This also verifies the results of the non radioactive runs [15].

The data related to the volatilization of the radionuclides and the various decontamination factors are plotted on Table V and Figs. 8, 9, 10, 11.

It appears the volatilization rate of the ruthenium, the main volatile element, varied from 39 % (57 % of which recycled) for the 1st campaign to 20 % (65 % of which recycled) for the 2nd one.

In the meanwhile the Ru decontamination factor of every parts of the equipment increased perceptibly.

The activity released in the main liquid waste (used water of the washing column) rose (in Ci/h)

	1st campaign	2nd campaign
106 Ru	$15.0\ 10^{-6}$	$0.7\ 10^{-6}$
137 Cs	$0.5\ 10^{-6}$	$1.9\ 10^{-6}$
90 Sr	$0.3\ 10^{-6}$	$< 0.4\ 10^{-6}$
144 Ce	$1.1\ 10^{-6}$	$0.3\ 10^{-6}$

The average flow-rate of this waste is 10 l/h. This ought to be considered satisfactory and to be credited to the good decontamination factors of the condensor and the recombination column. The Ru volatilization does not appear therefore as a shortcoming. Similar values related to the releasing of Ru were found out in

	% OF THE ELEMENTS				
	ESCAPED FROM THE CALCINER	RECYCLED IN THE CALCINER	IN THE CONDENSATE	IN THE ACID RECOVERY COLUMN	IN THE WASHING COLUMN
^{106}Ru 1st camp.	38.7	22.0	16.7	0.07	10^{-4}
^{106}Ru 2nd camp.	19.7	12.7	7.0	$2.28\ 10^{-2}$	10^{-4}
^{137}Cs 1st camp.	11.5	10.1	1.4	$3.3\ 10^{-3}$	$1.3\ 10^{-7}$
^{137}Cs 2nd camp.	6.4	5.2	1.2	$3.0.\ 10^{-3}$	$4.3\ 10^{-7}$
^{144}Ce 1st camp.	5.9	4.0	1.9	$0.40\ 10^{-3}$	$0.50\ 10^{-5}$
^{144}Ce 2nd camp.	3.9	2.2	1.7	$0.02\ 10^{-3}$	$0.13\ 10^{-5}$
^{90}Sr 1st camp.	5.2	5.0	0.2	$3\ 10^{-5}$	$0.7\ 10^{-7}$
^{90}Sr 2nd camp.	3.0	2.9	0.1	$3\ 10^{-5}$	$0.7\ 10^{-7}$

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BACK TO THE FP CONCENTRATION AND VITRIFICATION

TABLE V : EFFICIENCY OF THE OFF GAS TREATMENT
RESULTS OF THE AVM FIRST AND SECOND CAMPAIGNS

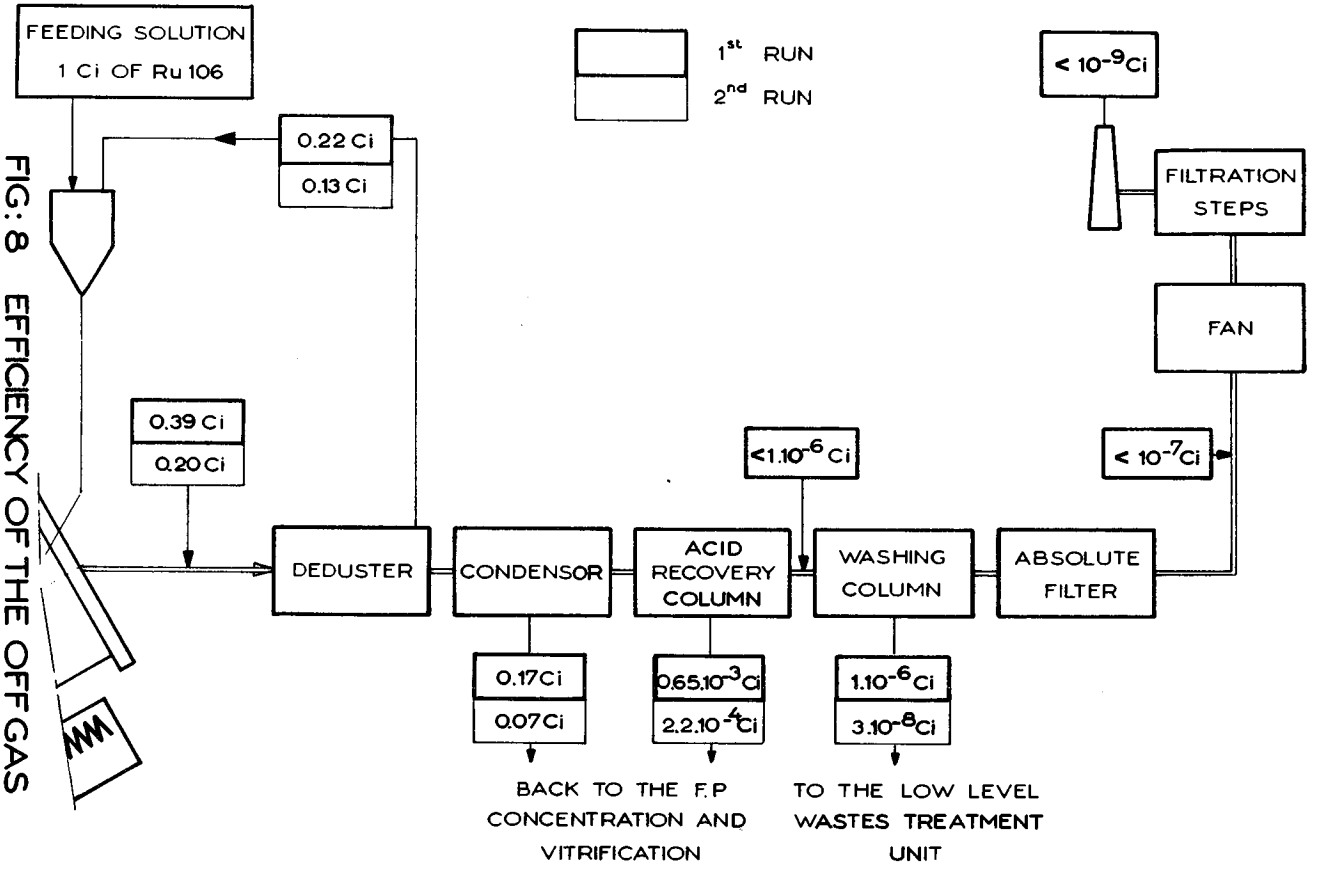

FIG: 8 EFFICIENCY OF THE OFF GAS TREATMENT FOR THE Ru 106

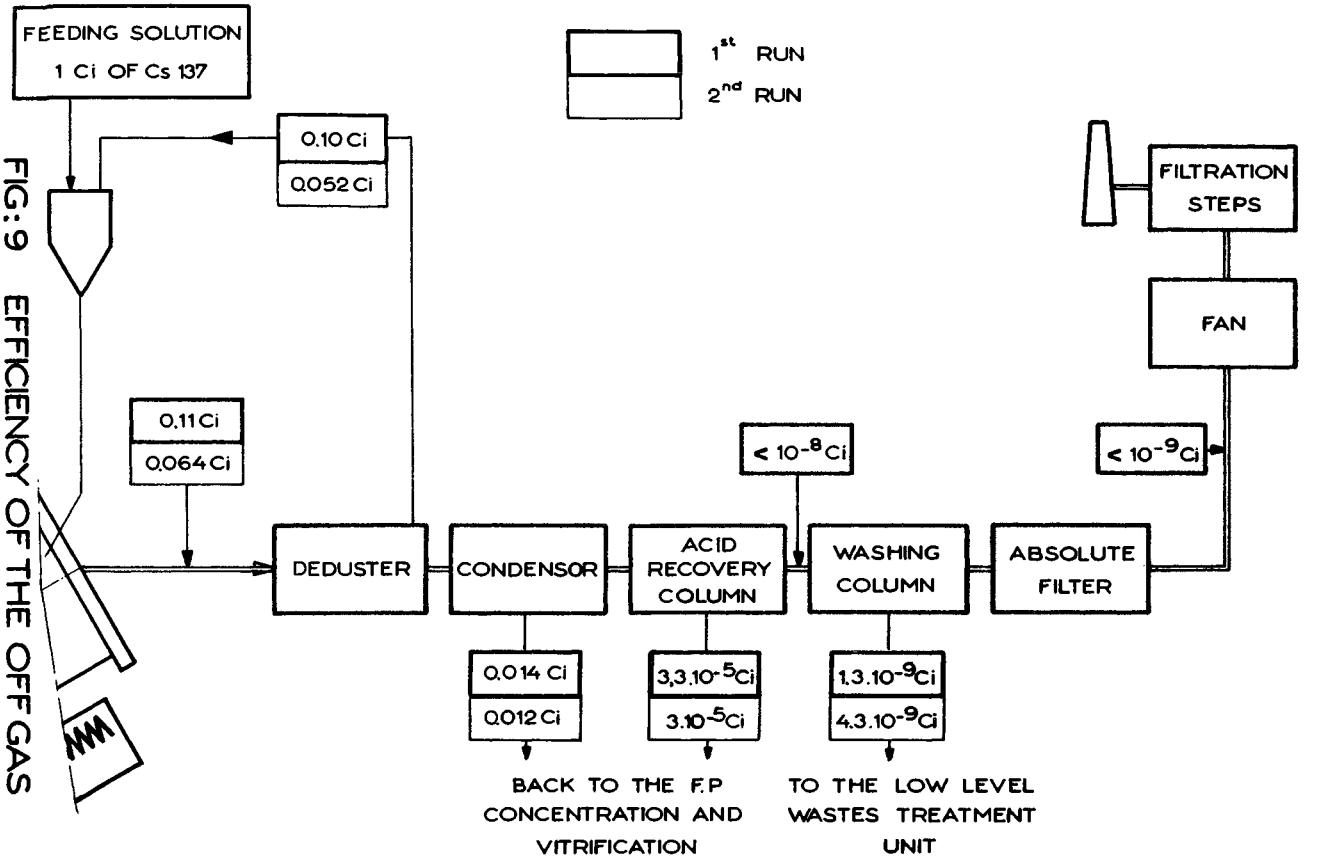

FIG: 9 EFFICIENCY OF THE OFF GAS TREATMENT FOR THE Cs 137

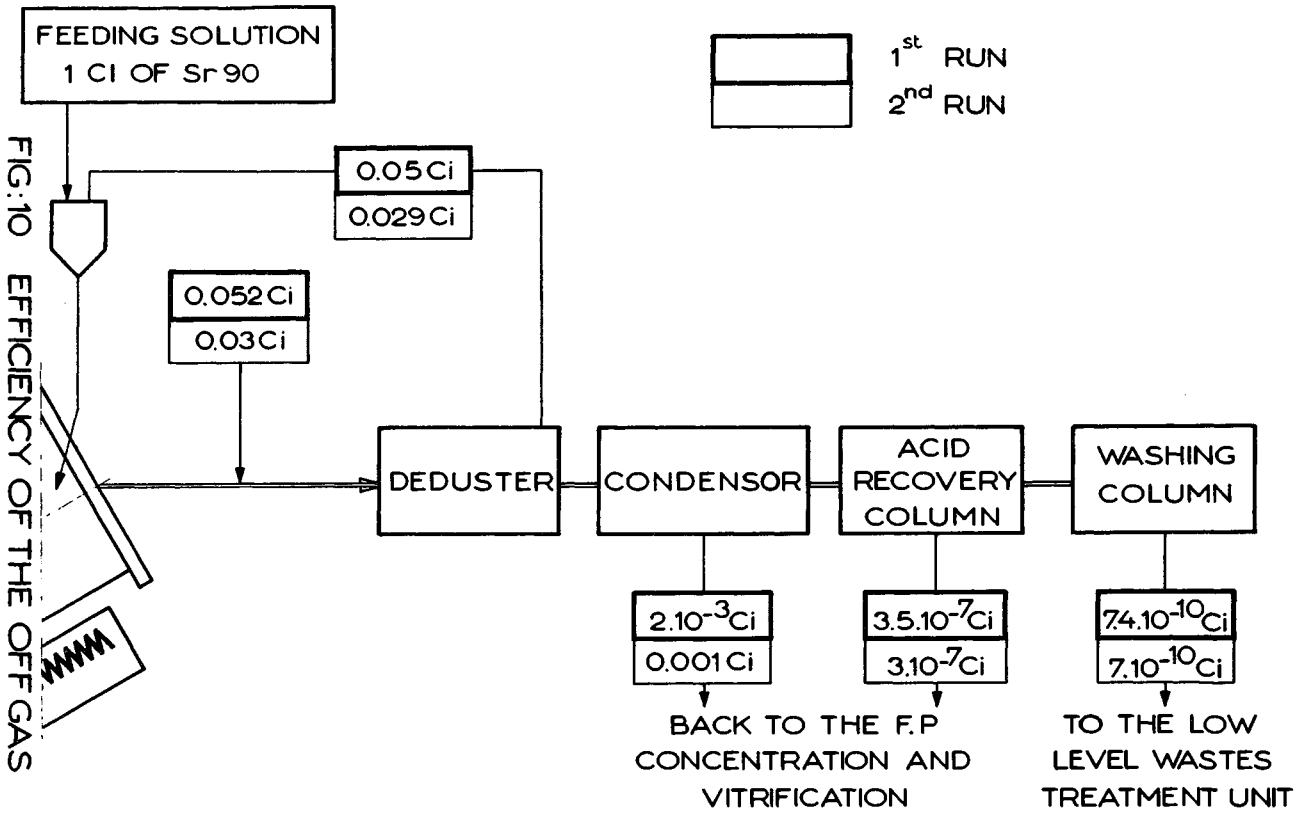

FIG:10 EFFICIENCY OF THE OFF GAS TREATMENT FOR THE Sr 90

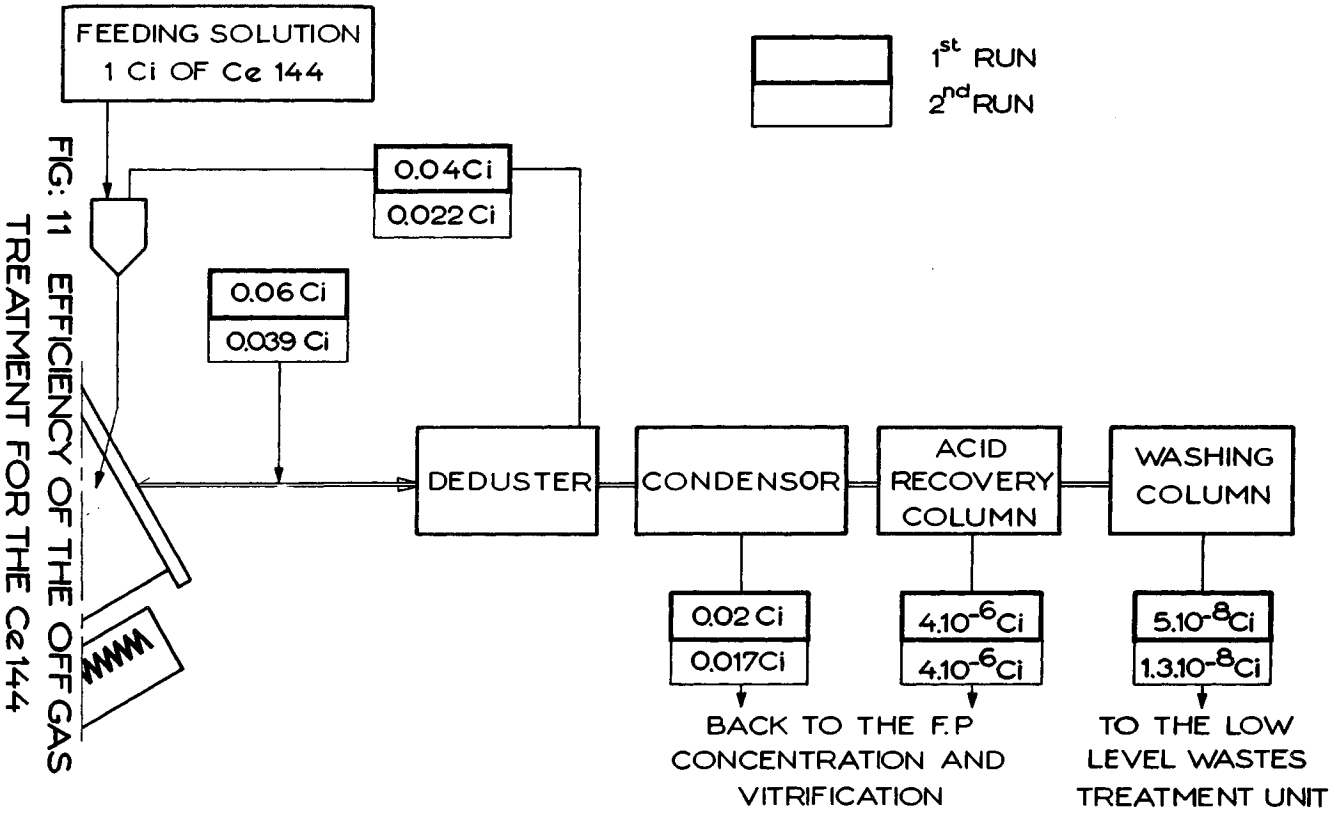

FIG: 11 EFFICIENCY OF THE OFF GAS TREATMENT FOR THE Ce144

the pilot plant ATLAS which is half scale AVM equipment.
In this pilot plant, attempts were made to reduce the Ru volatilization by adding sugar to the solution. Figure 12 shows the marked effect of this product upon the volatilization rate which is reduced to 2 % out of the calciner and less than 1 % in the condensate.

Gaseous wastes :

In order to evaluate the decontamination factor of the off-gas treatment equipment, some samples were taken in various places in the line. They were not able to give rise to any value owing to the very small activity of the stream but they provided the justification to assume minimum values.
Those ones are, just before the final absolute filter, as following.

	D.F
Ru	$\geqslant 1.2\ 10^7$
Cs	$\geqslant 1.2\ 10^9$
Ce	$\geqslant 2\ 10^7$

(These data are related to the 1st campaign, the measurement of the next ones is presently in progress).

About the possible contamination of the vitrification cell, an activity rate varying from 0 to 60 counts per second, has indicated a very low level contamination.

LA HAGUE NEXT VITRIFICATION PLANT

A vitrification plant, assigned to vitrify fission products solutions of LA HAGUE current reprocessing plant, is presently in design.
Another one is scheduled to vitrify the fission product solutions generated by the next reprocessing plant which will be located on the same site.

Scaling up to cope with the need of a 800 Mg/yr reprocessing plant could be done according to the diagram shown in Fig.13 with

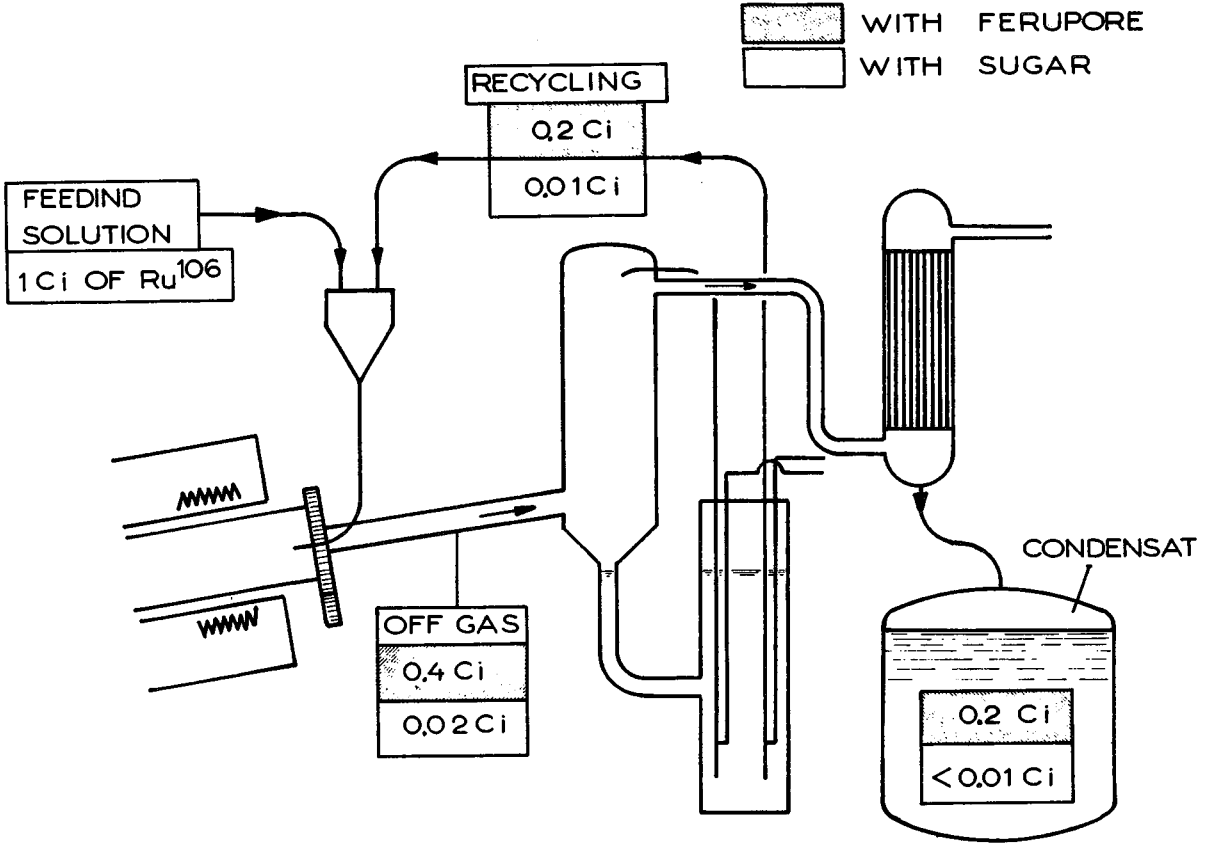

FIG. 12 EFFECT OF THE ADDITION OF SUGAR ON THE Ru VOLATILITY

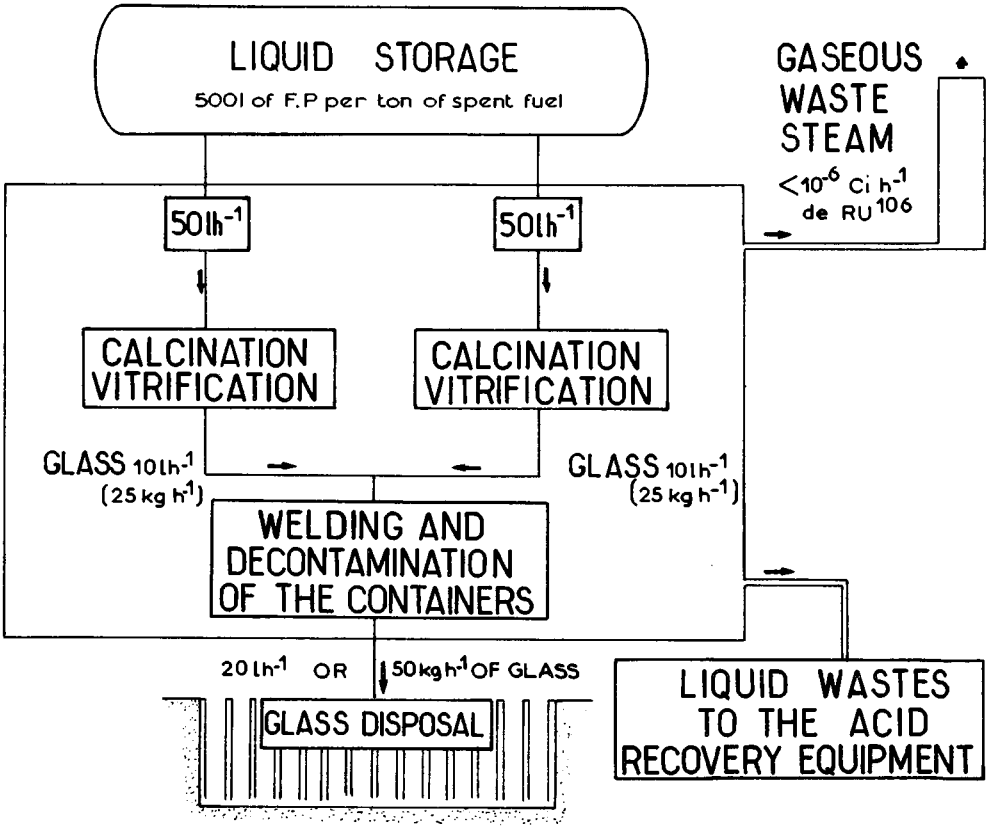

FIG. 13 DIAGRAM OF "LA HAGUE" VITRIFICATION PLANT (UP3 800 t/year)

the following assumptions :

- The spent fuel is reprocessed after 3 years cooling time
- The fission product solutions are concentrated to 500 l/Mg that sets a specific power of 7 w/l (at the time of reprocessing).

The vitrification plant will have then to face up solutions of a specific power of 5 to 6 watts per liter that will give rise to 25 to 30 w/l of glass, taking into account a volume reduction factor of 5.

Two reprocessing lines will be necessary. Each line will be composed of one calciner connected with a melting furnace. The rotary tube might be heated by induction in order to shorten the time needed to reach the steady state and to shut off. This heating device could by the way, when used, become a less bulky solid waste than a resistance furnace.

Every calciner will be fed at a flow-rate of 50 l/h with a 500 l/Mg concentrated solution. Every connected melting furnace will produce 25 Kg of glass per hour.

The whole throughput of the plant will be 600 m^3 per year with a loading factor of 250 days a year.

The glass will be cast in 40 to 45 cm diameter cylindrical canisters running 200 Kg batches. After welding of a lid, tightness test and outside decontamination, they will be transfered to a storage facility to be cooled for 2 years by forced air and further on by natural convection.

The resulting liquid wastes (mainly condensates), will be concentrated and recycled to the storage tanks to be vitrified. The only generated liquid wastes will be dubious ones which will be directed to the low level processing unit on site.

The gross decontamination factor of the plant for the off-gas could reach 10^{11} for ruthenium and at least matching values for the other nuclides.

CONCLUSION

The commission of A.V.M has opened the industrial development era for the continuous vitrification process.

The very good results which have been obtained until now in running the plant as well as the very small level of the activity of gaseous and liquid wastes may make thinking that the problem of the management of fission product solutions is going well.

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