La Hague Continuous Improvement Program: Enhancement of the Vitrification Throughput

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ABSTRACT

The vitrification of high-level liquid waste produced from nuclear fuel reprocessing has been carried out industrially for over 25 years by AREVA/COGEMA, with two main objectives: containment of the long lived fission products and reduction of the final volume of waste.

At the "La Hague" plant, in the "R7" and "T7" facilities, vitrified waste is obtained by first evaporating and calcining the nitric acid feed solution-containing fission products in calciners. The product -named calcinate- is then fed together with glass frit into induction-heated metallic melters to produce the so-called R7/T7 glass, well known for its excellent containment properties. Both facilities are equipped with three processing lines.

In the near future the increase of the fuel burn-up will influence the amount of fission product solutions to be processed at R7/T7. As a consequence, in order to prepare these changes, it is necessary to feed the calciner at higher flow-rates.

Consistent and medium-term R&D programs led by CEA (French Atomic Energy Commission, the AREVA/COGEMA's R&D and R&T provider), AREVA/COGEMA (Industrial Operator) and AREVA/SGN (AREVA/COGEMA's Engineering), and associated to the industrial feed back of AREVA/COGEMA operations, have allowed continuous improvement of the process since 1998:

- The efficiency and limitation of the equipment have been studied and solutions for technological improvements have been proposed whenever necessary,

- The increase of the feeding flow-rate has been implemented on the improved CEA test rig (so called PEV, Evolutional Prototype of Vitrification) and adapted by AREVA/SGN for the La Hague plant using their modeling studies; the results obtained during this test confirmed the technological and industrial feasibility of the improvements achieved,
- After all necessary improved equipments have been implemented in R7/T7 facilities, and a specific campaign has been performed on the R7 facility by AREVA/COGEMA. The flow-rate to the calciner was increased from 76 l/hr to 90 l/hr in three steps. Results of each step were followed by a team of experts from CEA, AREVA/COGEMA and AREVA/SGN analyzing the impact on the vitrification facility.

The results obtained at the end of this testing campaign were a success: the target feeding flow-rate was reached, improving the line productivity without consequences in equipment availability and off-gas treatment performances.

This paper presents the results obtained for each step of this technological improvement and illustrates the La Hague continuous improvement program performed in order to enhance the vitrification throughput.

INTRODUCTION

HLW Vitrification

Vitrification of high-level liquid waste is the internationally recognized standard to both minimize the impact to the environment resulting from waste disposal and the volume of conditioned waste. Many countries such as the USA, France, the United Kingdom, Germany/Belgium, Japan, Russia, have vitrified HLW (high level waste) and several more countries are studying application of the vitrification technology.

French Vitrification Foundations [1]

The first work on vitrification of radioactive waste began in France in 1957 at the Saclay nuclear center with early the selection by CEA of:

- Borosilicate glass as the most suitable containment matrix for waste from spent nuclear fuel.
- Induction-heated vitrification technology: the obvious advantage of this solution is the simplicity of the joule heating of a metallic melter by using electric inductors and the fact that the heating system is outside the metallic melter (melting pot).

Industrial French Vitrification Design

Learning from the PIVER vitrification experience (first industrial-scale prototype unit in the world intended for vitrification of concentrated fission product solutions in 1968 at Marcoule, in southern France), the basic principles leading to the choice and design of the French industrial two-step vitrification process with hot induction metallic melter [2] are:

- The separation of the processing functions (calcinations/melting), to have simpler and more compact equipment and to limit the size of the melter, allowing complete in-cell assembly and disassembly with moderate size overhead cranes, master-slave manipulators and remote controlled tools.
- Easy remote maintenance of the process equipment with minimization of solid wastes generated during operation

An overview of the two step vitrification process is shown in Fig. 1.

In the two step process the nitric acid solution containing the concentrated fission products solution coming from reprocessing operation is fed to a rotary calciner which performs evaporating, drying and calcining functions.

Aluminium nitrate is added to the feed prior to calcination to avoid sticking issue in the calciner (melting of NaNO3). Sugar is also added to the feed prior to calcination to reduce some of the nitrates and to limit ruthenium volatility.

At the outlet of the calciner, the calcine falls directly into the melting pot along with the glass frit which is fed separately. The melting pot is fed continuously but is batch poured. The melting pot is made of base nickel alloys; the glass in the melter is heated to a temperature of 1100°C and is fully oxidized.

Off-gas treatment comprises a hot wet scrubber with tilted baffles, a water and nitric acid vapor condenser, an absorption column, a washing column, a ruthenium filter and three HEPA filters. The most active gas washing solutions are recycled from the wet scrubber to the calciner. The other solutions are concentrated in an evaporator before recycling in the vitrification plant.

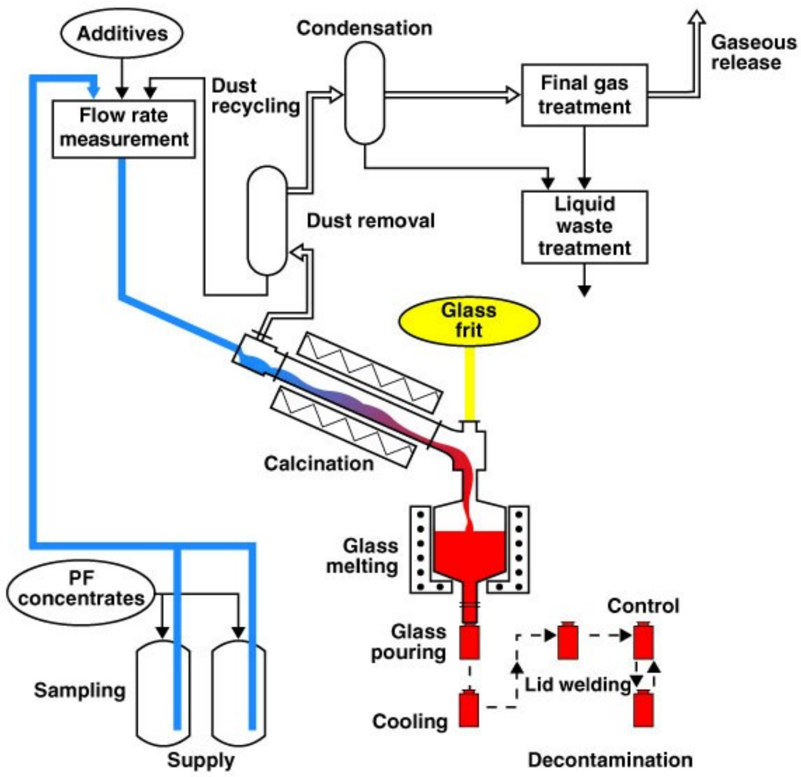

Fig. 1. Two step vitrification process.

The R7/T7 Facilities

Based on the industrial experience gained in the Marcoule Vitrification Facility (namely AVM), the vitrification process was implemented at larger scale in the late 1980's in the R7 and T7 facilities in order to operate it in line with the UP2 and UP3 reprocessing plants. Both vitrification facilities are equipped with three vitrification lines having each a maximal glass production capacity of 25 kg/h.

Continuous Improvement Management

In order to implement on commercial vitrification plant the improvements studied on R&D pilot, a specific organization is managed. It modules close links between research, engineering and operating team, and also judicious build up of results and experience so that R&D process optimisation stays compatible with industrial operation.

As a consequence, AREVA/COGEMA, CEA and AREVA/SGN have acquired a unique experience in the vitrification field of high-level waste coming from reprocessing activities through:

- The design and operation of three industrial vitrification facilities (AVM, R7 & T7) with high records of safety, reliability and product quality;

- The design of various glass formulations including those used in the AVM, R7 and T7 facilities which, together, have produced up to now more than 13000 glass canisters (corresponding to more than $180X10^6$ TBq immobilized in 5500 tons of glass);

- Continuous efforts to improve at the same time the technology (from hot to cold crucible) and the associated matrix formulations, with constant emphasis on quality and volume reduction.

New Production Requirements

The evolution of fuel characteristics and the vitrification of a new effluents generation lead AREVA/COGEMA to consider farther optimization of its vitrification lines in order to keep on managing on line High Level Wastes.

The vitrification of HLW with the increase of fuel burn up from 40 GW/t to 45 GW/t requires using the melter at its maximum capacity i.e. at a 25 kg/hr flow rate instead of the 20 kg/hr as used currently. To achieve this objective, the limiting equipment is the calciner, which feeding capacity has to be increased from a 76 l/hr HLW flow rate to 90 l/hr. This flow rate should also allow AREVA/COGEMA to vitrify new kinds of diluted acid or neutralised caustic effluents which require an increased evaporation capacity.

Consistent and medium-term R&D programs led by CEA, AREVA/COGEMA and AREVA/SGN, and associated to the industrial feed back of AREVA/COGEMA operations, have allowed continuous improvement of the process since 1998 in order to feed the calciner with an increased flow-rate. These improvements are detailed in the paper.

R&D ON CALCINER OUTPUT AND CONTROL

Calciner Description

The calciner process principles and design are shown in Fig. 2.

The calciner mainly consists of:

- An electrically heated cylindrical furnace equipped with 8 half-shells constituting 4 zones in the length of the calciner. These half-shells include heating resistance to heat the tube through radioactive transfer, thermal insulation and thermocouples.
- A rotating tube and a rabble bar
- Rollers bearing that support the rotating tube
- Upper and lower end-fitting ensuring off-gas containment, in spite of tube expansion and rotation.
- A tube motor drive unit rotating the tube at a constant speed between 20 and 30 rpm.

Process Description

The calciner functions are the following:

- Evaporating the liquids coming from constant volume feeder (in the so-called evaporation area).
- Decomposing nitrate salts partially (in the so-called calcination area).
- Elaborating satisfactory calcinate.
- Ensuring a continuous feeding of the melter furnace.

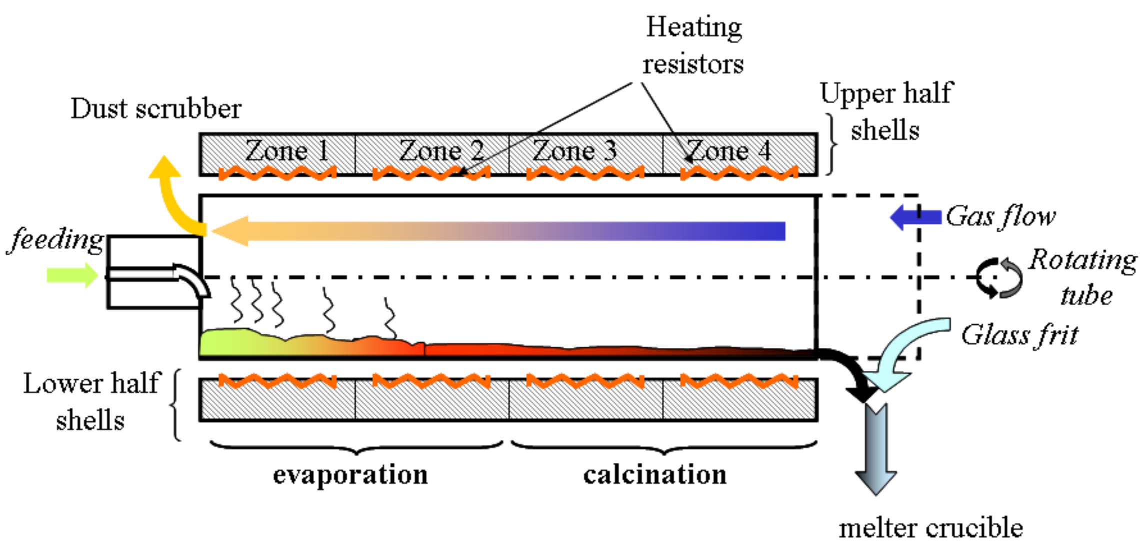

Fig. 2. Calciner process principles and design.

The calciner was initially designed to evaporate a 60 l/hr water feeding. A specific calcination campaign was carried out on the CEA test rig (so-called PEV, Evolutional Prototype of Vitrification) in 1993 with a 80 l/hr feeding flow in order to check the calciner capacity and the dust scrubber behaviour. The positive results of this test allowed AREVA/COGEMA to increase the R7/T7 calciner feeding flow up to 76 l/hr without equipments modification and without any consequences noticed on the process. AREVA/COGEMA vitrification facilities at La Hague are currently operated at this higher flow rate.

On the other hand, the increase of the calciner evaporative capacity from 76 l/hr to 90 l/hr led to operate calciner at its limits of operation (higher flow-rates and higher temperatures set points) which implied:

- a new specific testing program to determine a possible equipment limitation,
- a particular effort to better understand calciner behaviour in order to refine control and operations.

Technological Improvements

R&D program, led to increase calciner feeding flow-rate, began in 1998 by studying the efficiency and limitation of the equipment, and by finding solutions for technological improvements whenever necessary:

1. The increase of the feeding flow-rate has been implemented on the improved CEA test rig (PEV) to determine the operating conditions and the needed power supply which allow to evaporate and calcine a 90 l/hr feeding flow-rate ensuring the elaboration of a satisfactory calcinate that must:
   - be chemically homogeneous,
   - be sent continuously to the melter furnace,
   - have an homogeneous and satisfactory particle size.

   These test campaigns revealed that the heating system was the limiting factor.
2. New half-shell technology has been designed and manufactured in order to improve efficiency, by changing heating wires and insulating material. With this improved technology, thermal losses are reduced, half-shell strength is increased and maintainability is facilitated.
3. At first, these half-shells have been implemented and tested successfully in the CEA testing rig PEV.
4. In 2003 these new technological half-shells were implemented in the industrial R7/T7 AREVA/COGEMA calciners at normal operating conditions i.e. with a 76 l/hr HLW.

Calciner Modelling

In the meantime, a hydro-thermal model has been developed by SGN to describe the thermal behaviour of the tube and to analyse the role and interactions of various designs and operating parameters.

This model can determine optimal set points parameters for several kinds of operating conditions, by ensuring complete evaporation and efficient calcination and by avoiding film boiling occurrence or strong tube thermal stresses, phenomena which become more penalising at higher temperatures.

The model is based on the Aspen Custom Modeler ® solver, and among the other physical parameters accounted for by the model, one can list:

- Gravity flow of the product along the tube,
- Evaporation of the water from the feed solution,
- De-nitration and reduction reactions (38 reactions taken into account),
- Heat dissipation in the ½ shells,
- Longitudinal thermal expansion for the tube,

- Rotation of the calciner tube.

In order to qualify the calciner model, simulation results (power, temperatures, tube expansion) were compared to those measured on the PEV platform at various flow-rates and in various conditions. Those tests on the PEV platform were performed while actually measuring the tube temperature using 8 thermocouples positioned along its length, and the temperature on the inside face of the ½ shells. The other recorded parameters were the power delivered by the resistors and the feed-rate. The major results of these temperature comparisons are presented in Fig. 3 for different operating conditions (stand-by and HLW feed).

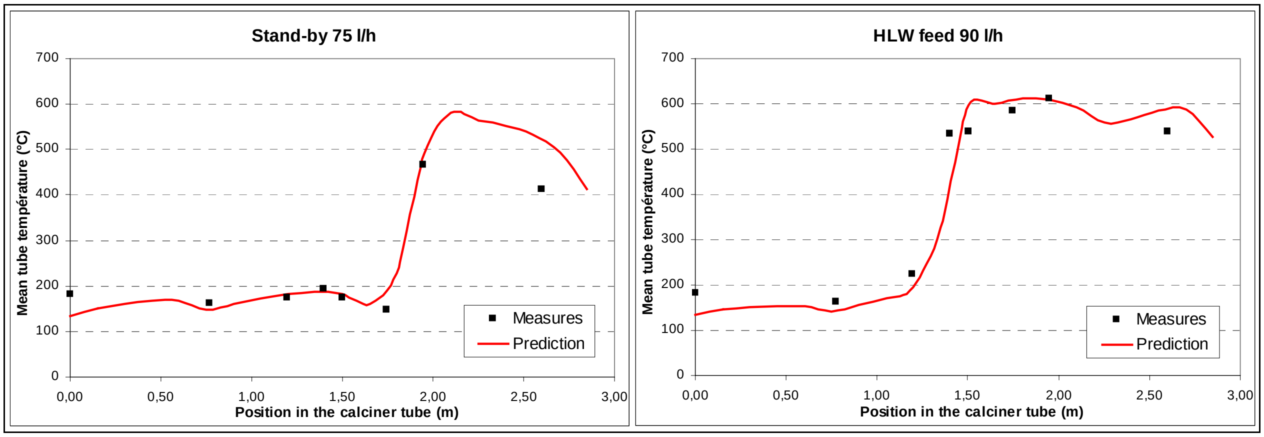

Fig. 3. Calciner model qualification - Comparison of measured and calculated calciner tube temperatures.

Also the simulation results (power, temperatures and tube expansion) agreed well with those measured on the various installations. For example, measured and predicted powers for R7 with a feed rate of 73 l/h and for T7 with a feed rate of 73 l/h are compared in Table I.

Table I. Calciner Model Qualification - R7 / T7 Half Shell Powers.

		R7 73 l/hr		T7 73 l/hr	
		Measured	Calculated	Measured	Calculated
Power delivered by the resistors (kW)	Zone 1	31	30.7	30	30.8
	Zone 2	24	23.6	24	20.8
	Zone 3	3.6	6.3	8.5	7.7
	Zone 4	5.4	5.9	6	5.2

ENHANCEMENT PROGRAM

Specific Organization

In order to perform the qualifying R7/T7 vitrification lines test campaign at the increased flow-rate of 90 l/hr, AREVA/COGEMA, CEA and AREVA/SGN have prepared a specific organization:

- Facility operation documents, explaining the enhancement program and including equipments set points for each operating conditions planned, have been written and diffused to the operators.
- AREVA/COGEMA lines shifts supervisors and operators have been informed of the industrial problematic and trained to manage successfully the enhancement program.
- Safety authorities have been informed.
- A team of vitrification experts from each entity has been involved in:
  - the project implementation, before the beginning of the enhancement campaign,
  - the parameters monitoring and checking (i.e. those that could be impacted by this throughput modification) during the campaign,
  - and the analysis of the functioning change impact after the campaign.

Campaign Layout

The preparation of the enhancement of the vitrification throughput required:

- The choice of the line on which the enhancement should be performed.
- The checking of equipments compatibility with the new functioning conditions (HLW feeder controls, calciner power supply checking, threshold parameters monitoring …).
- The inventory of normal conditions parameters that permit during the campaign the analysis of the increased flow-rate impact.
- The adaptation of the calciner model at the line configuration in order to specify all the calciner set points at the line shifts supervisors until the campaign start in terms of half shells power supply and stand-by operating conditions, according to CEA operating recommendations.

Campaign Program

The increase of the HLW flow-rate is realised by step from 76 l/hr up to 90 l/hr. For each step, calciner heating parameters are given by the adapted model. Minimum duration of each step is set at two weeks, to evaluate sufficiently campaign effect on equipments and off-gas treatment.

Sensitive parameters that can be followed with more attention are defined and sample requirements are also indicated to the AREVA/COGEMA laboratory in terms of:

- location: feeding, condenser and NOx column
- and frequency: each week

The samples analysis is realised, as the program advance, in order to determine the impact on equipments efficiency by determining their decontamination factors evolution.

A work session is planned at the end of each step to analyse all these parameters, and to decide if the next step can be realised, i.e. without negative effect on equipments behaviour, nor impact on activity releases.

RESULTS AND DISCUSSION

Production Records

The campaign has been performed on line B of the R7 facility, from February 14 to May 27 2005. The target flow-rate of 90 l/hr has been reached in three steps: 82 l/hr, 86 l/hr and finally 90 l/hr. During this period, 105 glass canisters have been produced (corresponding to approximately 1.6 millions TBq immobilized in 40 tons of glass). The evolution of canister production is presented in Fig. 4.

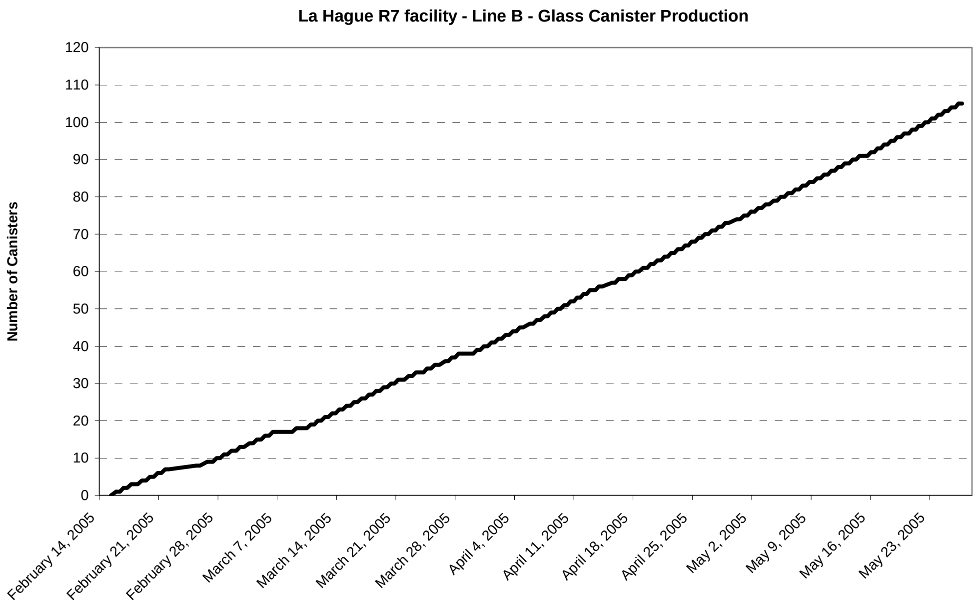

Fig. 4. Glass canister production during 90 l/hr campaign

From an industrial point of view this campaign has been successful, with satisfactory availability rate preserved during the whole test period, and without technical difficulties impacted by the functioning modification.

Table II presents the mean time required to fill a canister during each flow rate step.

Table II. Required Time per Canister Production during 90 l/hr Campaign.

Step	82 l/hr	86 l/hr	90 l/hr
Required time per canister production	26 hours	21.2 hours	19.1 hours

Calciner Behaviour

This equipment, modified in order to make this campaign possible, was particularly observed during this campaign, by checking regularly monitored parameters and by controlling sharply the evaporation length.

The calciner heating system behaviour agreed with predictions (the difference observed between measured and predicted powers was 1 kW or less for all zones and all flow-rates). It ensured evaporation without film boiling problems and calcination without clogging problems.

Off-gas Treatment Behaviour

The pressure regulation all along the off-gas treatment line has been controlled without particular difficulties during the campaign. The increase of off-gas rate did not affect equipments behaviour.

Based on analysis samples taken during each flow-rate steps, Table III shows the average decontamination factors [DF] of beta gamma activity in the off-gas system.

Table III. Evolution of Average Decontamination Factors [DF] of beta gamma Activity.

	76 l/hr [a]	82 l/hr	86 l/hr	90 l/hr
Calciner / Melter / Dust scrubber DF [b]	100	121	147	116
Condenser DF	200	230	143	183
From Calciner to Condenser DF	20000	27800	20900	21100

[a] Usual values at this flow rate.
[b] DF: for each equipment considered, beta gamma activity in the inlet divided by beta gamma activity in the outlet.

The results of this campaign are satisfactory from a technical point of view; all the line equipments behaviours have been in accordance with expected ones. The decontamination factors have not been impacted by the flow-rate increase, reaching during the whole campaign a good level of decontamination.

CONCLUSIONS AND PATH FORWARD

The 20% increase of the calciner feeding flow-rate realized in order to pursue in in-line vitrification in spite of increasing fuel burn-up, have been tested successfully in the La Hague R7 facility by the means of an efficient achievement of R&D program anticipated from 1998. This industrial improvement can be now implemented on the others R7 lines and then on the T7 facility.

This industrial successful campaign is another example of the La Hague continuous improvement program performed in order to enhance the vitrification throughput.

From Piver to T7, feedback from hot operations and the long-term R&D programs conducted jointly with the CEA have helped to continuously improve the vitrification process in all of its aspects (glass formulation, process, associated technologies, operations and maintenance).

The R7 and T7 vitrification facilities, in-line with AREVA/COGEMA’s two major commercial La Hague reprocessing plants, have had outstanding records of operation, not only from the standpoint of total glass production and plant availability but also with respect to safety (doses to personnel), remote in-cell maintainability, and secondary waste generated, demonstrating the soundness of French vitrification design choices.

The next major milestone in the evolution of the vitrification process will be the deployment in few years of the new generation of induction melter technologies (CCIM) able to operate at higher temperature than 1200°C to open new field of operation in terms of waste to be treated, glass form formulation and glass throughput, now also limited by melter capacity.

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