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ROLE OF ELECTROMAGNETIC FILTER IN LIMITATING RADIOACTIVITY IN THE PRIMARY CIRCUITS OF LIGHT WATER REACTORS (*)

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ABSTRACT

High temperature electromagnetic filtration of particulate corrosion products can be carried out with discharges up to 5 % of the cooling flow rate. It allows efficient extraction of particulate matter with rate constants required for considerable reduction of activable crud deposition in the core. The paper holds a review of the preventing operation in the primary circuit of a PWR, and reports experimental results of efficiency measurements with an electromagnetic filter set in out-of-pile and in-pile pressurized water loops.

The notable efficiencies towards radioactive fine grain and colloidal matter justify more extensive reactor scale application experiments.

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I. INTRODUCTION

In light water reactors primary circuits, only filtration with an adequate rate seems to be able to reduce considerably the radioactivity transportation by minimizing the crud deposition in the core, and then in the out-of-core components after the corrosion products and the radio-nuclides formed by neutron activation of the deposits have been released from the core.

Presently, water, used as coolant fluid in reactors, is purified by ordinary filtration followed by demineralization with ion-exchange resins. However the synthetic resins and the conventional filter materials cannot withstand high temperatures (not more than 70°C for the resins), and thus the cooling down of water before passage through resins beds is necessary. Under these conditions, this water purification is not possible with important output without inducing a prohibitive decrease of the thermodynamical yield in the power plant. Consequently, only a small fraction (about 0,05 %) of the main flow is treated. Moreover, the very fine particles in suspension and colloidal forms of corrosion products are not held back. The crud and activated products mostly consist of magnetite (Fe_2O_3), but also contain cobalt-60, cobalt-58, manganese-54, iron-59 and chromium-51. As the solubility of these various products is very slight, most of the activity is carried by particules in suspension. Magnetite is ferromagnetic, and the other constituants of the corrosion products are generally included with iron in a ferritic cristal lattice, ferromagnetic too. The size of the particles is submicronic.

II. EFFICIENT FILTRATION OF CORROSION PRODUCTS IN THE PRIMARY CIRCUIT

II.1. Required conditions

If the deposition of corrosion products has to be restricted, the filtration of crud particles must be quicker than

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their deposition.

In other words, the core acts as a filter, and the solution is therefore to place a more efficient filter in the circuit.

Fig. 1 represents an approximative comparison between the rates of water-borne particle disappearance due to deposit formation and different filtration rates, neglecting stationary corrosion and release. Current data were used for the deposit formation rates, which are very fast [1] [2]. It can be seen that an efficient filter must treat up to 1 to 5 % of the main flow rate, if the stationary concentration of insoluble corrosion products has to be reduced sufficiently, and their activation avoided by holding back most of them before they are deposited in the reactor core.

To treat such amounts of water, the filtration must be carried out at high temperature (around 300°C) and therefore under pressure (150 bar). The filtration process must also :

- introduce little pressure drop into the circuit,
- be efficient with suspensions of very fine particles (smaller than one micron) and colloïds
- be able of cleaning on line if possible, producing relatively little effluent.

These requirements severely narrow the choice of filtration methods. Two processes, one using the magnetic properties characteristic of the corrosion product suspensions, and the other the filtering effect of graphite granules, have been examined. The fast electromagnetic filtration is now by far the most developed, in spite of the need for a powerful electromagnet. During the International Conference on Water Chemistry of Nuclear Reactor Systems held last fall in Bournemouth, several papers have been given on this subject [3, 4, 5, 6, 7, 8, 9] and now the potentialities of the electromagnetic filter in high temperature filtration are well recognized. Research in primary circuit operating conditions is going to be accelerated. The present paper purposes to describe some recent results in high temperature efficient filtration of radioactive corrosion products.

II.2. Distribution of cruds between the core and the filter

In the primary circuit of volume V, the concentration C of suspended corrosion products, when the water is filtered with a discharge rate D, follows the equation :

$$\frac{dC}{dt} = \frac{P}{V} - \frac{K}{V} C - E \frac{D}{V} C \qquad (1)$$

where P represents the rate of crud release by corrosion
K the rate constant relative to crud deposition in the core
E the efficiency factor of the filter defined by the ratio
$\frac{C_i - C_o}{C_i}$ where C_i and C_o are the concentrations of particles at filter inlet and outlet.

The concentration C approaches a limiting value expressed by :

$$C_\infty = \frac{p}{a} \qquad (2)$$

when $p = \frac{P}{V}$ and $a = \frac{K}{V} + \frac{ED}{V}$

is written, and if C_o is the initial concentration of crud in the cooling water, the solution of equation (1) becomes :

$$C = C_\infty + (C_o - C_\infty) e^{-at} \qquad (3)$$

The quantity of crud retained by the filter is :

$$\int_o^t EDCdt = ED \left[C_\infty t + \frac{C_o - C_\infty}{a} (1 - e^{-at}) \right] \qquad (4)$$

If it is supposed that the core is by far the most efficient trap of crud in the primary circuit, the highest quantity of corrosion products deposited there, and susceptible to activation is :

$$\int_o^t KCdt = K \left[C_\infty t + \frac{C_o - C_\infty}{a} (1 - e^{-at}) \right] \qquad (5)$$

Since the time t expressed in hours is always long, equations (4) and (5) can be approximated by the following values of :

$$\int_0^t EDC\,dt \approx EDC_\infty t \qquad (4')$$

for the quantity of crud retained by the filter, and

$$\int_0^t KCdt \approx KC_\infty t \qquad (5')$$

for that deposited in the core.

In the case of a typical 900 MWe pressurized water reactor with three primary loops, having approximately 1400 to 1500 m^2 of 304 stainless steel and 14000 to 15000 m^2 of Inconel-600, with a steady corrosion product release rate of approximately 2 $mg.dm^{-2}$ per month for Inconel-600 and 1 $mg.dm^{-2}$ per month for stainless steel, a primary coolant volume of 360 m^3, a coolant circulation rate of 60000 $m^3.h^{-1}$ a 5 % fraction of which is filtered with an efficiency of 70 to 90 %, and a severe specified value of $C_o \approx 0{,}01.10^{-6}$ initial crud concentration at start, the limiting value of the crud expressed in particle concentration during operation becomes as low as :

$$C_\infty \approx 1{,}5.10^{-9}$$

and the corresponding quantities of crud picked up by the filter and deposited in the core amount in one year respectively to:

$$EDC_\infty t \approx 20{,}8 \text{ kg}$$

and

$$KC_\infty t \approx 8{,}9 \text{ kg}$$

if the value of the deposition rate constant is supposed to be about 10^{-4} [1] [2].

The quantity of 20,8 kg of crud retained in the filter is activated only to a slight extent, because this part of corrosion products is irradiated during a very short time of approximately 24 seconds in the core.

II.3. High temperature operation of the electromagnetic filter

Basically two antagonistic forces take effect on ferromagnetic particles while they are crossing the magnetized filter matrix, both influenced by the temperature :

- the magnetic force depending on the magnetic susceptibility of the particles, through their magnetization,
- the sweeping force depending on the strongly variable viscosity of water.

Fig. 2 shows a plot of the variation in the ratio of particle magnetization to water viscosity with temperature for magnetite and some other ferromagnetic spinels. The increasing value of this ratio characterizes an improved efficiency of an electromagnetic filter at elevated temperatures, far enough from the Curie point of the ferromagnetic species.

III. EFFICIENCY FACTORS OF AN ELECTROMAGNETIC FILTER

III.1. Reduced scale prototype filter

A reduced scale prototype electromagnetic filter has been tested at ordinary temperature, and on the out-of-pile pressurized water loop DOLMEN with synthetic magnetite and spinel suspensions, and then on the in-pile pressurized water fuel testing loop IRENE (OSIRIS reactor) with radioactivated corrosion products. The matrix of ferritic steel balls is 420 mm high and 78 mm in diameter ; the balls are 6 mm in diameter. The magnetic field is generated by a set of three coils supplied with direct current and the magnetic field attains 2700 gauss with 25 ampere intensity on each coil. The pressure drop in this filter matrix is especially low, about 120 mbar per meter balls height and per dm^2 cross-section at 250°C, with a specific water flow-rate of 5 $m^3.h^{-1}dm^{-2}$. Fig. 3 shows the filter fitted into the IRENE in-pile loop.

A new method has been developed to clean the filter matrix at working temperature of power reactor primary circuits; it makes use of turbulent rinsing during the direct expansion of the contained hot water to a water-cooled condenser. Several successive sequences, which proceed by filling the filter with hot water from the pressurizer, and expansion to the condenser, regenerate the filter with a yield higher than 90 %. This operation is slightly longer and more difficult than simple cleaning by cold rinsing, but avoids to lose time to cool and reheat the filter ; it also reduces the water volume containing the sludges, an appreciable advantage in view of their radioactivity. On the other hand, the high temperature rinsed matrix needs a much shorter filter pressure vessel, and reduces appreciably the cost of it.

III.2. Results

III.2.1. Fundamental studies on electromagnetic filtration efficiency

When tests were done with synthetic magnetite and spinel suspensions, the injections took place upstream from the filter. The particles were mostly smaller than 0,2 micron. The purpose of the efficiency measurements at ambiant temperature was mainly to study the quantitative role of the different physical filter constants, for instance the ball diameter, the length of the matrix, the applied magnetic field, together with the effect of the linear velocity of the suspension bearing water and the particle concentration. The results have been already reported elsewhere [8] [9].

III.2.2. High temperature application on the pressurized water loop DOLMEN

This loop is working at 250°C, with water flows up to about 2,75 m^3 per hour.

The filter efficiencies reach normally 97 % and can be as high as 99,5 % for magnetite or cobalt ferrite concentrations above 40 ppb. At low concentrations (25 ppb), they drop somewhat but still are around 72 %.

The efficiency of the reduced scale prototype filter at 250°C remains constant at more than 97 % if the linear velocity of the water varies from 12 to 20 $cm.s^{-1}$ (from approximately 36 to 60 $cm.s^{-1}$ in the matrix of 6 mm balls).

The filtration yield, the ratio:

$$\frac{C_i - C_f}{C_i}$$

of the difference in the initial C_i and final particle concentration C_f to the initial concentration in the circuit, is higher than 99 %. Since this characteratic is not a kinetic quantity, it is better to specify the time needed for the filter to remove a fixed fraction of the suspension under the working conditions. The filtration of one gram of suspension with such a yield takes less than 20 mn . One can also determine in this way the filtration half-life, or time necessary for the filter to lower the concentration of an impurity to half its initial value.

III.2.3. Filtration of radioactive corrosion products in the pressurized in-pile water loop IRENE

The insoluble fraction of representative highly radioactive corrosion products in the circulating water, chromium-51, cobalt-60 and zirconium-95, are filtered with efficiency factors ranging from 80 to 95 %, with little difference when the concentration decreases at the end; yet zirconium is not ferromagnetic and probably not in a ferritic lattice with iron. Notably increased quantities of radioactive corrosion products are retained in the filter when the loop is slowly cooled down from 270°C to 100°C.

Further noteworthy results are obtained by the radiochemical analysis of the concentrated slurry after filter regeneration. During the cool-down of the loop, the filter picked up approximately 25 to 30 times more chromium-51 activity than during loop operation at steady temperature.

In the slurry, the other long lived radioactive corrosion products originating from stainless steel are iron-59, cobalt-58 and manganese-54.

Besides zirconium-95, they contain zinc-65 and also cesium-132, with traces of lanthanum, cadmium, cerium, hafnium and ruthenium. This latter result allows hopes of worthy efficiency towards fission products.

The clean-up of the primary water in regard to radioactive crud has consequently a most favorable effect during a load change, especially during reactor shutdown.

Nevertheless, the important quantity of non activated crud retained on the filter limits seriously the bulk of radioactivated corrosion products inventory of the primary circuit.

IV. DISCUSSION

As far as industrial application of electromagnetic filters on primary circuits of pressurized water reactors is concerned, some attention has to be called on a profit which can be brought in with very fast filtration.

The efficiency variation of the electromagnetic filter is approximately :

$$\log \frac{C_o}{C} = - \log (1 - E) = AL d^2 \chi \frac{1}{\nu} \cdot \frac{1}{v} \vec{H} \, \text{grad} \, \vec{H} \qquad (6)$$

where A is the kinetic constant of the filter comprising its physical constant characteristics like the volume fraction occupied by the matrix balls material, the ball diameter and the saturation magnetization particularly

χ the magnetic susceptibility of the crud particles
d their diameter
L the length of the matrix
ν the dynamic viscosity of water
v the linear velocity of water in the empty filter vessel
$\vec{H}$ the magnetic field in the matrix cavities
$\vec{\text{grad}}$ H the field gradient locally achieved in the matrix.

When the kinetic constant A is calculated in case of the reduced scale prototype filter operated on the DOLMEN loop at 250°C, the relative variation of the filter efficiency with increasing water velocities in a constant length of filter matrix is shown in fig. 4.

Very high velocities are allowed, provided highest efficiencies are achieved through the highest value of the product of magnetic field with its gradient in the matrix. Of course, the effect of water velocity on the capacity of the filter has to be simultaneously taken into account.

The reduced size of an electromagnetic filter, operating at the highest possible water velocity and high temperature regenerated, lowers appreciably the cost of a high pressure filter vessel and of the powerful electromagnetic equipment. Projects with the aim of costs and benefits estimations concerning an industrial scale high temperature filter of primary circuit qualification have now to be conducted actively.

V. CONCLUSION

The electromagnetic filter loaded with a steel ball matrix offers an excellent efficiency towards corrosion products in the primary circuit of pressurized water reactors. It can be cleaned remotely and quickly on line. Since the pressure drop in the ball matrix is very low, it can accept the extremely large water outputs required for efficient competition with crud deposition in the core. Strong magnetic fields may be applied, favouring retention of the finest or less magnetic particles, they mainly ameliorate the efficiency at very high specific flow rates in the filter.

The electromagnetic filtration operating at primary circuit temperature seems the most direct continuously operating way permitting an appreciable reduction in the bulk inventory of radioactive corrosion products in primary circuits of nuclear power plants with light water reactors.

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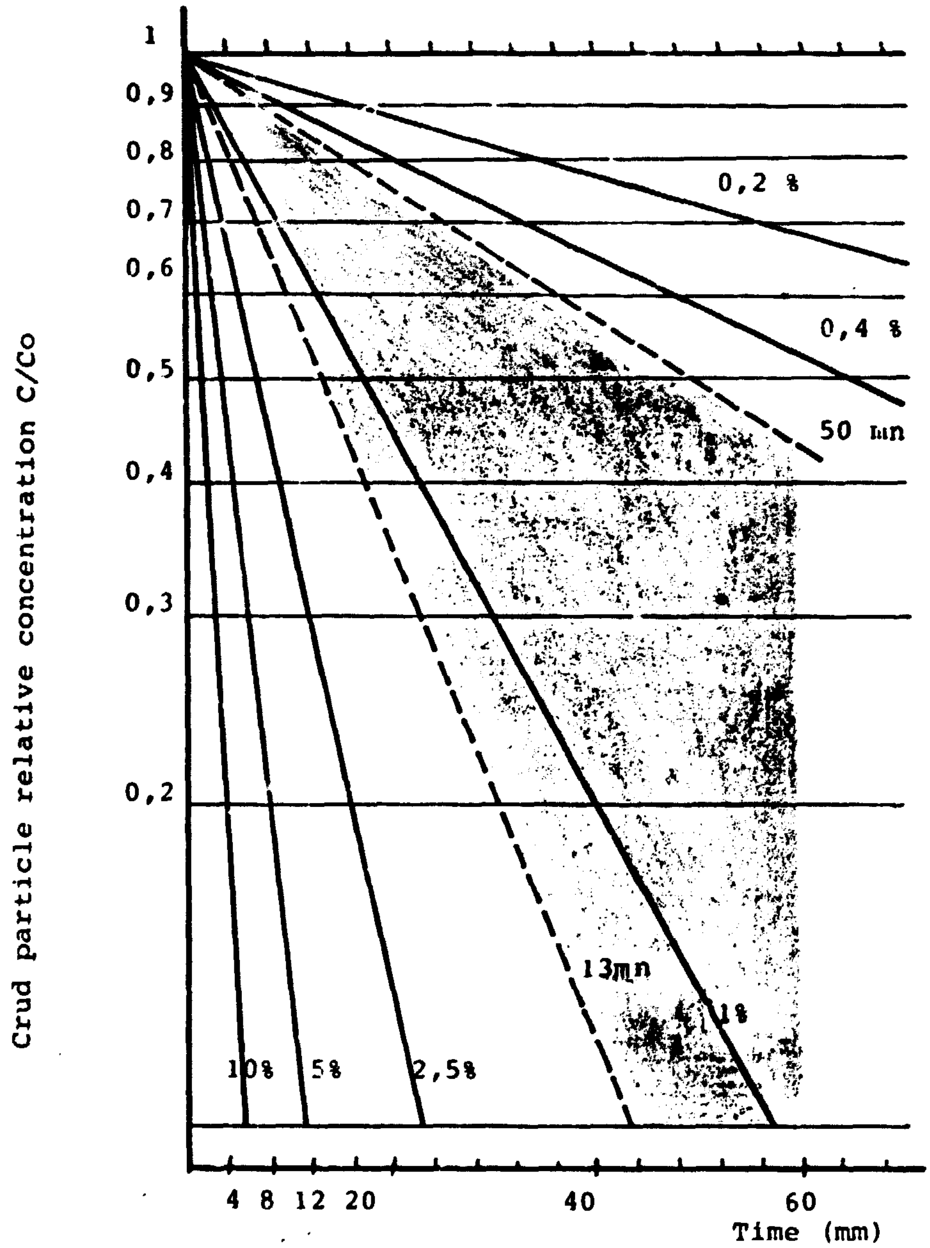

Fig. 1 - Approximate relative change of crud particle concentration in water versus time.

------- by deposition with half-lives between 13 and 50 mn.

——— by filtration with a rate expressed in percent of cooling flow rate.

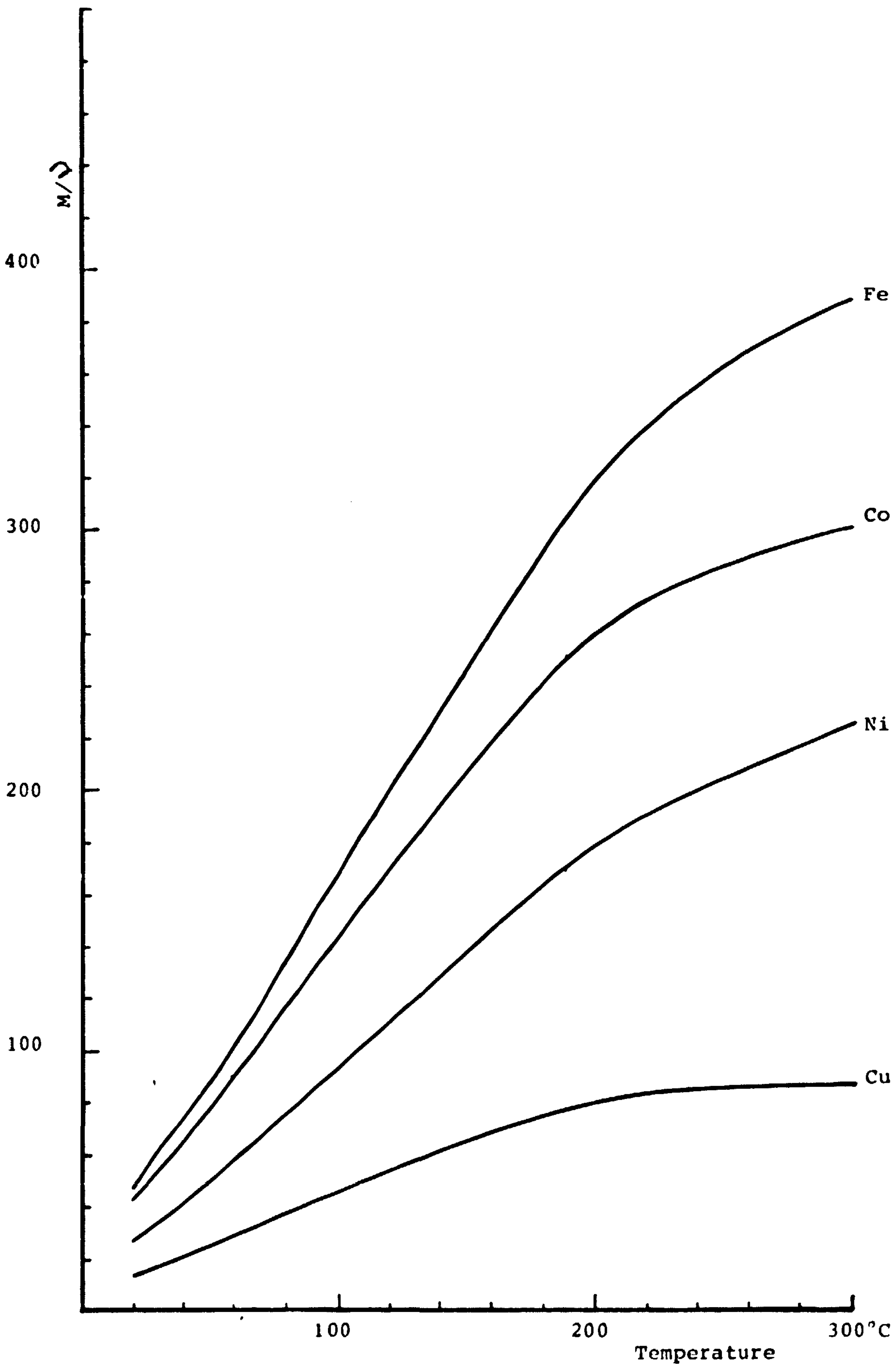

Fig. 2 - Variation of the ratio M/ν of ferrite magnetization to water viscosity with temperature.

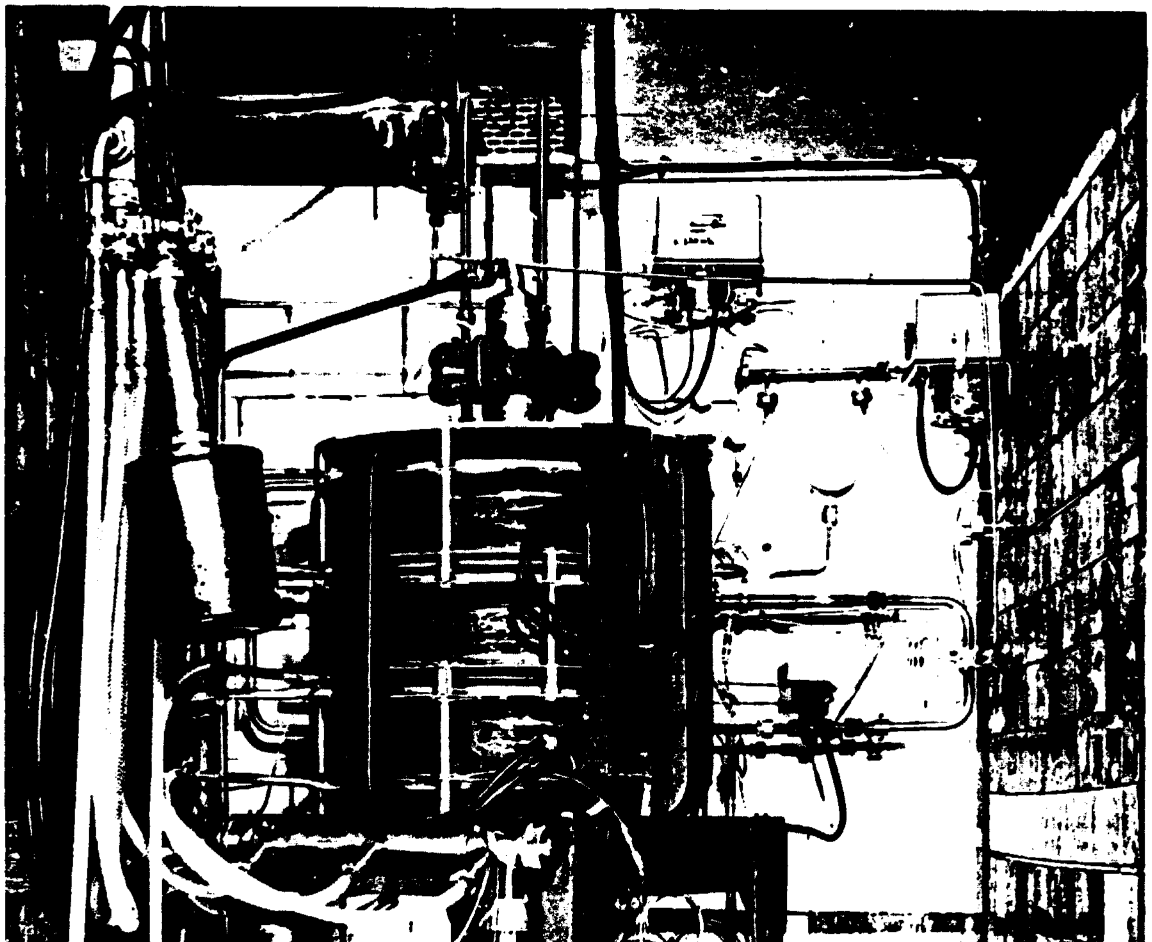

Fig. 3 - Electromagnetic filter fitted into the IRENE in-pile loop.

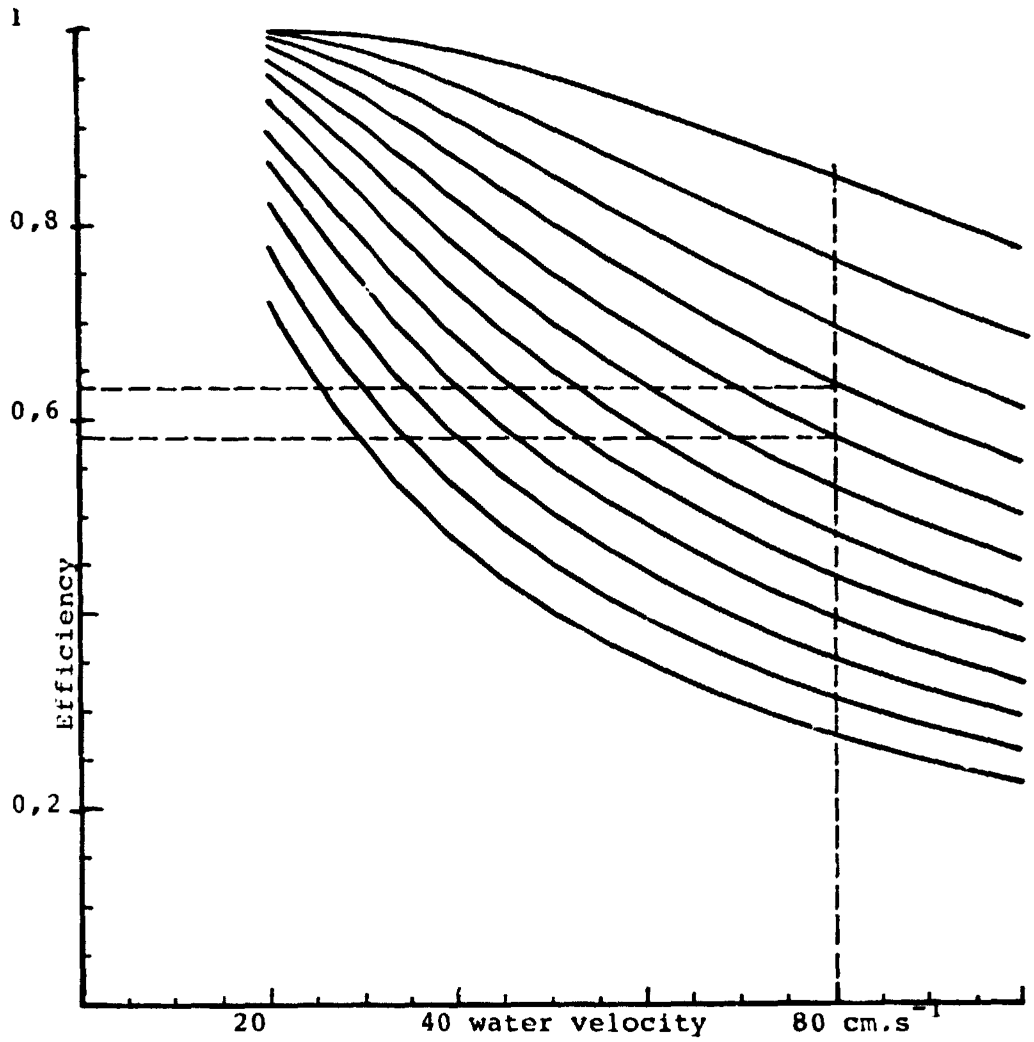

Fig. 4 - Variation of the efficiency factor with water velocity for different values of the kinetic filter constant.