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A SIMPLISTIC VIEW OF THE IODINE CHEMISTRY INFLUENCE ON SOURCE TERM ASSESSMENT

Luis E. Herranz
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ABSTRACT

The intrinsic characteristics of iodine make it a relevant concern as to its potential radiobiological impact in case of a hypothetical severe accident in nuclear power plants. This paper summarizes the major results drawn from a very simple but illustrative calculation exercise aimed at weighing how significant could be taking iodine chemistry in containment into account for source term assessments in case of a postulated severe reactor accident. The scenario chosen as representative of expected conditions in containment was LA-4 test of LACE programme. Several approximations and hypothesis concerning the scenario were necessary.

Iodine chemistry analyses were performed with IODE code, as long as thermalhydraulic and aerosol behaviour analyses, providing initial and boundary conditions for iodine calculations, were carried out with CONTEMPT4/MOD5 and NAUA/MOD5 codes, respectively. In general, the results obtained agreed qualitatively with the current knowledge on the area; from a quantitative point of view, one of the major results was that iodine chemistry on acidic conditions could provide a substantial increase in the leaked mass from containment under the postulated circumstances. Hence, this study underlines the need of including iodine chemistry in source term assessments

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1. INTRODUCTION

Chemical behaviour of some fission products affects strongly their evolution in case of hypothetical severe accidents. In this regard one of the major radionuclides is iodine. Its significant yield in the fission process, its volatility, its great reactivity and its potential radiobiological impact have made it one of the most important issues of nuclear research programmes in recent years. As a result several computational tools to predict iodine chemistry during severe accidents are being developed by different organizations.

This study was aimed at analyzing how iodine chemistry in containment can influence source term assessments in case of a postulated severe reactor accident. In order to do so, a coupled code system covering thermohydraulics (CONTEMPT4/MOD5), aerosol behaviour (NAUA/MOD5) and iodine chemistry (IODE) was used.

The scenario chosen was closely related to LA-4 test of LACE programme. Even though such an experiment was essentially devoted to examine specific aspects of aerosol behaviour, some assumptions and approximations allowed to use it as representative boundary conditions for iodine behaviour in case of a hypothetical severe accident. On the other side, such an approach made meaningless any comparison between predictions and experimental results.

2. SCENARIOS ANALYZED

The scenario analyzed can be seen as a modification of LA-4 experiment. LA-4 test is one of the six experiments which composed LACE (LWR Aerosol Containment Experiments) programme matrix [1]. The general objective of the project was to provide a suitable data base to validate thermohydraulic and aerosol codes. The tests were carried out in CSTF (Containment System Test Facility) vessel, whose essential features are summarized in Tables I and II.

TABLE I
Properties of vessel materials

Material	Conductivity (W.m ⁻¹ .K ⁻¹)	Heat capacity (J.m ⁻³ .K ⁻¹)
Phenolic paint	3.00 10 ⁻¹	1.60 10 ⁶
Carbon steel	5.12 10 ¹	3.85 10 ⁶
Insulator	4.67 10 ⁻²	7.23 10 ⁴

TABLE II
Vessel characteristics

Design pressure	517. KPa at 433.15 K
Nominal leak	2%/day at 69 kPa overpressure
Internal diameter	7.62 m
Height	20.3 m
Transference surface	741 m ²
Total volume	852 m ³

2.1. LA-4 Experiment

LA-4 experiment simulated typical accident conditions of a late failure of a LWR (Light Water Reactor) containment [2]. A couple of aerosol species were injected into the vessel in an overlapped manner: one soluble, cesium hydroxyde (CsOH), and another insoluble, manganese oxyde (MnO). The test consisted of several periods: heating-up (-50 - 0 min), aerosol injection (0 - 80 min), steady state (80 - 280 min), vent (280 - 600 min) and cooldown (600 - 5700 min). The injection phase was dividen into three subphases: CsOH input (0 - 30 min), CsOH and MnO input (30 - 50 min) and MnO input (50 - 80 min). Table III shows some of the thermal variables at -50 min (initial time of thermohydraulic analysis) and 0 min (initial time of aerosol and chemistry analysis).

TABLE III
Initial thermalhydraulic conditions

Time (min)	T _g (K)	T _w (K)	T _i (K)	P _T (kPa)	M _{sump} (kg)	X _{H2O}
-50.	315.7	315.0	315.7	107.0	950.	0.071
0.	371.1	329.3	366.8	212.5	1760.	0.443

2.2. Approximations and hypothesis

The major approximation taken concerned the aerosol species: the soluble aerosol (CsOH) was substituted by another soluble species containing iodine, cesium iodide (CsI). This compound is thought to be one of the most representative iodine species entering the containment in case of an accident. Rates and timing were maintained as in the original test, as well as the insoluble compound (MnO).

Chemical aspects were ignored in LA-4 test. This forced to take additional hypothesis and approximations to can analyze iodine chemical behaviour in a prototypical accident environment:

- A dose rate within the range expected in accident conditions [3] was imposed (1 Mrad/h).
- No organic substances were assumed due to the lack of data on this regard and to the unaccurate theoretical approach for the time being on formation and destruction of organic iodine compounds.
- Two extreme pH were considered to cover both acidic (pH=5) and alkaline (pH=9) conditions (both anticipated under accident circumstances [4]).

2.3. Modelling

Several codes were used sequentially to analyze the scenario: CONTEMPT4/MOD5 (thermohydraulics), NAUA/MOD5 (aerosol behaviour) and IODE (iodine chemistry). A diagram of code interfaces can be seen in Figure 1. Except for thermalhydraulic analysis, which took into account the heating-up phase, the calculations extended from 0 to 450 min (time at which leaks through the vent became null).

In order to evaluate the impact of iodine chemistry on source term assessment two types of calculations were performed: without and with chemical considerations. The second one consisted of two different calculations, one in acidic conditions and the other in alkaline conditions.

Next some specifics of modelling in each of the three areas covered are described. Input data not commented below were taken from ref. [2].

CONTEMPT4/MOD5 [5] describes the thermohydraulic response of a multicompartimental containment of a LWR under accident conditions in terms of thermodynamic variables such as temperature, pressure, relative humidity and so on.

The containment vessel was modelled as an atmosphere region ($\sim 851 \text{ m}^3$) composed by air and steam, and an aqueous region ($\sim 1 \text{ m}^3$ at -50 min), both enclosed in an only one cell which was surrounded by environment. In both zones a uniform temperature was assumed, being the interface temperature equal to the liquid one. Six structures were considered: cylinder, upper head, submerged lower head, non-submerged lower head and internal structures (in both sump and atmosphere). Uchida correlation was used to describe the heat transfer between atmosphere and surfaces contacting with, as long as liquid phase was assumed to be in equilibrium with the surrounding walls (a heat transfer coefficient of $57000 \text{ W/m}^2\text{K}^1$ was imposed). The vent from the vessel was modelled as a homogeneous gas flowing through an orifice.

NAUA/MOD5 [6] predicts aerosol behaviour within LWR containments during severe accidents. Their results are expressed in terms of airborne concentration, particle size and depleted mass. Themohydraulic input data were taken from CONTEMPT4/MOD5 output except steam condensation rate onto particles which was taken from a theoretical adjustment carried out in the frame of a european benchmark exercise on LA-4 test [7]. Concerning particle properties the main assumptions were: extention of size range from 10^{-4} to $5 \times 10^{-2} \text{ cm}$, partition of such an interval into sixty bins, spherical shape (shape factors equal to 1.0) and particle density reduced to 50% of its theoretical value for insoluble particles [8].

summarizes these and other thermalhydraulic results at the end of each test phase.

TABLE IV
Thermalhydraulic results (CONTEMPT4/MOD5)

Variables	Time (min)			
	0.	80.	280.	450.
T _g (K)	371.1	374.1	375.7	364.3
T _i (K)	329.3	339.1	351.8	346.6
T _w (K)	366.8	373.4	375.4	360.9
P _T (kPa)	212.5	271.8	285.8	105.6
P _{H₂O} (kPa)	94.1	105.0	111.0	61.53
P _{nc} (kPa)	118.4	166.7	174.8	44.1
RH (%)	100.	100.	100.	84.0
WCR (kg/s)	0.22	0.02	0.01	0.00
LR (kg/s)	0.00	0.00	0.45	0.00

The concentration of CsI particles followed the expected pattern (Fig. 4). A steep slope was obtained during its injection up to attaining a maximum of 2.76 g/m³ at ~50 min. Next, it diminished in a exponential way till the vent start. At 280 min the decay was accelerated by the leaks.

The major depletion process was settling. Relative humidity conditions allowed an intense steam condensation onto particles which made them grow and, subsequently, be removed by sedimentation. In Figure 5 settled mass (M_{sed}) and settling rate (V_{sed}) versus time are plotted. It can be readily seen that most of settling happened between 50 and 100 min, reaching the maximum intensity at 60 min. At 450 min the total mass settled is ~2380 g. Table V collects results of the most significant variables concerning aerosol behaviour.

TABLE V
Aerosol behaviour results (NAUA/MOD5)

Time (min)	Concentration (g/m ³)	Sedimentation		Leaked Mass (g)
		Mass (g)	Rate (g/s)	
50.	2.745	4.58 10 ²	2.37 10 ⁻¹	0.
280.	4.521 10 ⁻¹	2.24 10 ³	7.32 10 ⁻²	0.
450.	3.961 10 ⁻²	2.36 10 ³	1.82 10 ⁻³	2.21 10 ²

IODE [9] simulates iodine chemical behaviour within a PWR (Pressurized Water Reactor) containment under accident conditions. Its results were expressed in terms of species concentration in aqueous and gaseous phases.

The chemical behaviour consisted of six chemical reactions:

- Hydrolysis of molecular iodine (I_2)



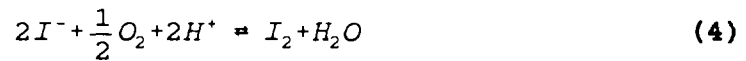
- Ionization of hypoiodous acid (HOI)



- Disproportionation of hypoiodous acid



- Thermal oxidation of iodide (I)



- Radiolytic oxidation of iodide (I)



- Radiolytic reduction of iodate (IO_3^-)



In Figure 2 the chemical model is plotted. A total of four flows were taken into account: I_2 transfer from sump to atmosphere (ϕ_1), I_2 deposition onto surfaces by steam condensation (ϕ_2), I_2 diffusion towards the walls (ϕ_3) and I_2 leak through vents (ϕ_4).

3. RESULTS

A thorough description of every variable of each of three fields modelled is out of the scope of this paper. Attention is mainly focused on those variables closely related to iodine chemical behaviour.

Figure 3 shows gas, sump and wall temperatures as a function of time. Gas temperature (T_g) curve showed different slopes in each test period: a sharp rise during the heating-up, a moderate increase in the injection phase and an almost constant value up to the beginning of the leaks, during which an initial rapid decrease is then softened. Sump temperature (T_s) exhibited a smoother evolution given that there were no heat sources injected into it and, hence, T_s variation depended upon energy exchanges with atmosphere and surfaces. Wall temperature (T_w) showed similar trends to T_g , and only during the heating-up and the vent periods there were differences caused by thermal inertia of the surfaces. Table IV

The results on iodine chemistry were dramatically different depending on pH. A couple of iodine scenarios were studied: the first one, characterized by an acid pH (QA); the second one, by an alkaline pH (QB).. In both cases I mass input rate into the sump was derived from CsI settling velocity.

Figure 6 shows the evolution of total iodine concentration in the sump along with those of each species (I_2 , HOI, I^- , IO_3^-) at pH=5. As can be seen, iodine was mainly as I^- and I_2 . Both species concentrations increased considerably during the injection period. Their maxima were attained during the stationary: at ~200 min for I^- (~ 3×10^2 g/m³) and at ~235 min (~ 3×10 g/m³). On the contrary, IO_3^- concentration increased slowly along the time but, as in the case of HOI, it was so low that it hardly affected the total iodine concentration. I_2 concentration in the gas phase followed that of liquid phase since they were linked each other by the aqueous-gaseous transference; at ~260 min $I_2(g)$ reached a maximum (~ 3.5×10^{-2} g/m³) and 20 min later it experienced a sudden decrease due to the vent. Table VI shows the results obtained at the end of each phase.

In Figure 7 the different flows affecting $I_2(g)$ at pH=5 are plotted. As can be seen, the most significant transfereces occurred between the sump and the atmosphere (ϕ_1) and between the atmosphere and the walls (ϕ_2) (interaction with paints). The amount of iodine carried to the walls by steam was negligible, as well as leak flow rate (even at 280 min).

TABLE VI
Iodine species concentration at pH 5

Time (min)	LIQUID PHASE (g/m ³)				GAS PHASE (g/m ³)
	I_2	HOI	I^-	IO_3^-	I_2
50.	6.15	$3.09 \cdot 10^{-3}$	$9.61 \cdot 10^1$	$4.97 \cdot 10^{-4}$	$2.10 \cdot 10^{-3}$
280.	$5.32 \cdot 10^1$	$2.12 \cdot 10^{-2}$	$2.81 \cdot 10^2$	$3.91 \cdot 10^{-1}$	$3.51 \cdot 10^{-2}$
450.	$3.17 \cdot 10^1$	$1.81 \cdot 10^{-2}$	$1.50 \cdot 10^2$	$8.53 \cdot 10^{-1}$	$2.50 \cdot 10^{-2}$

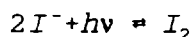
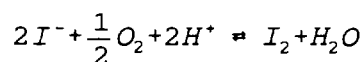
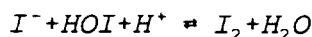
TABLE VII
Iodine species concentration at pH 9

Time (min)	LIQUID PHASE (g/m ³)				GAS PHASE (g/m ³)
	I_2	HOI	I^-	IO_3^-	I_2
50.	$1.23 \cdot 10^{-2}$	$5.68 \cdot 10^{-2}$	$1.04 \cdot 10^2$	$1.76 \cdot 10^{-1}$	$4.31 \cdot 10^{-6}$
280.	$3.14 \cdot 10^{-2}$	$6.21 \cdot 10^{-2}$	$5.68 \cdot 10^2$	$9.72 \cdot 10^0$	$3.10 \cdot 10^{-5}$
450.	$5.76 \cdot 10^{-2}$	$8.25 \cdot 10^{-2}$	$5.97 \cdot 10^2$	$1.95 \cdot 10^{+1}$	$4.17 \cdot 10^{-5}$

At pH=9 iodine speciation changed dramatically respect to the acidic scenario (Fig. 8). Ionic forms of iodine (I^- , IO_3^-) were predominant in the sump. Almost 100% of iodine existed as I^- (I^- concentration was more than one order magnitude above that of IO_3^-). Even though both increased steadily, I^- reached its order of magnitude at the beginning of stationary state as long as IO_3^- rised smoothly along all the time. Molecular species (I_2 , HOI) had negligible concentrations compared to I^- one. As for $I_2(g)$ the major observation referred to its very low concentration, being its fluctuations a consequence of existing flows (Fig. 9). Table VII collects the concentration of every species at the end of each period.

4. DISCUSSION

Once CsI came into the liquid it underwent ionization and iodine dissolved as I^- . Both at acid pH and alkaline one, I^- is then submitted to the following chemical reactions:



Namely, a certain fraction of I^- was transformed into I_2 . Among these reactions the most efficient way of converting I^- into I_2 was the radiolytic oxidation of I^- . However, in alkaline conditions the yield of I_2 formation through any of the three reations was quite low.

The iodine fraction transformed into I_2 was instantaneously hydrolyzed. The extention of this reaction depended also on pH. At acidic pH the continuous supply of I^- into the sump and the high concentration of protons (H^+) caused a noticeable I_2 build up in the sump. On the contrary, as H^+ concentration was low (basic solution) hydrolysis equilibrium shifted towards HOI formation, producing a decrease in I_2 concentration.

Another potential source of $I_2(l)$ is the radiolytic reduction of IO_3^- . However, this reaction was not crucial in any of the cases analyzed. In acidic conditions, the concentration of IO_3^- was too low and, thus, this reaction was negligible compared to radiolytic oxidation of I^- . In basic conditions the formation of IO_3^- by HOI disproportionation was favoured but the IO_3^- reduction was seen to be quite unefficient at high pH.

As to $I_2(g)$ concentration, discrepancies between acid and basic cases were a consequence of the aqueous chemistry of I_2 given that the only source of I_2 into the gas phase was its partition bewtween sump and atmosphere,



and that no chemical reactions were considered in the gaseous phase. The removal mechanisms of $I_2(g)$ were formulated as linear functions of $I_2(g)$ concentration whose rates

were defined in the input deck of IODE, so that differences in their intensity was essentially caused by those of $I_2(g)$ source from the pool.

In both cases the predominant species was I^- . However, as long as in acidic conditions its evolution reached a maximum, in basic conditions it tended to attain an asymptotic value. The most intense process which underwent I^- was the radiolytic oxidation, whose velocity is a function of pH,

$$V_{I^- \rightarrow I_2} \propto [H^+]^{0.25} \quad (7)$$

Such a dependence makes the conversion to I_2 more intense at low pH. At a certain time the I^- input rate into the sump did not compensate I^- losses by oxidation and I^- concentration began to decay. This situation did not exist at high pH, because the oxidation rate was lower than I^- input rate during all the transient.

Acid and alkaline IO_3^- concentration curves were qualitatively similar. However, they were dramatically different from a quantitative standpoint. The responsible process of IO_3^- formation was HOI disproportionation that is proportional to the square of HOI concentration,

$$V_{HOI \rightarrow IO_3^-} \propto [HOI]^2 \quad (8)$$

I_2 hydrolysis was the source of HOI, so that [HOI] can be written as,

$$[HOI] = \frac{K_{hyd} [I_2]}{[I^-] [H^+]} \quad (9)$$

Thus, it can be said that the formation of IO_3^- is inversely proportional to the square of H^+ concentration. So, a decrease in pH led to an increment of [HOI] that caused an iodate concentration much higher at basic pH.

A global comparison between acid and basic scenarios can be done through the overall partition coefficient of iodine:

$$H_T(I) = \frac{\sum [I]_l}{\sum [I]_g} \quad (10)$$

As it is readily seen H_T is a measure of system volatility: the higher H_T , the lower volatility. Figure 10 shows that the system volatility was much lower under alkaline conditions than under acidic ones (roughly three orders of magnitude).

From the safety point of view another variable worth to be analyzed is the iodine leaked mass. As leaked mass obeys a first order law in $[I_2(g)]$,

$$M_{Ik} \propto [I_2]_g \quad (11)$$

speciation at the beginning of vent period was crucial. Figure 11 shows that at low pH $I_2(g)$ was a relative important iodine compound as long as it was not at high pH. This fact and the performance of sump as a source of $I_2(g)$ during the vent (as a consequence of the shift of

partition equilibrium to the right), explains the quantitative discrepancy found between M_{ik} both cases (Fig. 12): at acid pH M_{ik} is approximately 25% higher than at basic pH.

Finally, Figure 13 compares the three cases studied. The curves belonging to the amount of iodine suspended as particles and the amount of airborne iodine both as particles and as gaseous form under basic conditions were practically equal. This fact indicated that basic conditions enhance stability of ionic forms of iodine in the sump, whose negligible volatility makes iodine to be efficiently retained once it comes into the sump. On the contrary, if both curves are compared to that of the total iodine suspended (particles and I_2) under acidic conditions a growing divergence from the end of the injection can be observed. Such a difference became more relevant as the vent started because, as said above, a fraction of the I_2 inventory in the sump was transferred to atmosphere trading off somehow the $I_2(g)$ loss through the leak path; so to say, under acidic conditions sump can behave as a source of I_2 into the atmosphere of containment vessel whenever the partition equilibrium is shifted to the right due to $I_2(g)$ removal.

The results discussed along the preceding paragraphs underline a well known statement [9,10]: pH as a crucial magnitude for iodine behaviour. Achievement of alkaline conditions in the containment sump would produce a substantial reduction in iodine source term compared to that expected under acidic conditions. This fact makes pH be one of the most important variables needed to be taken into account in the definition of accident management strategies into the containment aimed at reducing the potential radiobiological impact.

In addition, iodine nature as particles or gaseous species is a relevant factor in its filtration. Hence, another interesting point is that iodine to be emitted under acidic conditions has a large fraction of gaseous components (25%) as long as at basic pH most of iodine available to leak would be in particulate form. This fact should be considered as well when mitigative measures are being planned.

5. CONCLUSIONS

In this paper a simplistic view on how iodine chemistry in containment can influence the source term assessment in case of a hypothetical severe reactor accident has been summarized. The codes used were: CONTEMP4/MOD5, NAUA/MOD5 and IODE. In general, the results obtained agreed with the current knowledge on this area.

The major conclusions drawn from this study were the following:

- According to the scenario analyzed and under the hypothesis and approximations taken, the quantitative contribution of chemical factors to the leaked mass from containment could reach values around 25% if an acidic pH prevails in the containment sump from the beginning of the transient.
- Under alkaline conditions, containment sump behaves as a very efficient sink of iodine. Only a very small fractions is transferred from the sump to the atmosphere in form of volatile species (I_2, HOI), remaining most of iodine dissolved as ionic species (I^- and IO_3^-).
- Sump efficiency as iodine sink becomes much lower under acidic conditions.

An acid pH gives certain stability to molecular forms of iodine, whose volatility results in a substantial transport of these species into the containment atmosphere. This sump behaviour as a source of molecular species can effect significantly source term magnitude in case of a containment venting.

Finally, it can be concluded that an accurate estimation of source term in case of hypothetical accidents requires chemical considerations, in particular those concerning iodine behaviour in containment.

ACKNOWLEDGEMENTS

The authors are indebted to Inmaculada L. Díez and Antonia Martínez for their contribution to this work. Likewise, The authors wish to thank Commissariat à l'Énergie Atomique and Kernforschungszentrum Karlsruhe laboratories for making the IODE and NAUA/MOD5, respectively, available.

NOMENCLATURE

LR	Leak rate	RH	Relative humidity
M_{sed}	CsI Settled mass	T_g	Gas temperature
M_{lk}	Leaked mass	T_l	Liquid temperature
M_{sump}	Water mass in the sump	T_w	Wall temperature
P_T	Total pressure	V_{sed}	Settling rate
P_{H_2O}	Steam partial pressure	X_{H_2O}	Steam fraction
P_{nc}	Non-condensibles pressure	ϕ	Flow rate

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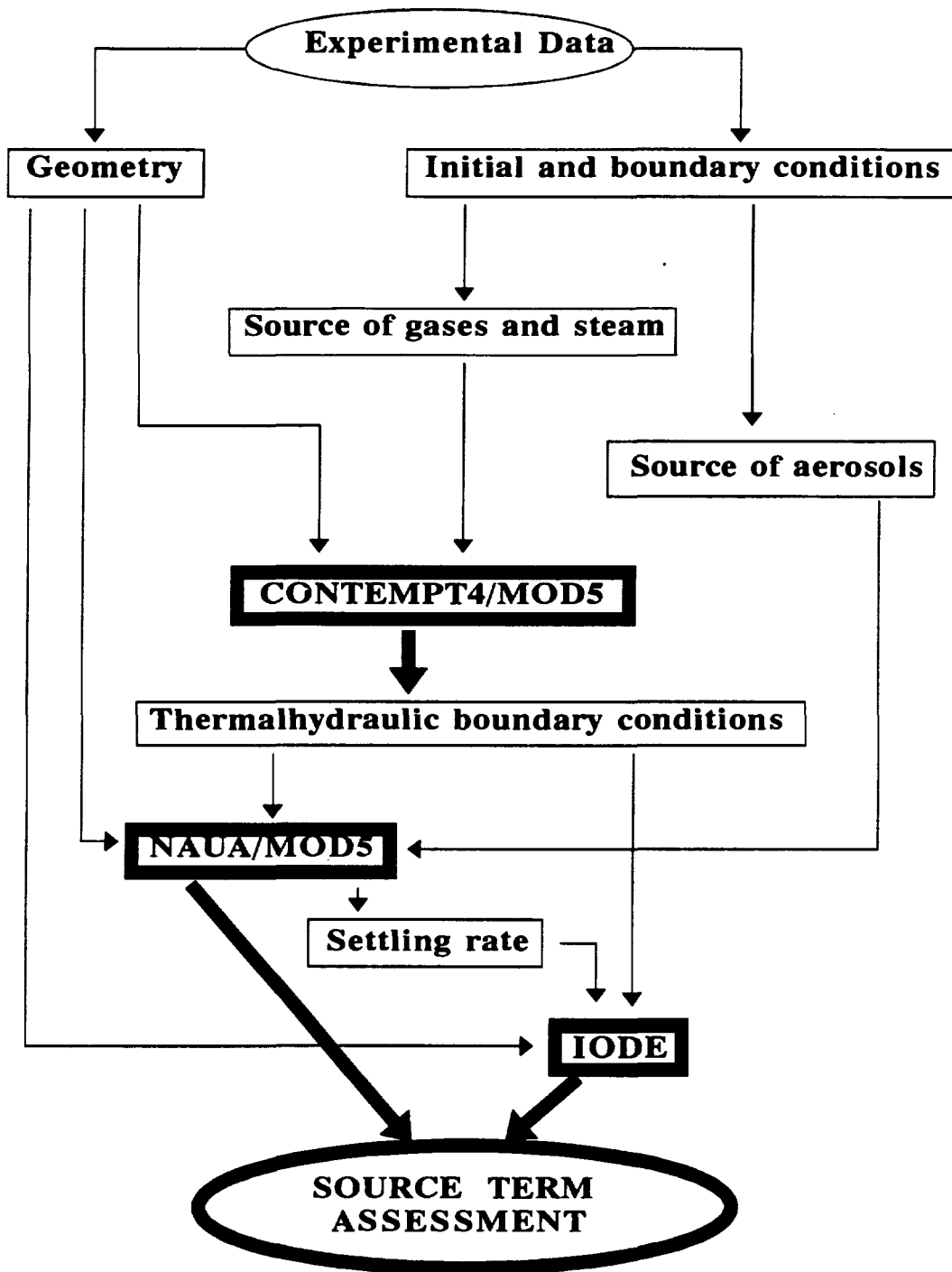


Figure 1. Diagram of code coupling

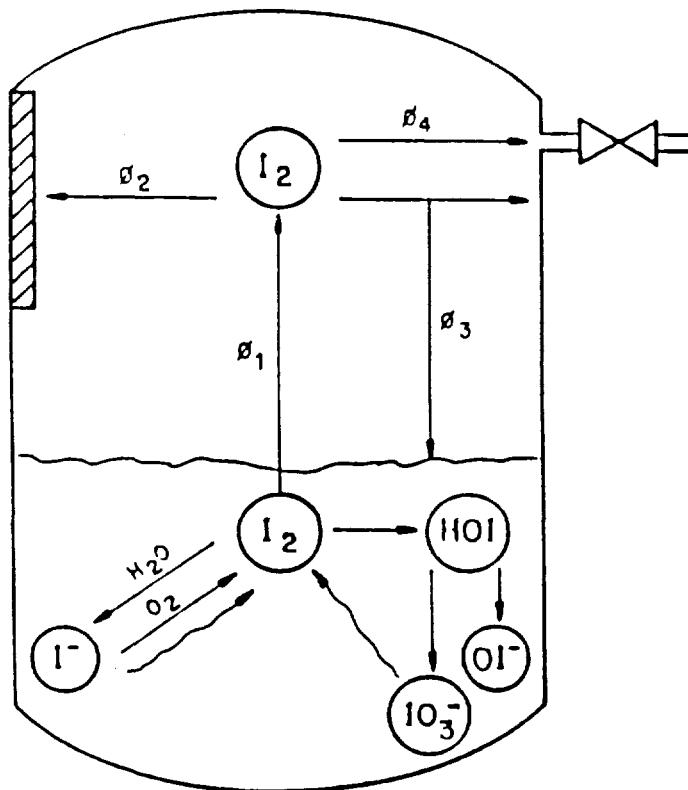


Figure 2. Chemical model of the scenario

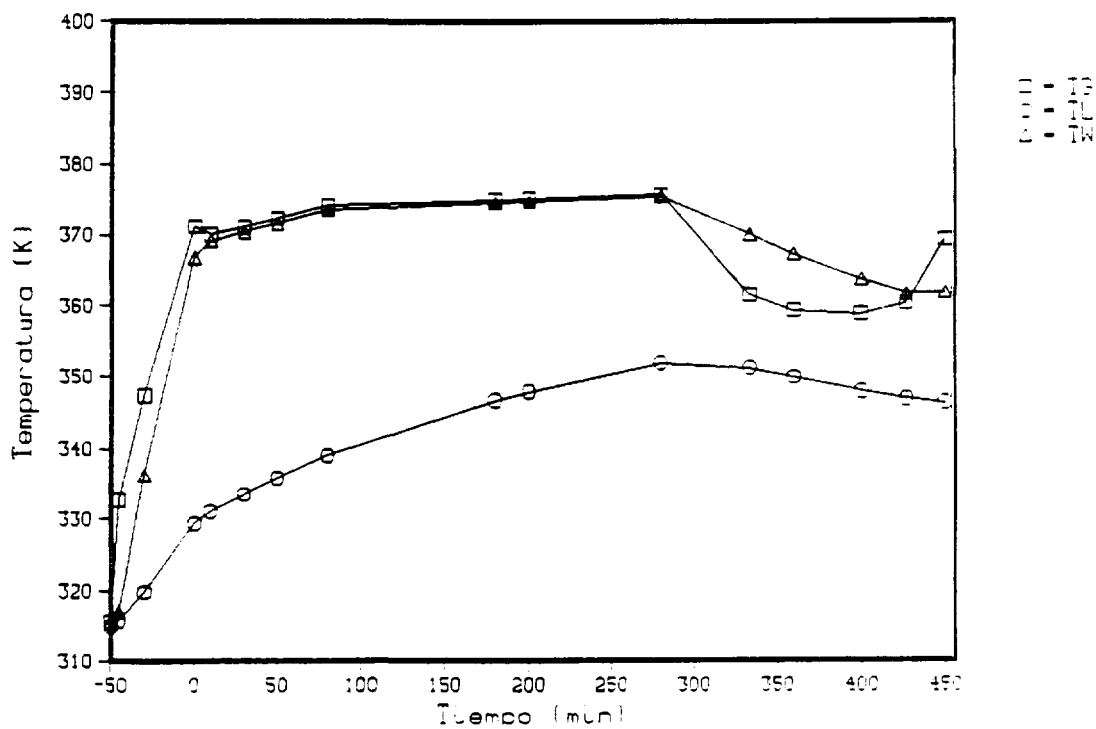


Figure 3. Gas, sump and wall temperatures vs time

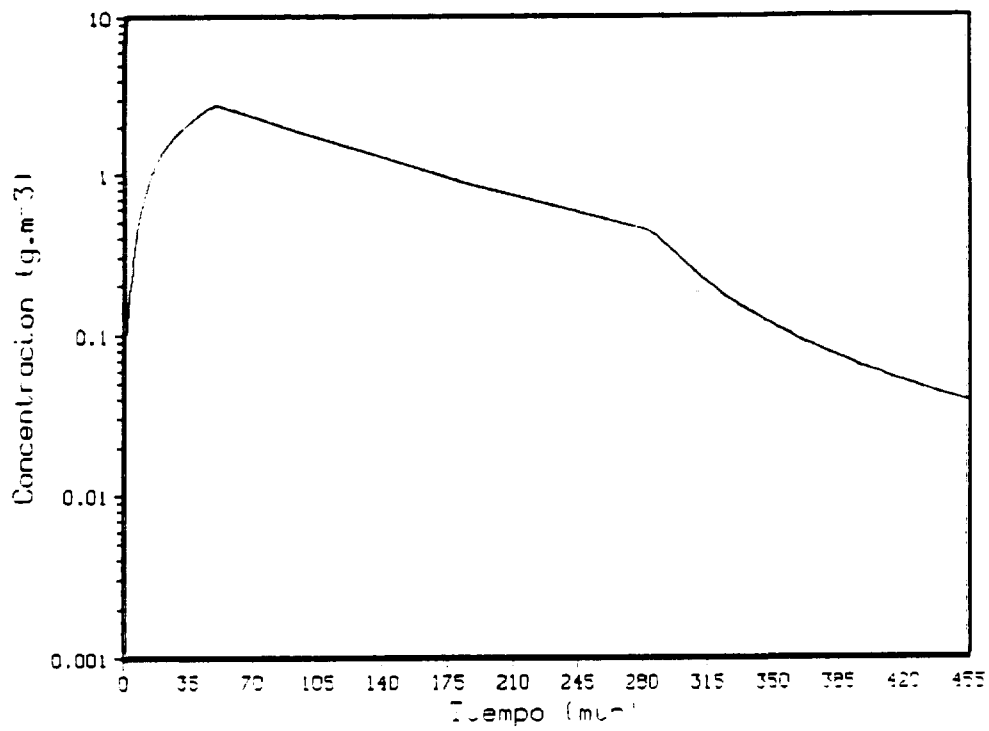


Figure 4. CsI concentration vs time

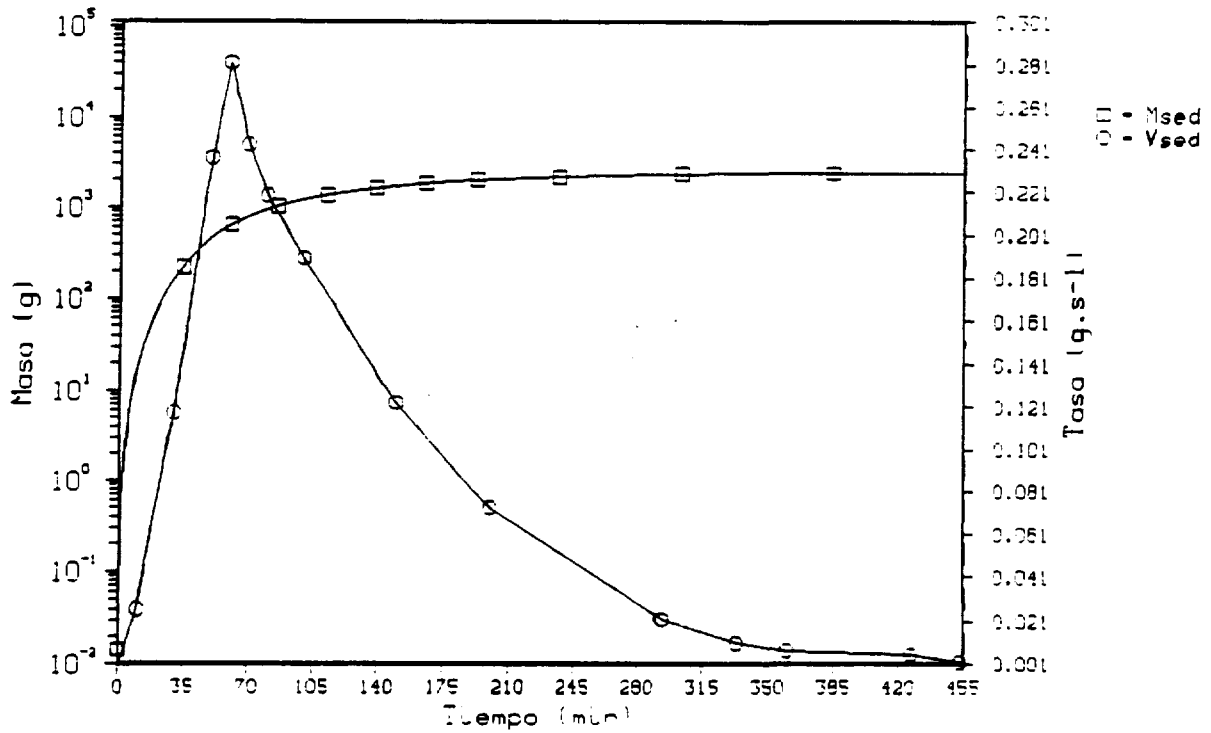


Figure 5. Settled mass and settling velocity vs time

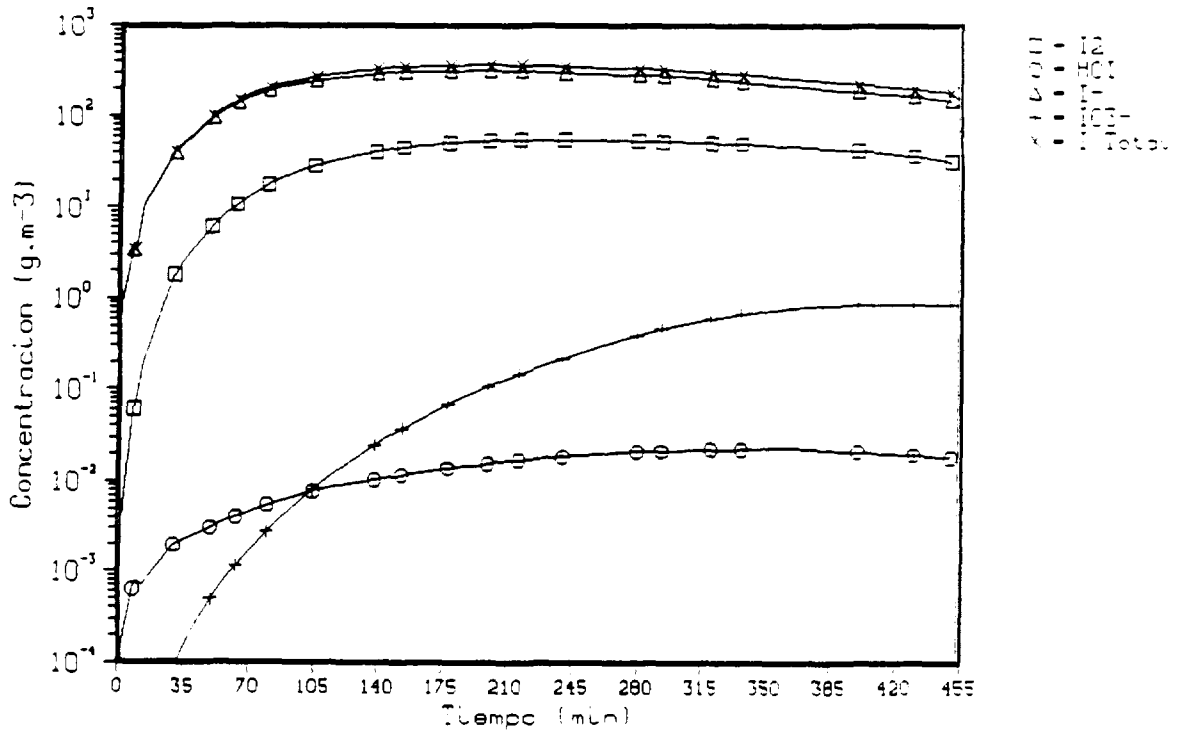


Figure 6. Iodine distribution vs time at pH=5

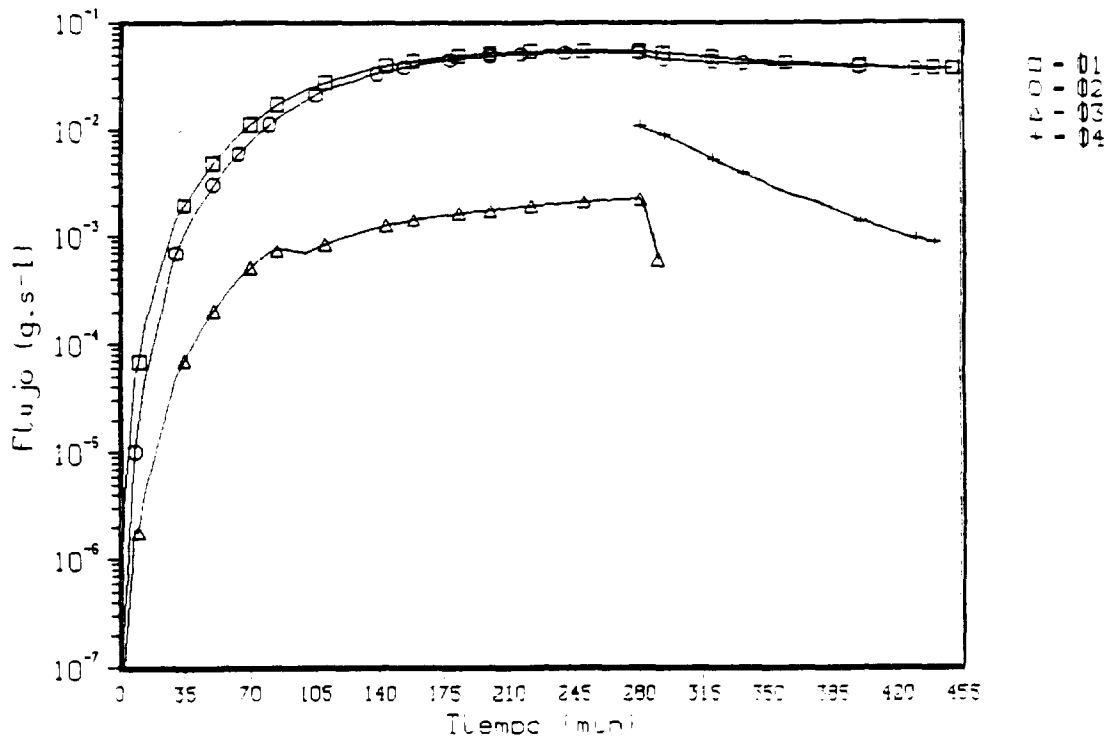


Figure 7. Flow rates affecting I₂(g) vs time at pH=5

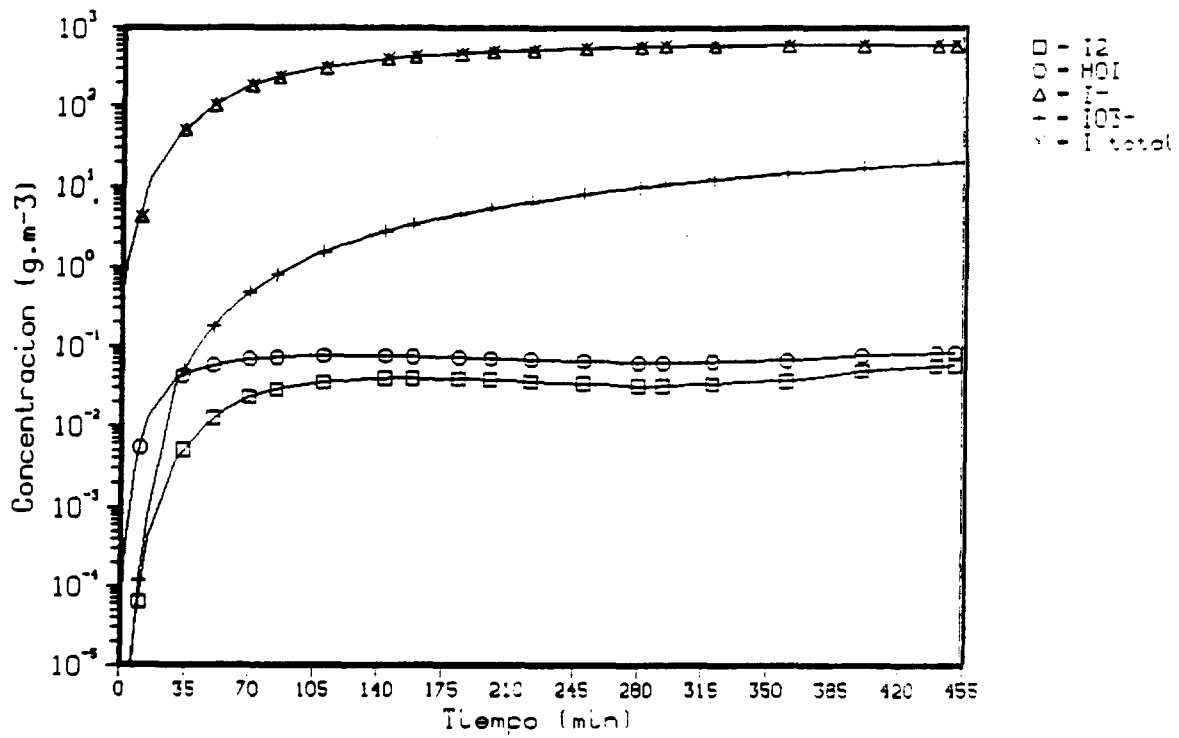


Figure 8. Iodine distribution vs time at pH=9

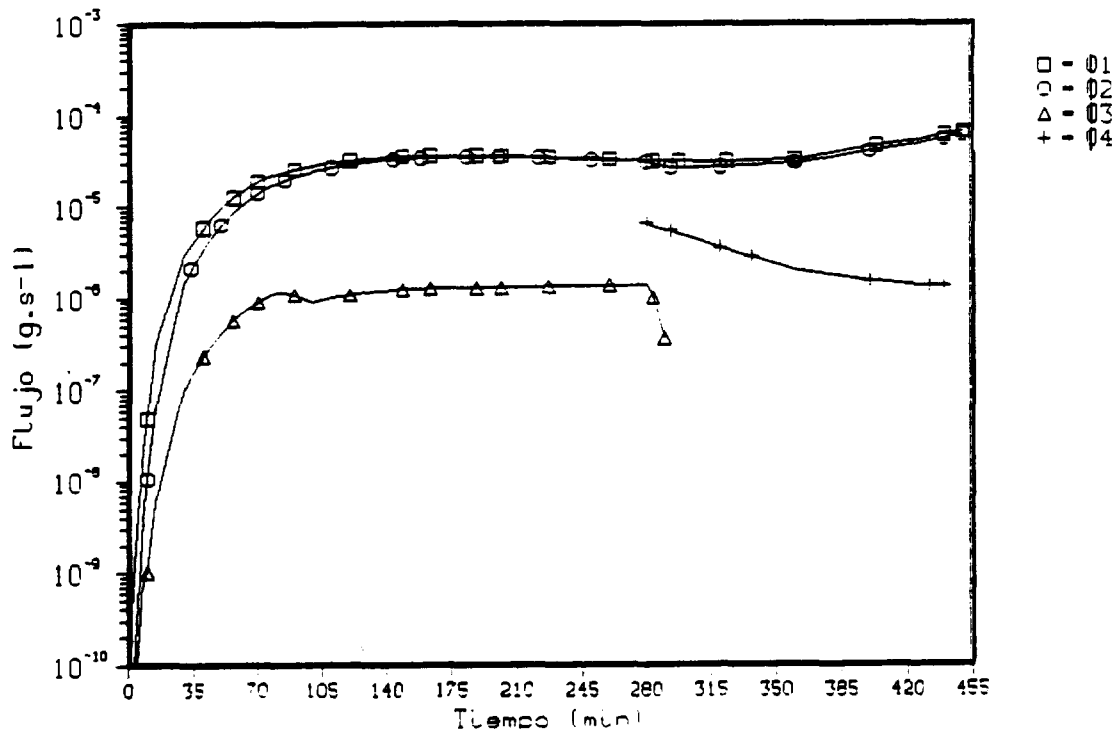


Figure 9. Flow rates affecting I₂(g) vs time at pH=9

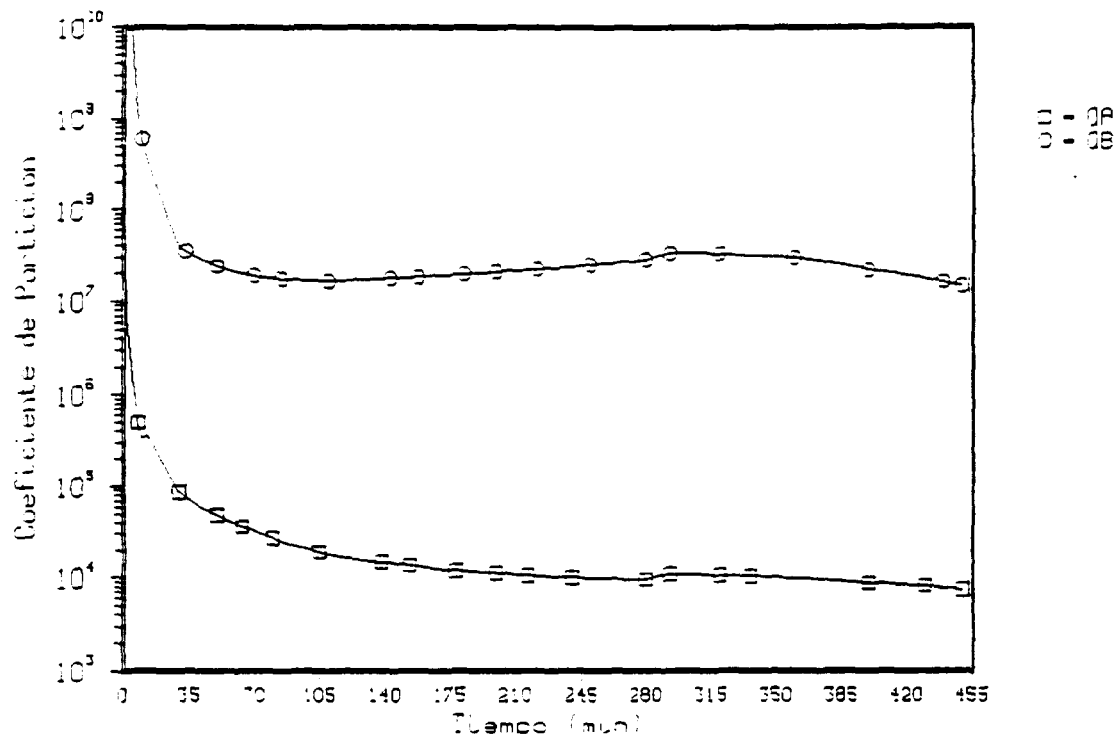


Figure 10. Iodine total partition coefficient vs time (QA - acid pH; QB - basic pH)

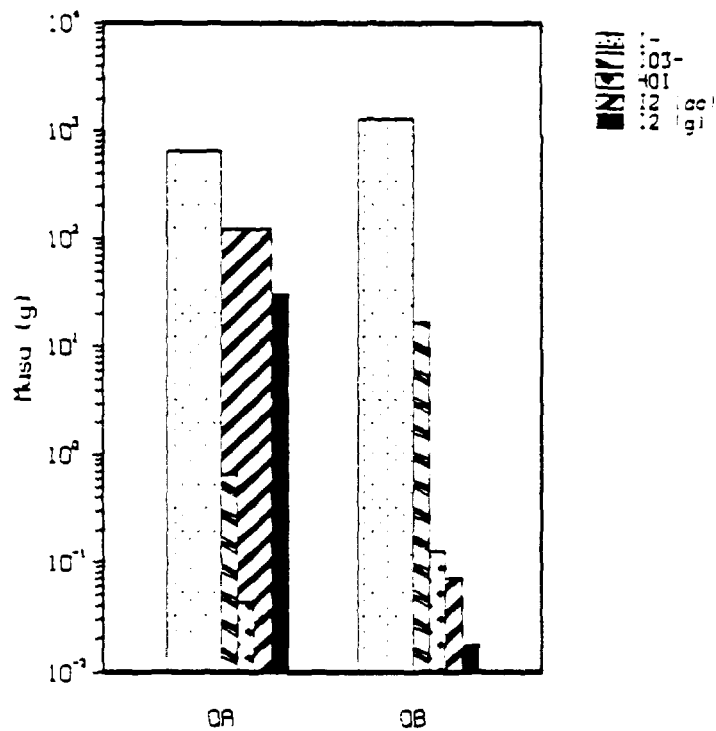


Figure 11. Iodine speciation at $t=280$ min under acidic (QA) and basic (QB) conditions

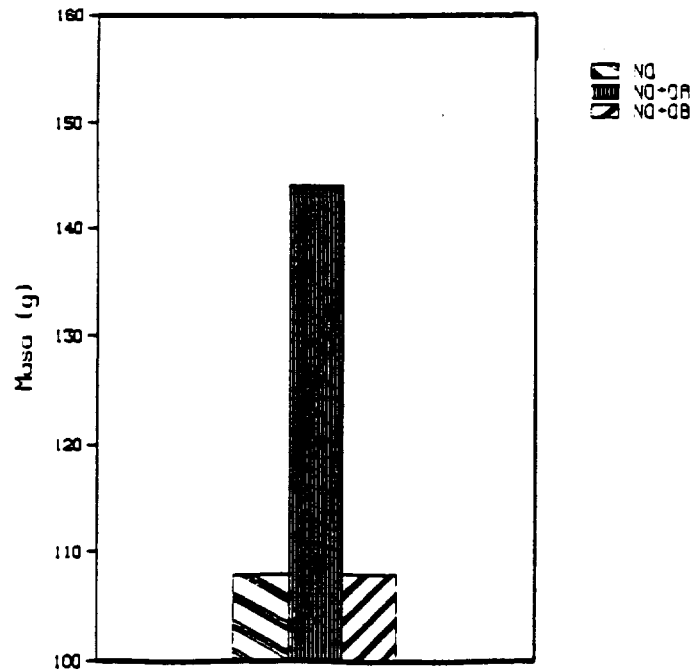


Figure 12. Total leaked mass without chemistry (NQ) and under acidic (QA) and basic (QB) conditions

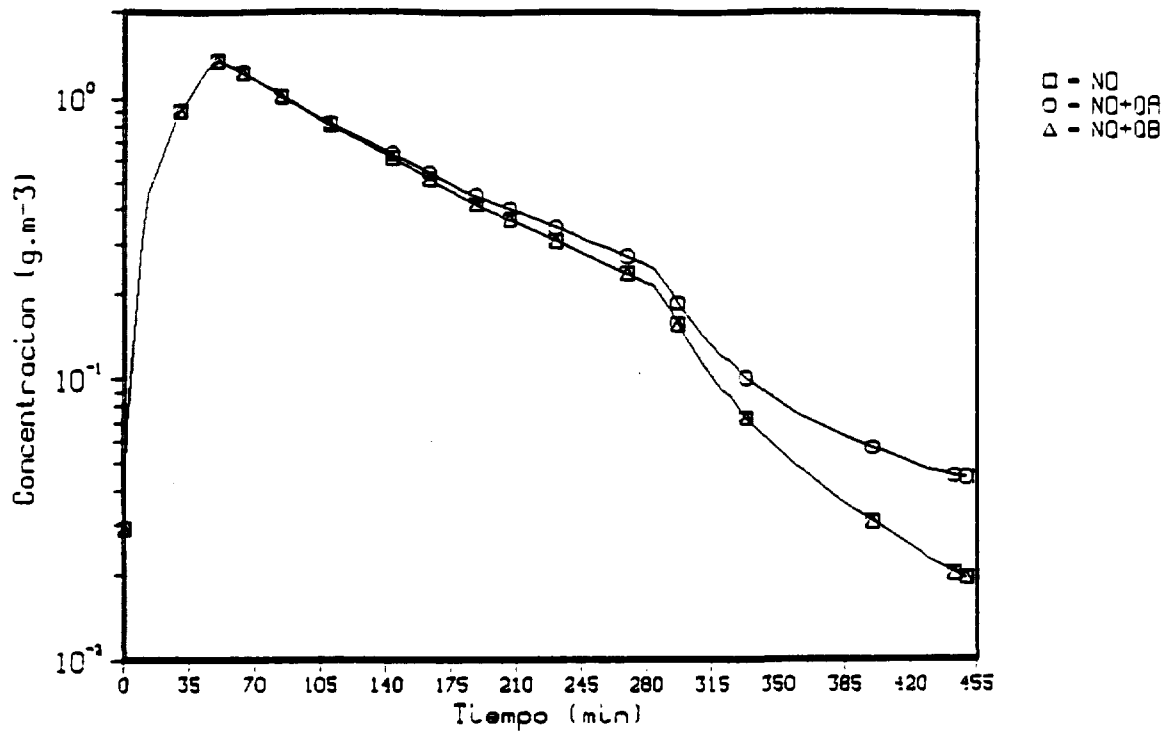


Figure 13. Total concentration of iodine in atmosphere vs time (NQ - particles; QA - particles + I₂(g); QB - particles + I₂(g))