

**RADIOISOTOPE TRACER APPLICATIONS IN INDUSTRY**

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Abstract

Radioisotope tracers have many advantages in industrial trouble-shooting and studies on process kinetics. The applications are mainly of two types: one leading to qualitative (Yes or No type) information and the other to quantitative characterisation of flow processes through mass balance considerations and flow models. "Yes or No" type methods are mainly used for leakage and blockage locations in pipelines and in other industrial systems and also for location of water seepage zones in oil wells.

Flow measurements in pipelines and mercury inventory in electrolytic cells are good examples of tracer methods using the mass balance approach. Axial dispersion model and Tanks-in-Series model are the two basic flow models commonly used with tracer methods for the characterisation of kinetic processes. Examples include studies on flow processes in sugar crystallisers as well as in a precalcinator in a cement plant.

## **I. Introduction**

Radioisotope tracers, by virtue of their special characteristics are ideally suited for fast diagnosis of faults in operating industrial systems and for studying the kinetics of industrial processes. The radioactive tracer methodology is often applicable for 'on-stream' application without having to shut-down normal operations.

There are two types of radiotracer methodologies. One is aimed at obtaining qualitative (Yes or No) or semi-quantitative information. Here the radiotracer is injected into one part of an industrial system and its appearance in another part of the system is monitored for radioactivity. The detection or otherwise of the tracer is directly interpreted for fault diagnosis. In the second type of applications, the input function of the tracer is related to its output function through simple mass balance or a suitable flow model.

## **II. Yes or No Type Applications**

This group of applications covers a wide range, but the following are representative of the potentialities.

1. Leak detection and location
2. Blockage location
3. Location of water seepage zones in oil wells

### **II.1 Leak detection and location**

This is probably the most common of radiotracer applications in industrial trouble-shooting. Detection of leak, if any, is unambiguously achieved by injecting the radioactive tracer in the part suspected to be leaking and monitoring for the tracer in the suspected part. Care should be taken to inject adequate quantity of the tracer so that the lowest suspected leak rate will result in tracer detection. Otherwise the interpretation of non-detection of tracer will no longer be unambiguous.

Location of actual leak position is a little more complicated as the monitoring procedure needs to be designed to suit each specific application. Following examples are illustrative.

### II.1.1 Leak detection in buried pipelines

There are many methods of leak detection in buried pipelines. Some are

- "Radiotracer-Detector Pig" Method
- "Velocity drop" Method
- "Tracer patch Migration" Method

#### II.1.1.1 Radiotracer-Detector Pig Method

This method is suitable for location of leaks in piggable pipelines (dia > 20 cm). The general procedure is as follows:

The suspected section (upto a few km) of the buried pipeline is isolated with jumpers provided to the fore and aft sections (fig.1). Small trenches are dug upto the pipeline at several points along the line in which small sealed sources of gamma emitting cobalt-60 or cesium-137 are placed as "marker" sources. The section is then filled with water along with the radiotracer (usually Br-82 as  $\text{NH}_4\text{Br}$ ). The section is then

pressurised to the line's normal operational level and held at that pressure for a few hours to allow adequate labelled water to leak to enable easy detection. The line is then depressurised and is washed with fresh water till the radiation level at the exposed part of the section returns to near background levels. Then a pig loaded with a detector-recorder package is sent through the line using water pressure. The pig is recovered at the other end and the tape is played on to a strip-chart recorder through a pulse integrating circuit. Fig.2 shows a typical record. The time elapsed between two marker peaks gives the speed of the detector pig from which the position of the leak is located.

#### Application

A few years back, a large scale investigation was carried out to locate leaks in a 140 km long buried crude oil pipeline in Western India. During the pre-commissioning trials, the pipeline was found to be leaky in a few sections. The radiotracer method described above was applied to the pipeline divided into seven sections. Five leaks were detected in about six weeks time (1). The size of leak varied from about 20 ml/min to 200 ml/min. The economic benefits from the investigation were estimated to be considerable; \$300,000 alone in investigation costs.

Another example is the detection of a small leak (about 40 ml/min) in the 200 km long buried Bombay-Poona pipeline carrying petroleum products from Bombay refineries to the Poona distribution terminal.

### II.1.1.2 Velocity-Drop Method

This method was found useful in the case of large leaks which cause significant fall in the velocity of flow after the leak. Here, a pulse injection of the tracer is made into the flow line and the tracer passage is monitored at feasible points by locating gamma ray detectors in trenches dug upto the pipeline. Once the section in which a fall in velocity is detected, the suspected section can be further narrowed down by repeating the test with a larger number of detectors in that section.

#### Application

The method was applied to locate the position of a leak in a 300 m long buried water main in a dyestuff factory (2). The main was carrying treated water from a filtration plant to a boiler with considerable loss of water due to the leak.

Fig.3 shows the alignment of the main and the pit positions. The first tracer test with Na-24 as  $\text{Na}_2\text{CO}_3$  indicated a sharp fall in the velocity after pit no.5. When the test was repeated with additional pits between 5 & 6, the leak was located between pits 5A & 5B.

In addition, the leak was found to be the source of a nearby pond.

### II.1.1.3 Tracer Patch Migration Method

This method is useful for minute leaks in small diameter (non-piggable) pipelines. The technique consists of injecting the radiotracer at the inlet of the pipeline, already filled with water, while keeping the outlet end blinded and observing the movement of the tracer patch at suitably placed trenches. The line is maintained at constant pressure throughout the test by continued replenishment of water lost through leakage. Under these static conditions, the tracer would fail to appear beyond the leak.

#### Application

The technique was applied to a 10 cm dia, 2 km long pipeline carrying naphtha from an oil refinery to a chemical industry in Bombay. The line was filled with water and blinded at the refinery end. Pits were placed at about 16 m spacing along the line. 3.5 mCi of Er-82 as  $\text{NH}_4\text{Er}$  was injected into the

pipeline and was pressurised to about  $4 \text{ kg/cm}^2$ . The tracer patch travelled with a reasonably steady speed of about 70 m/h to a

distance of 1.5 km and failed to progress any further. The suspected section was then dug up and the leak was visually located. The leak rate was about 15 l/min.

### II.1.2 Leak Detection in Industrial Systems

Two examples are given here:

#### II.1.2.1 Nylon Intermediates Plant

Charlton et al (3) used Argon-41 to sort out a leak detection problem in a nylon intermediates plant. Gaseous products from two reactors were fed to an absorber column. The absorber (fig.4) was protected from excess pressure by two control valves V1 & V2, one for each reactor. Excess flaring rates indicated that one or both the valves was "passing".

A pulse injection of Argon-41 was made into the relief line downstream of valve V1 and data from detectors D1, D2 & D3 were used to compute the volume flow rates down the individual relief lines and down the flare stack. The flow in the common line was much more than that down the line from reactor 1, thus suggesting that valve V2 was responsible for excess flaring. This was confirmed by a further test with tracer injected at point 2.

#### II.1.2.2 Rubber Internal Mixer

This test (4) was carried out at a rubber tyre factory situated near Delhi. The internal rubber mixer mixes natural rubber, carbon and some chemicals at high shearing forces to provide the main starting material for tyre manufacture. This causes heating and the temperature rises to about 150 deg C in the mixer. A chilled water jacket cools the mixer. The batch mixing of 200 kg is complete in about 2 minutes. Water droplets sometimes noticed in the mixture were attributable either to leaks in the coolant circuit or to inherent moisture in the feedstock.

Connecting the coolant system to a temporary 2000 litre capacity tank, injecting about 100 mCi of Br-82 as  $\text{NH}_4\text{Br}$  into the

tank and monitoring 6 batches of the product (sample size: 25 kg) for radioactivity, it was conclusively proved that the coolant system was not leaking. The sensitivity of the technique was demonstrated by introducing a small amount (1 litre) of labelled water directly into the feed and monitoring the product for radioactivity. The level of radioactivity indicated that even if 50 ml of water had leaked from the coolant, it would have been detected.

## II.2 Blockage Location

Blockages occur in industrial systems for a variety of reasons, obstructing the free flow of materials. In pipelines, blockages occur due to inadvertent introduction of foreign materials during construction and due to scaling on walls during operation. Radiotracers offer elegant approaches for easy location of blockages. The actual design of the test would depend on the specific nature of the problem. Two examples are given here:

### II.2.1 Location of Blockages in Pipelines

Tracer method is particularly suited to buried pipelines since the alternatives such as radiometry are possible for overground pipelines. In the tracer method, a small sealed source (about 0.1 to 1 mCi) of a gamma emitter like cobalt-60 or cesium-137 is loaded into a "pig", introduced into the pipeline and is pushed along by air or water pressure. The progress of the pig is monitored by radiation detectors located at suitable trenches along the pipeline. Non-arrival of the pig at any point indicates blockage upstream of that point.

The tracer method was applied for detection of blockages in a 17 km long aviation turbine fuel (ATF) pipeline between a petroleum refinery and the storage terminal at Bombay airport. During precommissioning trials, it was found that the pipeline was blocked at several points because of extraneous materials left inside during construction. The tracer test enabled speedy detection of the blockages at low cost.

The same procedure was used to locate the interface between ATF and water ahead of it at the time of final commissioning of the line.

### II.2.2 Nitric Acid Tower

A nitric acid tower at an explosives factory developed a blockage obstructing free flow of distilled water. Samples drawn from different levels in the tower indicated a blockage between the trays 12 & 20 (fig.5).

For the purpose of the tracer test, the tower was emptied. Distilled water input from the top and air inlet from the bottom were started just before injecting 10 mCi of Br-82 as  $\text{NH}_4\text{Br}$

through the nozzle available at the 20th tray. Radiation monitoring was then carried out continuously at each tray position using shielded scintillation detectors. The tracer profiles given in fig.6 clearly indicate that the outlet of tray no. 15 was blocked. This information avoided opening the entire

tower for examination. It was opened at the 15th tray position for speedy remedial measures.

### II.3 Location of Water Seepage Zones in Oil Wells

Cementation behind casings in oil wells is intended to isolate oil and water bearing zones in oil fields. Poor cementation can cause channeling behind casings resulting in water being mixed up with the oil. All efforts are made to locate such channels and seal them.

Radiotracers are commonly used to locate channeling. A gamma ray logging tool coupled with a tracer injector is used in such investigations.

After obtaining a background gamma ray log in the suspected well, an adsorbable gamma tracer (like iridium-192 as  $\text{Na}_2\text{IrCl}_6$  in  $\text{HCl}$ ) is injected at the suspected channel position.

The well is then washed by replacing the well fluids atleast three times and then a final gamma ray log is taken. High gamma intensity will be recorded along the channel, if one is present.

Fig.7 shows the background and final gamma ray logs obtained in a radiotracer test (5) of a well in South Gujarat field. The s.p and resistivity logs are also included in the figure.

The well was producing gas around  $25,000 \text{ m}^3/\text{day}$  from  $\text{H}_2$  layer, but ceased to flow after 9 years. Since the estimated reserves far exceeded the cumulative production from the well, channeling was suspected and the isotope tracer survey was carried out.

The tracer test indicated channeling between the gas layer and water layer  $\text{H}_1$ . The channel was then sealed by squeezing cement slurry and the well started to produce again.

A similar approach is used in water injection wells where water is lost to thief zones. In the Peoples' Republic of China, over 4000 such tests are reported to have been carried out using Barium-131 labelled microspheres as the radioactive tracer.

### III Tracer Methods Based on Mass Balance and Flow Models

Here we aim at discussing the tracer methodologies intended for a quantitative characterisation of the dynamics of flow processes in industrial systems. This is based on tracer input-output correlations with the flow characteristics (fig.8) such as the mean velocity, flow rate, axial and longitudinal dispersion, mixing etc.

"Mass balance" i.e. the mass of both tracer and bulk flow entering the system are totally accounted for at the outlet is not sufficient to make tracer input-output correlations. It is also necessary that the tracer flow pattern be identical to that of the bulk flow at the inlet itself (condition of good mixing).

The mass balance concept (with good mixing) is sufficient to determine such flow characteristics as mean velocity, flow rate and batch mixing. However for the characterisation of dispersion in continuous flow systems, we need to use suitable flow models.

### III.1 Mass Balance

#### III.1.1 Flow Measurement

A large percentage of radiotracer tests carried out in industrial systems appear to pertain to measurement of fluid flow rates for one reason or the other. It could either be for calibration or recalibration of installed flow meters or as a primary method wherever there are no flow meters or for trouble-shooting in industrial systems. The tracer methods of flow measurement are either based on tracer dilution or on transit time measurement.

##### III.1.1.1 Tracer Dilution Methods

Let a tracer of mass  $m$  be injected into a system of flow rate  $Q$ . The tracer concentration-time ( $c-t$ ) response (fig.9) at any point downstream is related to the mass of tracer injected as

$$m = \iint [c(t) dt] v d\sigma \quad (1)$$

$d\sigma$  is an elemental cross-section of flow

If complete mixing of the tracer is achieved at the point of measurement, we have  $(dQ/Q) = (dm/m)$ . We can then simplify equation (1) to

$$Q = \iint v d\sigma = \frac{m}{\int c(t) dt} \quad (2)$$

since  $\int c(t) dt$  will be constant at any point in the cross-section.

If  $c_m$  is the mean concentration of all the samples collected over a period of time  $t$ , then  $\int c(t) dt = c_m t$  and



$$Q = \frac{m}{c_m t} \quad (3)$$

The above equation is particularly useful for an instantaneous input of the tracer or in principle for any arbitrary input function. This is called the **Tracer integration Method** of flow measurement.

On the other hand, if we make a continuous injection of the tracer of concentration  $C$  at a constant input flow rate  $q$  and measure the the tracer concentration  $c$  during the plateau period  $t$  (fig.10) at a point downstream where complete mixing is achieved, we can show by mass balance, for  $q \ll Q$

$$qC = Qc \quad \text{or} \quad Q = (qC/c) \quad (4)$$

This is called the **Constant Rate Injection Method**.

### III.1.2 Transit Time Method of Flow Measurement

In many industrial systems such as pipelines, it is usually not possible to take samples and in such cases we use this transit time method (also called pulse velocity method) taking advantage of the unique detectability of gamma emitting tracers from outside the system. Here an instantaneous injection of the tracer is made at cross-section and two detectors placed at and downstream record the passage of the tracer (fig.11). If complete mixing of the tracer is achieved by the time the tracer reaches ,  $t_m$  is the mean transit time between and and  $V$  is the volume of the system between the two points, then we have

$$Q = (V/t_m) \quad (5)$$

Though the method is called Transit time or Pulse velocity method, it is in fact based on the mean residence time of the tracer between two given cross-sections (6).

### III.1.1.3 Examples of Flow Measurement

#### Test of Waste Heat Boiler

Charlton et al (3) described the use of the pulse velocity method to investigate the cause of recurrent tube failure in a waste heat boiler (fig.12). Velocity of flow and hence the volume flow rate in each take-off (AB, CD etc) was separately measured. In each case a pulse injection of an aqueous tracer (Na-24 as sodium carbonate solution) was made upstream of the feed pump.

The velocity in each take-off was measured by the two detector system as shown in the figure. The results clearly indicated that the distribution of water flow in the six take-offs was uneven, with the last two IJ & KL taking excessively high quantity of water and the first four correspondingly starved. This tube dry-out was precisely the cause of the problem and subsequent modifications made on the geometry of the pipe-work helped to greatly reduce the frequency of failure.

### Calibration of Flow Meters

This is an area of large application of the transit time method of flow measurement. In many industries with on-line computers for process control, the need for reliability of flow data has greatly increased. For several reasons the installed flow meters appear to need recalibration at regular intervals to ensure their measurement accuracies.

Kurten (7) described the use of the pulse velocity method for the calibration of magnetic flow meters at three different paper mills.

The flow measurements were made using an aqueous solution of  $\text{NH}_4^{82}\text{Br}$  as the tracer. The tracer was injected instantaneously into the pipe, usually immediately upstream of a pump and the tracer concentration pulses were recorded at two points sufficiently downstream to ensure complete mixing of the tracer. Eq(5) was used to calculate the flow rate in the pipeline. The measurement was repeated four or five times and the mean value was compared with the flow meter reading. The procedure was repeated for different flow rates within the working range of the flow meters. The difference between the flow meter reading  $Q_F$  and the tracer-measured flow rate  $Q_T$  i.e  $Q_F - Q_T = \Delta Q$  was used to

build a calibration line by linear regression. One such calibration is shown in fig.13. The experience of the author was that the the flow meters were fairly stable for long periods, but suddenly changed giving a different calibration slope. The overall deviation of the flow meter reading varied between -20% to +30% compared to the tracer measurement (error : about 0.5%). The flow meter accuracy was not as good as claimed.

### Testing a long distance pipeline

Recently the 200 km long Bombay off-shore crude oil pipeline was tested for any loss of volume due to waxy deposits. Transit time from Bombay High to the on-shore terminal at Uran was determined using paradibromobenzene labelled with Br-82 as the radiotracer. This transit time was compared with what was

expected from the flow rate and the cross-section of the pipeline. The results indicated that the pipeline was free from any deposits.

### III.1.2 Material Inventory

Radioisotope dilution method based on mass balance provides an elegant technique for taking material inventory in a closed or recirculating system where other conventional methods may not be practicable. The main advantages of the radiotracer method are that the procedure can be carried out on-stream without shutting-down the plant and the amount of tracer added is negligible compared to the bulk material in the system.

In this method, a radiotracer of mass  $m$  and specific activity  $C_1$  (Bq/g) is added to the bulk and is allowed to mix

thoroughly. At the end of the mixing period an aliquot of the bulk is taken and its specific activity  $c_s$  is determined. From

mass balance and tracer balance considerations, the mass  $M$  of the bulk can be determined from

$$(M + m) c_s = m C_1 \quad (7)$$

or

$$M = m \left[ \frac{C_1}{c_s} - 1 \right] \quad (8)$$

#### III.1.2.1 Mercury Inventory in Electrolytic Cells

This is probably the most common application of the isotope dilution method. In caustic soda plants and for chlorine production, brine is electrolysed using flowing mercury as the cathode and carbon as the anode. From consideration of the economics of the process and environmental pollution it is necessary to take inventory of mercury at regular intervals.

The conventional gravimetric method of mercury inventory involves shutting-down the plant, draining the mercury and weighing. This causes loss of production, needs more manpower and the method is not very accurate as some mercury usually remains trapped inside the cells.

On the other hand, in the isotope dilution method, radioactive elemental mercury (Hg-197 or Hg-203) is injected into an operating cell, allowed to mix thoroughly over a period of a day or two and at regular intervals thereafter samples are taken out for concentration measurement. Successive samples should show same concentration within counting statistics. Standards prepared from samples of injected mercury are also counted along

with the cell samples to avoid problems of radioactive decay and instrumental drift. Eq (8) is then used to obtain the total quantity of mercury in each cell.

Hg-203 with a half-life of about 47 days and 280 keV gamma energy is the usually preferred tracer. The shorter lived Hg-197 (Half-life : 2.7 days, Gamma : 77 keV) is selected (4) whenever the inventory has to be taken more frequently. The accuracy obtainable varies from 1 to 1.5%.

### III.2 Tracer Methodologies Based on Basic Flow Models

We have so far seen how the tracers can be used to measure flow rates and velocities of fluids in process systems. It did not matter to us as to what type of temporal distribution of tracer molecules took place within the systems. This is, however, of large importance to the designer and operator of a processing system since the distribution of time spent by a given group of feed molecules in the system has a large bearing on the quality of the final product and on the economy of the process.

We can visualize two ideal conditions. One is called "piston flow" or "plug flow" in which all the molecules entering the system at any time  $t$  spend exactly the same time  $T$  within the system i.e

$$c_1(t) = c_0(t+T) \quad (9)$$

is the relationship between the tracer input concentration and the tracer output concentration.

The second ideal condition is called "completely mixed flow at all times". In this, the input is completely mixed instantaneously with the bulk in the processing system. Here the tracer output  $c(t)$  for an instantaneous tracer input is

$$c(t) = c(0) e^{-(Qt/V)} \quad (10)$$

where  $c(t)$  &  $c(0)$  are the output concentrations at any time  $t$  and  $t=0$  respectively,  $Q$  is the flow rate and  $V$  is the volume of the process vessel.

In practice, however, the above ideal conditions are only rarely achieved and the situation is usually somewhere between the two extremes (fig.14) (3).

Before we start discussing characterisation of flow processes in terms of deviations from the above two, ideal conditions, let us see what information we can derive from the

concentration - time response of a system for a familiar instantaneous injection.

### III.2.1 Mean Residence Time (MRT)

It is desirable to define the average time a statistically representative number of molecules spends in a processing system. The centroid (first moment) of the concentration - time distribution curve is defined as the Mean Residence Time (MRT) of the system. Thus

$$\text{MRT} = t_m = \frac{\int_0^{\infty} c(t)t dt}{\int_0^{\infty} c(t) dt} \quad (11)$$

This is usually compared with the theoretical mean residence time

$$t_m(\text{th}) = (V/Q) \quad (12)$$

Any deviation of  $t_m$  from  $t_m(\text{th})$  is an indication of some dead volume within the system.

### III.2.2 Variance

This is another important characteristic which describes the spread of the distribution. This is obtained from the second moment of the c-t curve and is expressed as

$$s^2 = \frac{\int_0^{\infty} (t - t_m)^2 c(t) dt}{\int_0^{\infty} c(t) dt} \quad (13)$$

This is particularly useful for matching experimental c-t curves with a family of theoretical curves.

s is called the standard deviation

One important and useful property of  $t_m$  and of  $s^2$  is that they are additive for a flow through a series of interconnected vessels 1,2,3 etc. So

$$\begin{aligned} t_m &= t_m(1) + t_m(2) + t_m(3) + \dots \\ s^2 &= s^2(1) + s^2(2) + s^2(3) + \dots \end{aligned} \quad (14)$$

Let us now discuss two simple flow models which are often used to characterise a flow process in terms of deviations from the ideal plug or fully mixed flow conditions.

### III.2.3 Basic Flow Models

Any process system can be represented this way

Input  $X(t)$  --- (SYSTEM) ----> Output  $Y(t)$

The relationship of  $Y(t)$  with  $X(t)$  is a characteristic of the system. Any mathematical simulation of  $Y(t)$  for an input  $X(t)$  through an appropriate transfer function  $H(t)$  can yield valuable information about the system.

The transfer function  $H(t)$  is defined as the response of the system for a  $\delta$ -input function (instantaneous injection of the tracer). So, for a  $\delta$ -input of tracer

$$H(t) = Y(t) \quad (15)$$

Any arbitrary but known input function  $X(t)$  can be considered as composed of  $n$  pulses occurring at  $T_1, T_2, T_3 \dots T_n$ , each of duration  $\Delta T$  and amplitude  $X(T_i)$ .

The response of the system for such an arbitrary input can be expressed in the following convolution integral form

$$Y(t) = \int_0^t X(t-T)H(t-T)dT \quad (16)$$

The upper limit of the integral is  $t$  since  $X(t)$  cannot contribute to  $Y(t)$  if  $T > t$

The two basic models which are often used to obtain the transfer function  $H(t)$  are

1. Axial Dispersion Model
2. Tanks-in-Series Model

#### III.2.3.1 Axial Dispersion Model

As the material passes through a vessel, it moves along the longitudinal direction primarily by advection as well as it tends to mix in the axial direction due to slippages, eddies etc., analogous to molecular diffusion. This advection-dispersion can be expressed by the following differential equation.

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - u \frac{\partial c}{\partial x} \quad (17)$$

where

$c$  is the concentration at a distance  $x$  from the input and at time  $t$

$D$  is the axial dispersion coefficient and

$u$  is the mean velocity of advective transport

For an instantaneous and planar injection at  $t=0$  and  $x=0$ , the solution for eq (17) has the form

$$c(x,t) = \frac{M}{A \sqrt{4\pi Dt}} \exp \left[ -\frac{(x - ut)^2}{4Dt} \right] \quad (18)$$

where  $M$  is the mass of tracer injected into the cross-section  $A$  at the inlet

The right hand side of eq (18) is the transfer function  $H(t)$  which is appropriately used in eq (16) for modelling the process. The model parameter normally used is the non-dimensional Peclet number  $P_e = (ux/D)$

It can be shown that

$P_e \rightarrow \infty$  for plug flow

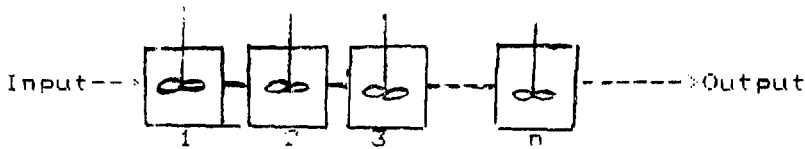
$P_e \rightarrow 0$  for fully mixed flow

The numerical value of  $P_e$  obtained in the model fitting process represents the degree of deviation of the process from either of the ideal conditions.

It is often difficult to apply the simple solution given in eq (18) since the boundary conditions are not usually satisfied, particularly in the case of closed vessels. Numerical methods have been attempted in such cases (8).

### III.2.3.2 Tanks-in-Series Model

This is the other one parameter model most widely used to simulate flow processes. Here we view the fluid passing through a series of perfectly mixed tanks of equal volume  $v$ . It is easier to simulate the  $c-t$  curves with this model since problems of proper boundary conditions and methods of tracer injection and measurement do not disturb.



For a  $\delta$ -input function at the input of the first tank, the analytical solution for the concentration  $c_n(t)$  in the  $n$ th tank is given by

$$\frac{c_n(t)}{c_1(0)} = \frac{(Qt/v)^{n-1} e^{-t/v}}{(n-1)!} \quad (19)$$

where  $c_1(0)$  is the concentration in the first tank at time  $t=0$

RHS in eq (19) is the transfer function  $H(t)$  which is appropriately used in eq (16) for curve fitting. It can be shown that

$$\begin{aligned} n & \text{-----} \infty && \text{for plug flow} \\ n & \text{-----} 1 && \text{for fully mixed flow} \end{aligned}$$

As in the earlier model, the numerical value of  $n$  obtained in curve fitting using this model represents the degree of deviation of the process from either of the ideal conditions.

It can be shown that the two models when applicable have the following equivalence:

$$P_e = 2n \quad (20)$$

### III.2.3.3 Examples of Application

#### Sugar Crystallisers

In continuous sugar crystallisers, it is desirable to obtain as near a plug flow as possible with complete lateral mixing. Smith et al (10) describe model based interpretation of radiotracer data obtained from a continuous crystalliser installation. Fig.15 shows the layout of seven crystalliser units

(each  $26 \text{ m}^3$ ) connected in series. Initially all the crystallisers were equipped with reciprocating stirrers and the performance of the installation was poor from the point of view of heat transfer. On an experimental basis, the stirrer in the sixth crystalliser was changed to the conventional rotating type and the heat transfer efficiency improved. A radiotracer test was carried out to characterise the flow process in the digester units with different type of stirrers.



About 20 mCi of iodine-131 as aqueous iodide was first mixed with 1 litre of viscous sugar crystal slurry and instantaneously injected with compressed air into crystalliser no.5. Lead shielded NaI scintillation detectors monitored the passage of the tracer at the outlets of crystallisers 5,6 and 7.

A modified form of axial dispersion model (eq.18) was used to simulate the c-t curve obtained at the outlet of crystalliser No.5(fig.16). The modification was necessitated by the double peak response. It is assumed that there were two flow channels a & b carrying tracer quantities  $M_a$  &  $M_b$

$$M_a + M_b = M$$

$$M_c(x,t) = M_a c_a(x,t) + M_b c_b(x,t)$$

Assuming a  $\delta$ -input function and using the above modification for the axial dispersion model, the best fitting curve obtained using the least squares method is plotted on the same fig.16.

In the case of crystalliser 6, the tracer input is not a  $\delta$ -function and hence the convolution integral (eq.16) was used to simulate the c-t curve as shown in fig.16. Tracer data is given in the following table.

Cryst'iser No	Stirrer Type	$P_R$	$f^*$	$t_m$ (min)	% Dead volume
5a	Reciprocating	100	0.73	380	2.6
5b	"	35	0.27	"	"
6	Rotating	5	1.0	128	67

$f^*$  is the fraction of the tracer passing through each of the split channels

It is seen that the flow process in crystalliser No.5 is closer to plug flow compared to crystalliser 6 which shows higher mixing. The dead volume was also none in no.6. The study showed that the change-over to rotating type from reciprocating type would lead to unacceptable changes in flow patterns.

### RTD in Precalculator

A radiotracer study was recently carried out in a cement plant in India to investigate the residence time distribution of raw feed in a precalcinator. Two separate experiments were carried out, one with reactor irradiated raw feed as the tracer (mainly Na-24) and the second using raw feed surface labelled with La-140. Fig.17 shows the sketch of the precalcinator and fig.18 shows the c-t curve obtained with the irradiated raw feed as well as the model simulations. The data obtained are as follows:

	<u>Na-24</u>	<u>La-140</u>
Mean Residence Time	9.3 sec	9.9 sec
Standard Deviation	4.4 sec	5.2 sec
Peclet No.	14	12
No. of Tanks	8	7

The special feature of the experiment was the measurement of such a short residence time as 10 seconds. To do this the detectors were connected to a specially built data acquisition system which accumulated counts in every 0.1 sec duration and stored the data for later retrieval. Both the tracers yielded similar results and model simulations show considerable back-mixing in the precalcinator.

### **IV Conclusions**

From the foregoing it is clear that radioactive tracers have a unique role to play in industrial trouble-shooting, particularly for location of leaks and blockages and also in studies on industrial process kinetics. They also find extensive application in flow measurements and for mercury inventory in electrolytic cells. The amounts of radioactivity used in most of the studies are so small that they do not pose any undue hazard.

The simple models discussed in this paper are not always applicable to more complicated systems. Advanced models which take into account exchanges between "dead" and "flow" zones and recirculation, if any as well as models for variable flow processes have also been developed.

## V References

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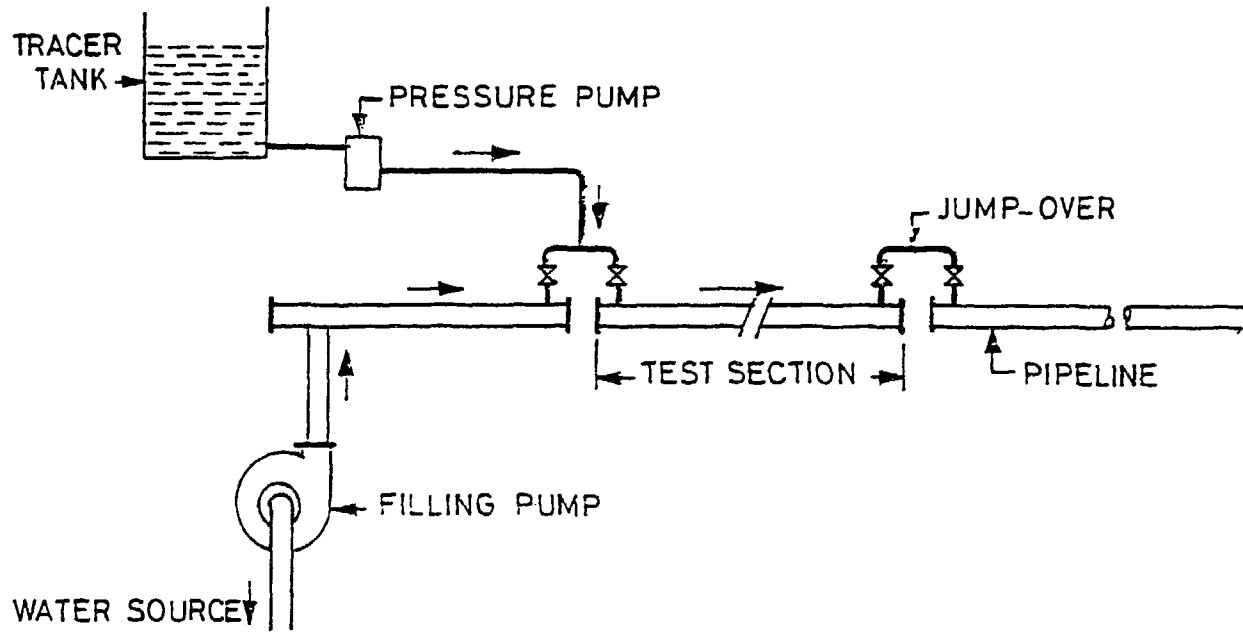


Figure 1. Tracer injection arrangement for leak detection by "Radiotracer-Detector pig" method

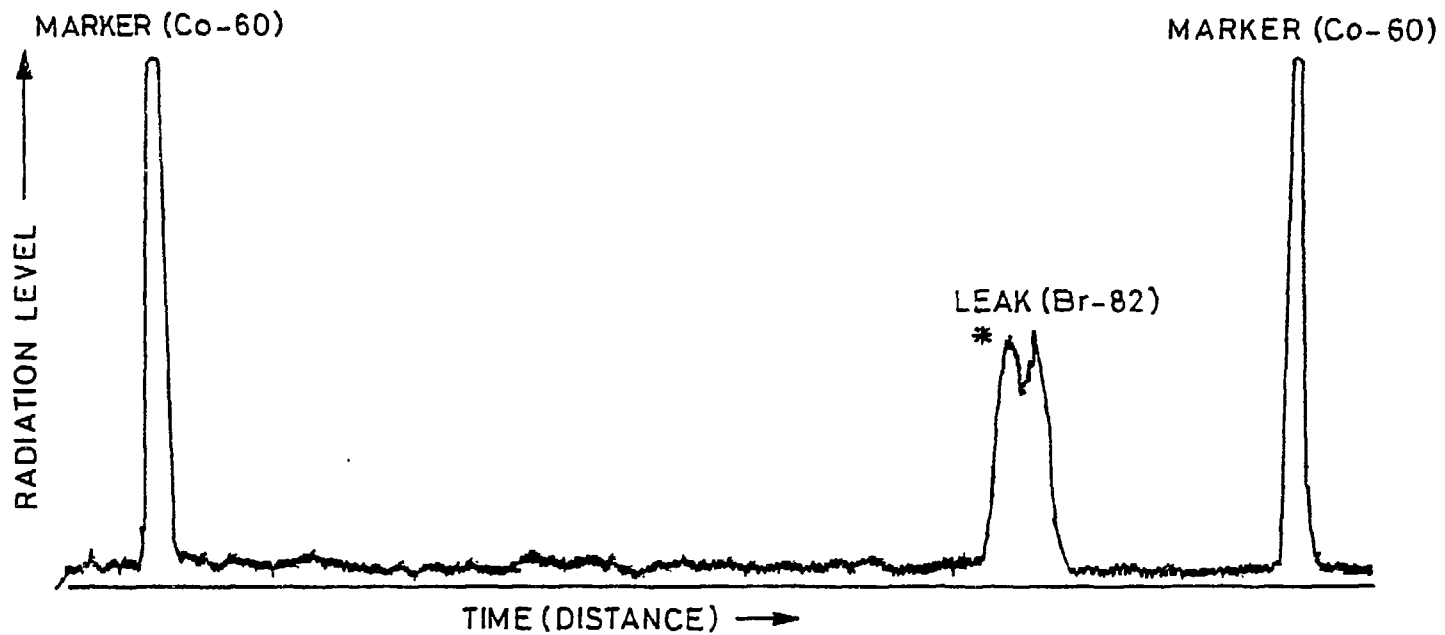


Figure 2. Typical recording of tracer at leak in the "Radiotracer-Detector pig" method

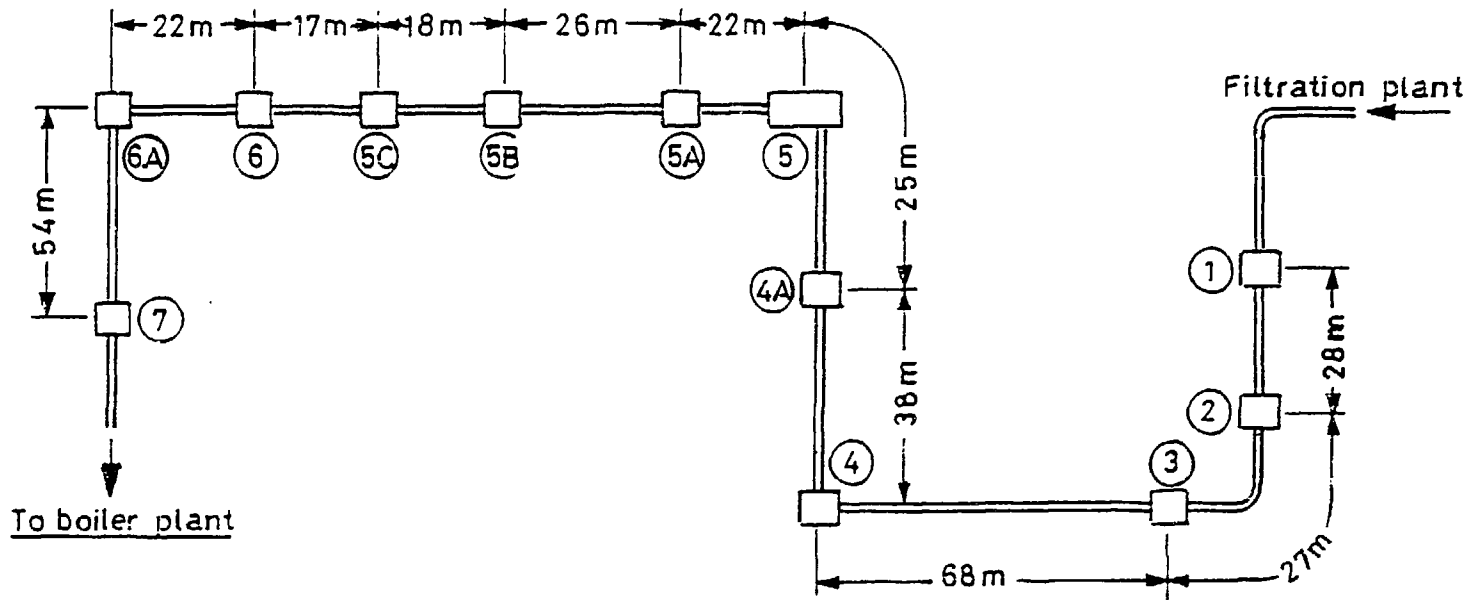


Figure 3. Pit positions on buried water main

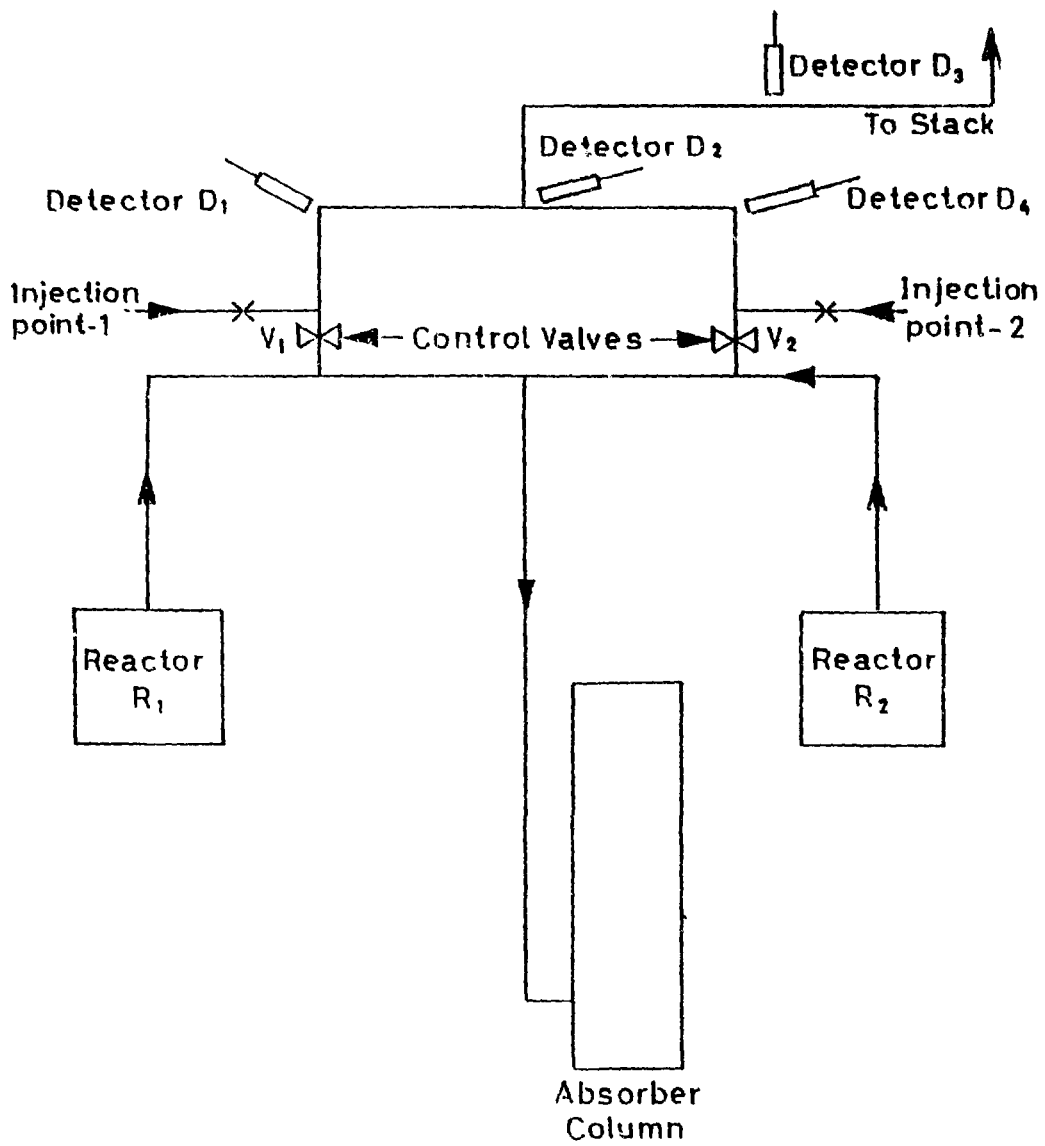
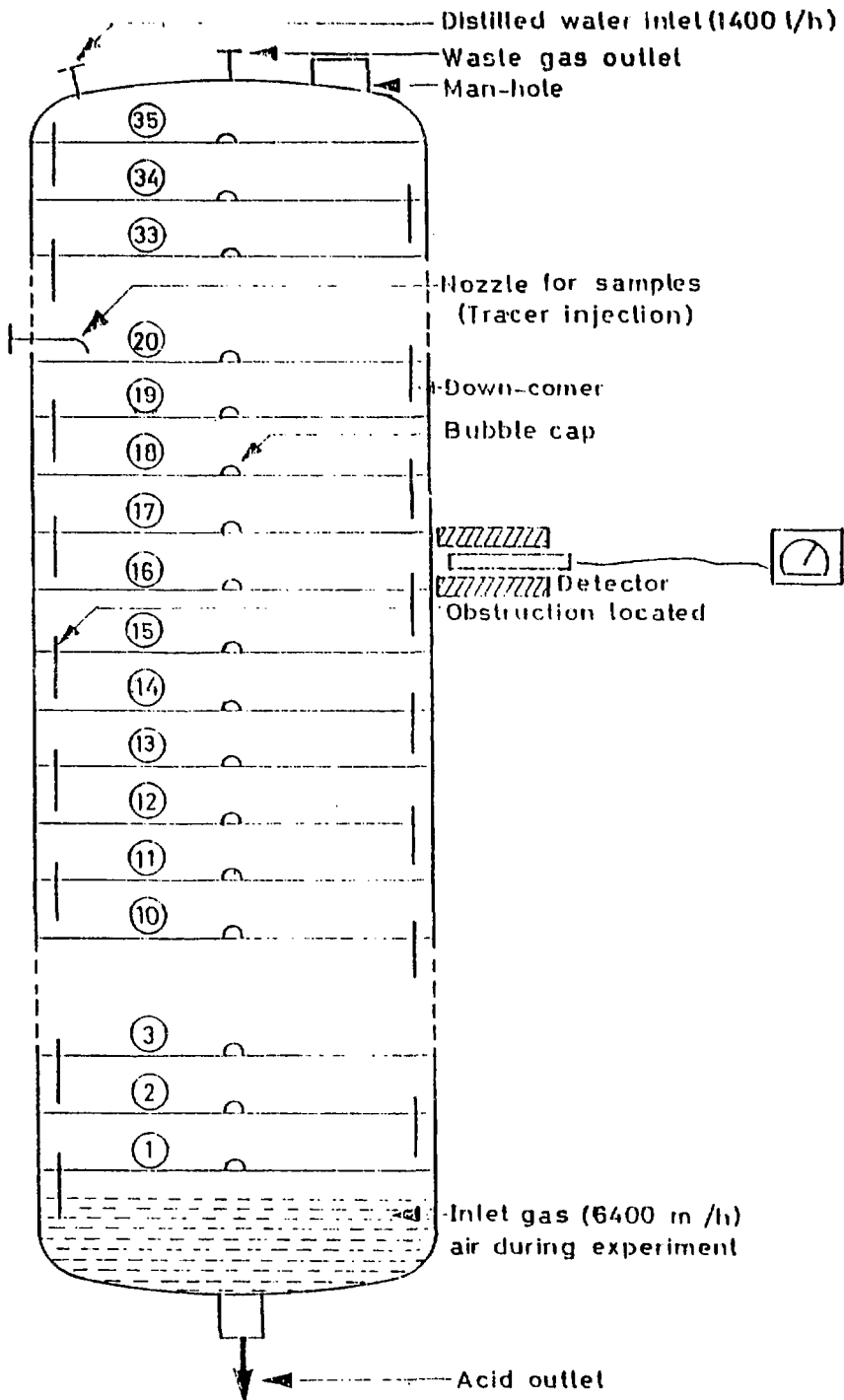


Figure 4. Leak detection in a nylon plant



**Figure 5. Nitric acid tower:Tracer injection position and detector arrangement**



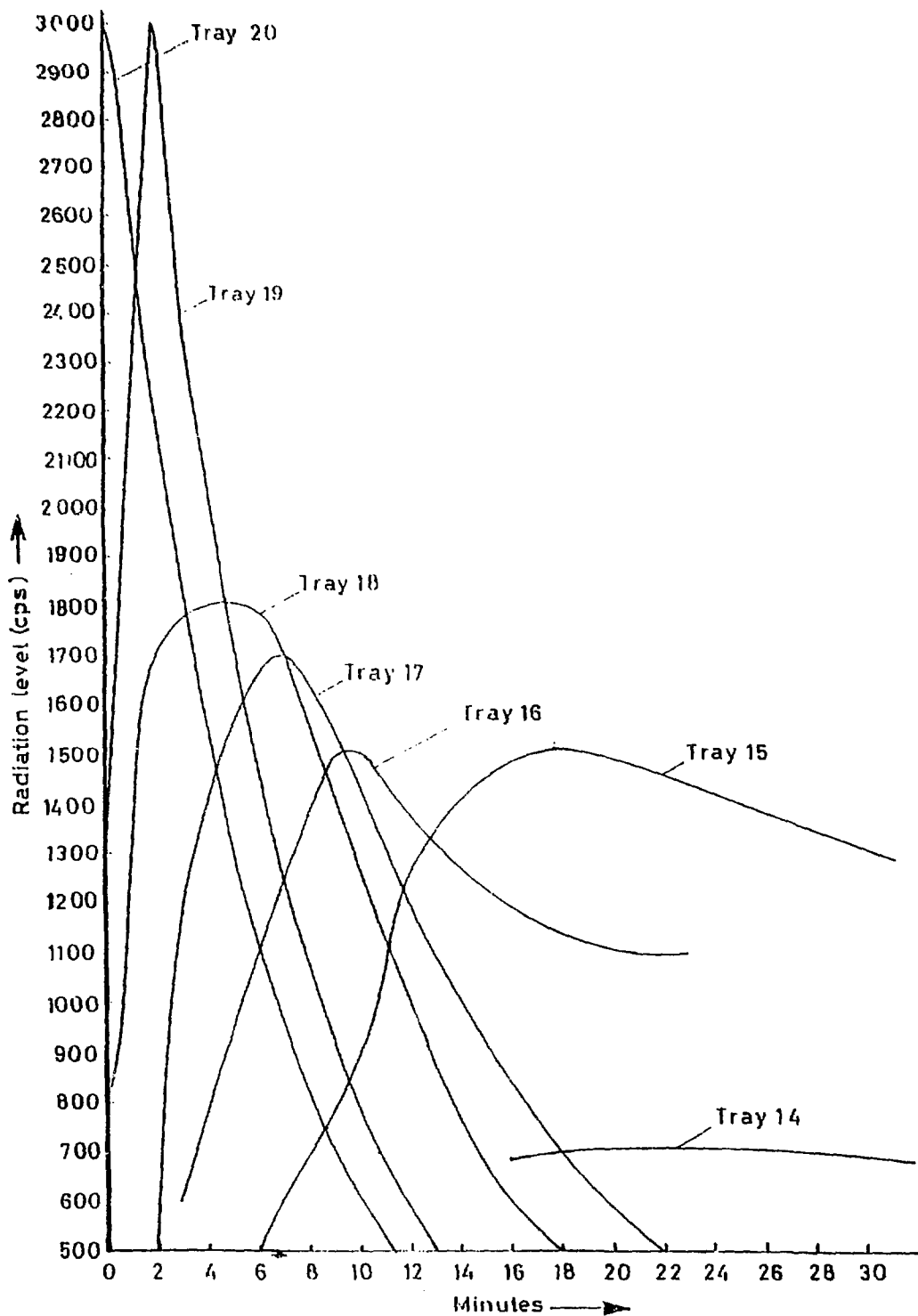


Figure 6. Tracer responses at different tray positions in the nitric acid tower

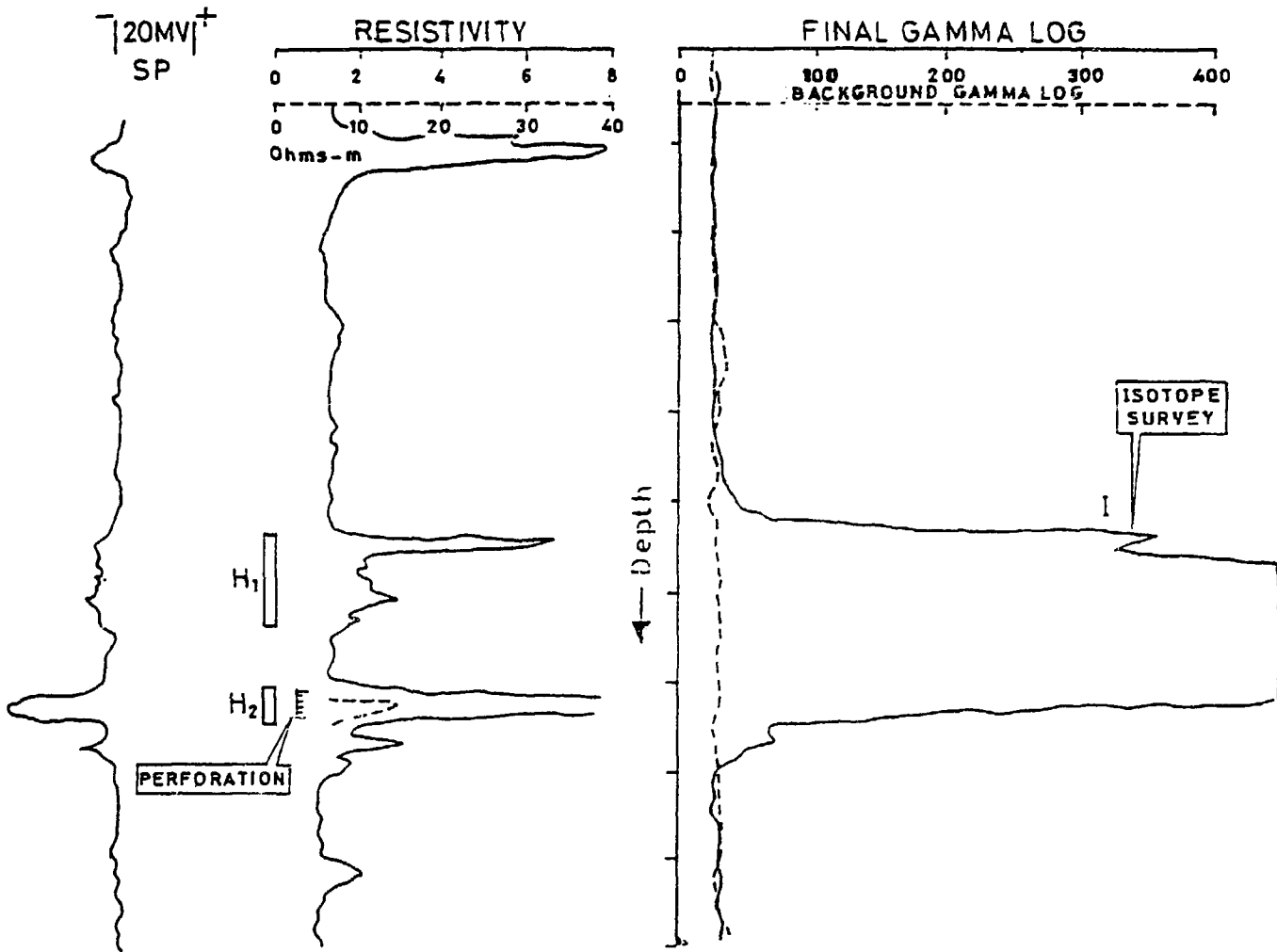


Figure 7. Detection of channeling in an oil well

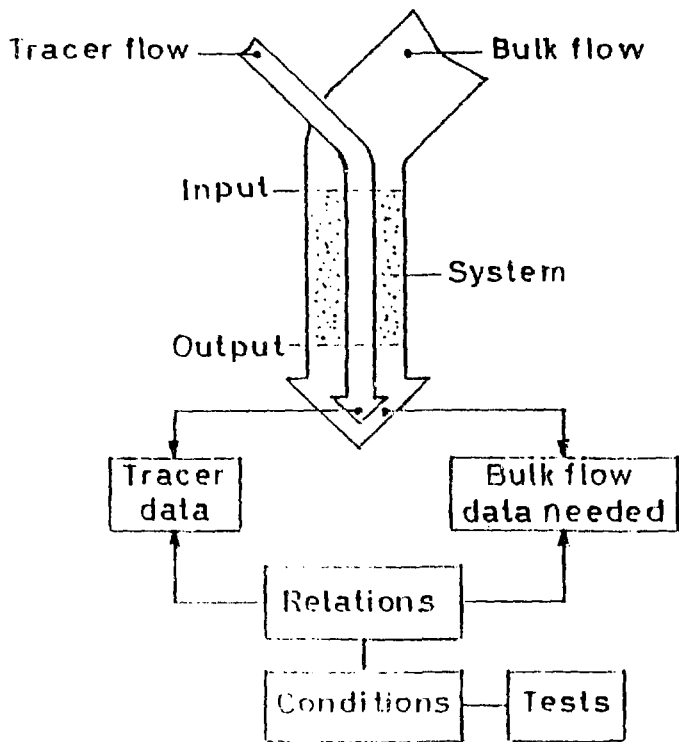


Figure 8. Tracer methodology to study a flow system

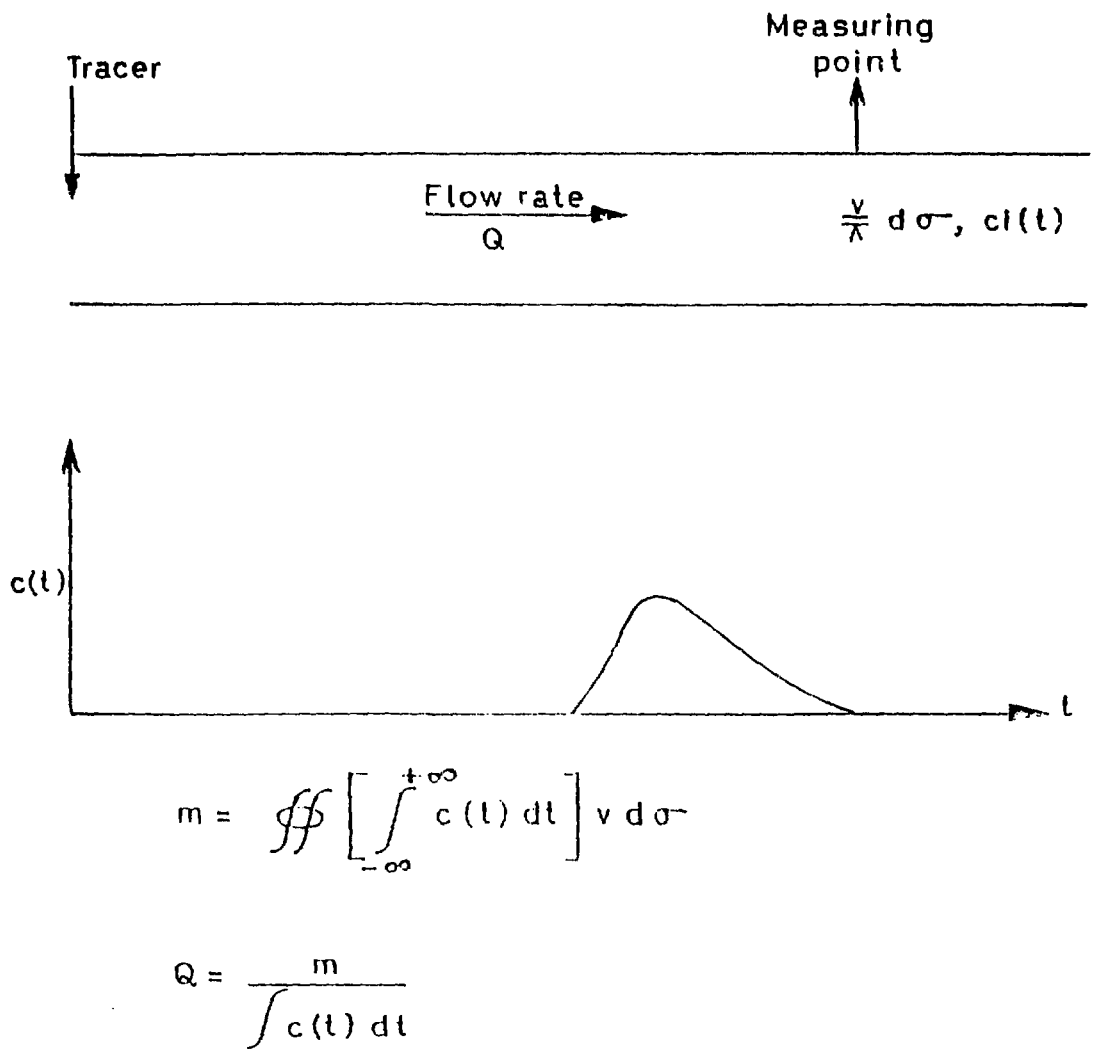
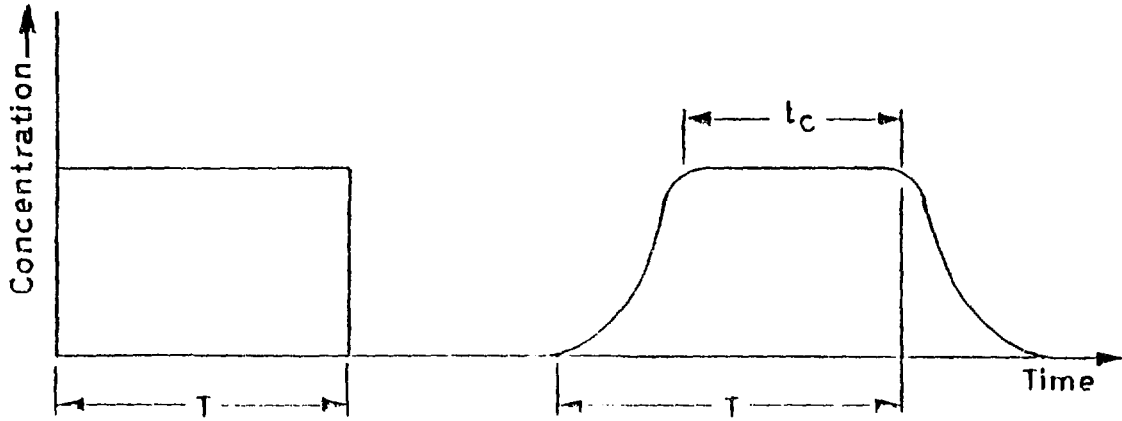
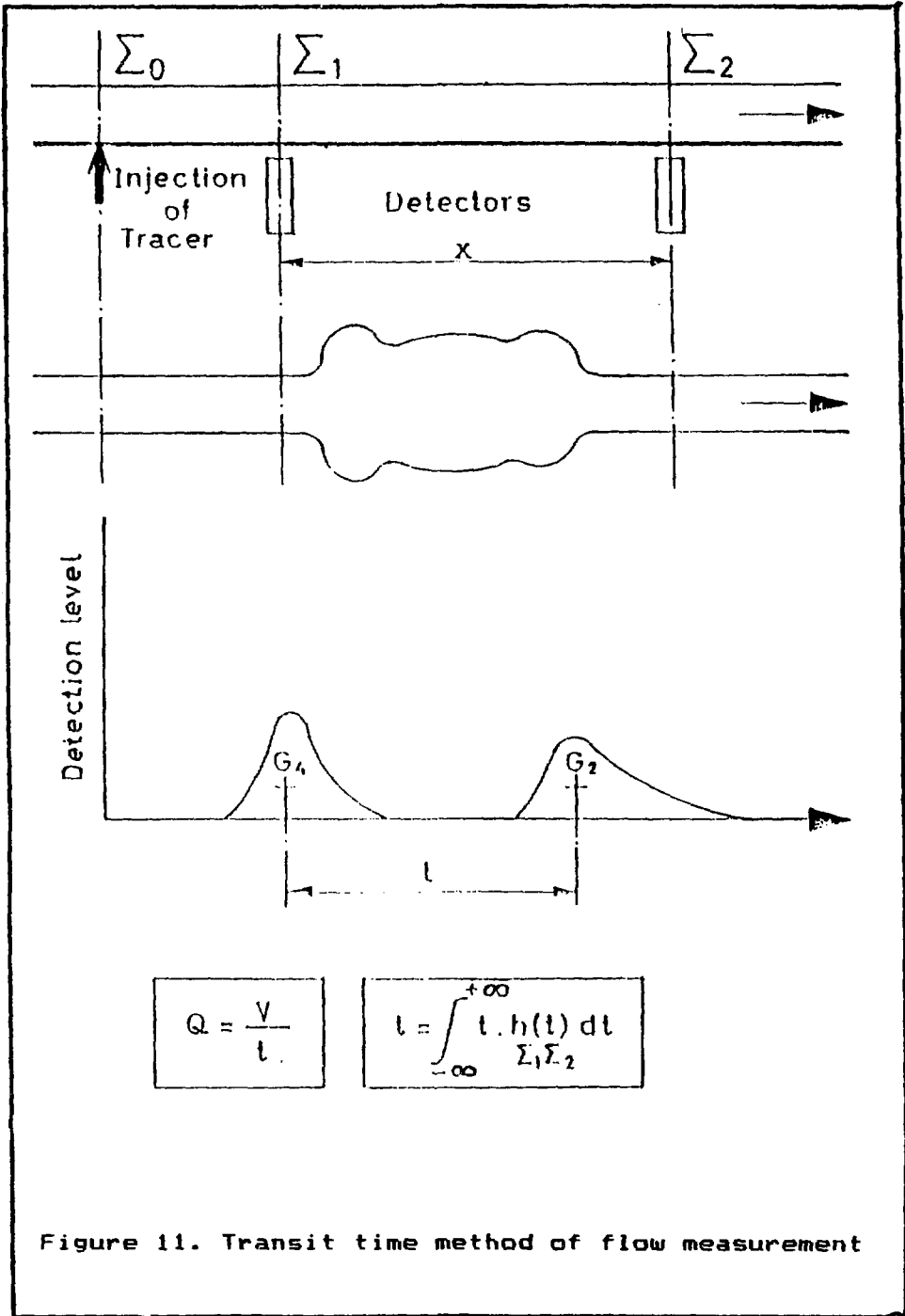


Figure 9. Tracer integration method of flow measurement



$$qC = \iint cv \, d\sigma \quad \text{or} \quad Q = \frac{Cq}{c}$$

Figure 10. Constant rate injection method of flow measurement



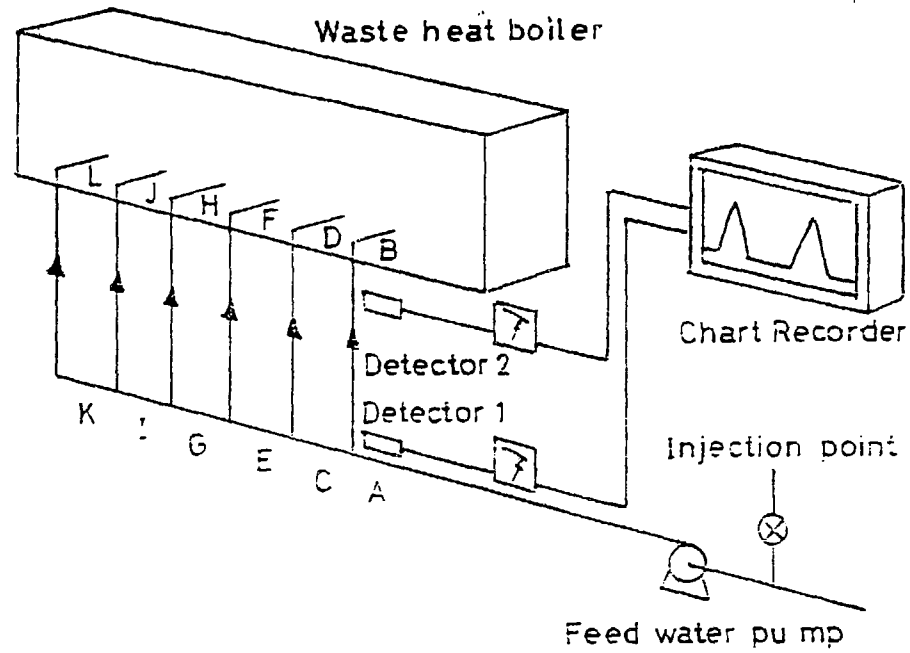


Figure 12. Distribution of feed water to waste heat boiler

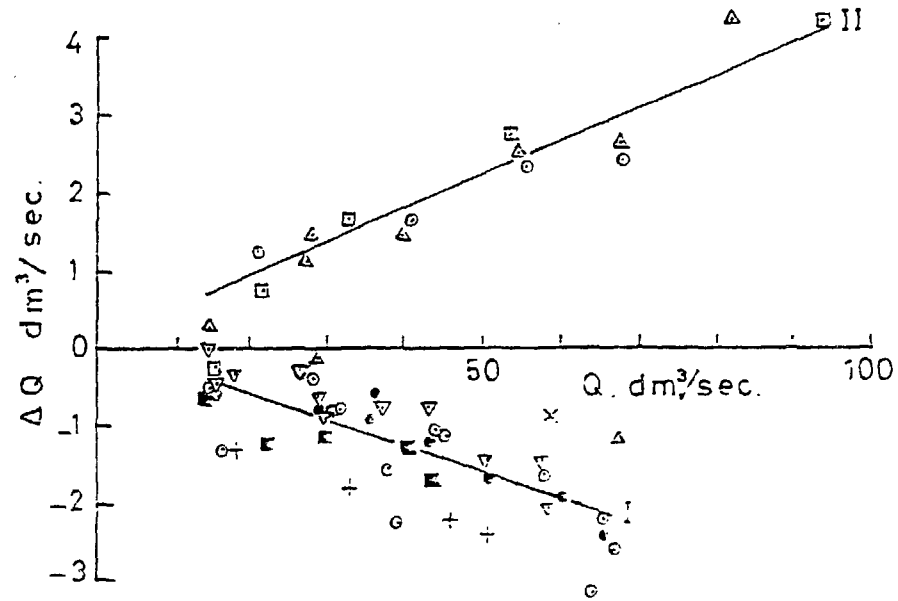


Figure 13. Calibration regression lines obtained for a magnetic flow meter.

Calibration lines: (a)  $\Delta Q = 0.1 \text{ dm}^3/\text{sec} - 0.034 Q_T$   
 (b)  $\Delta Q = 0.1 \text{ dm}^3/\text{sec} + 0.042 Q_T$



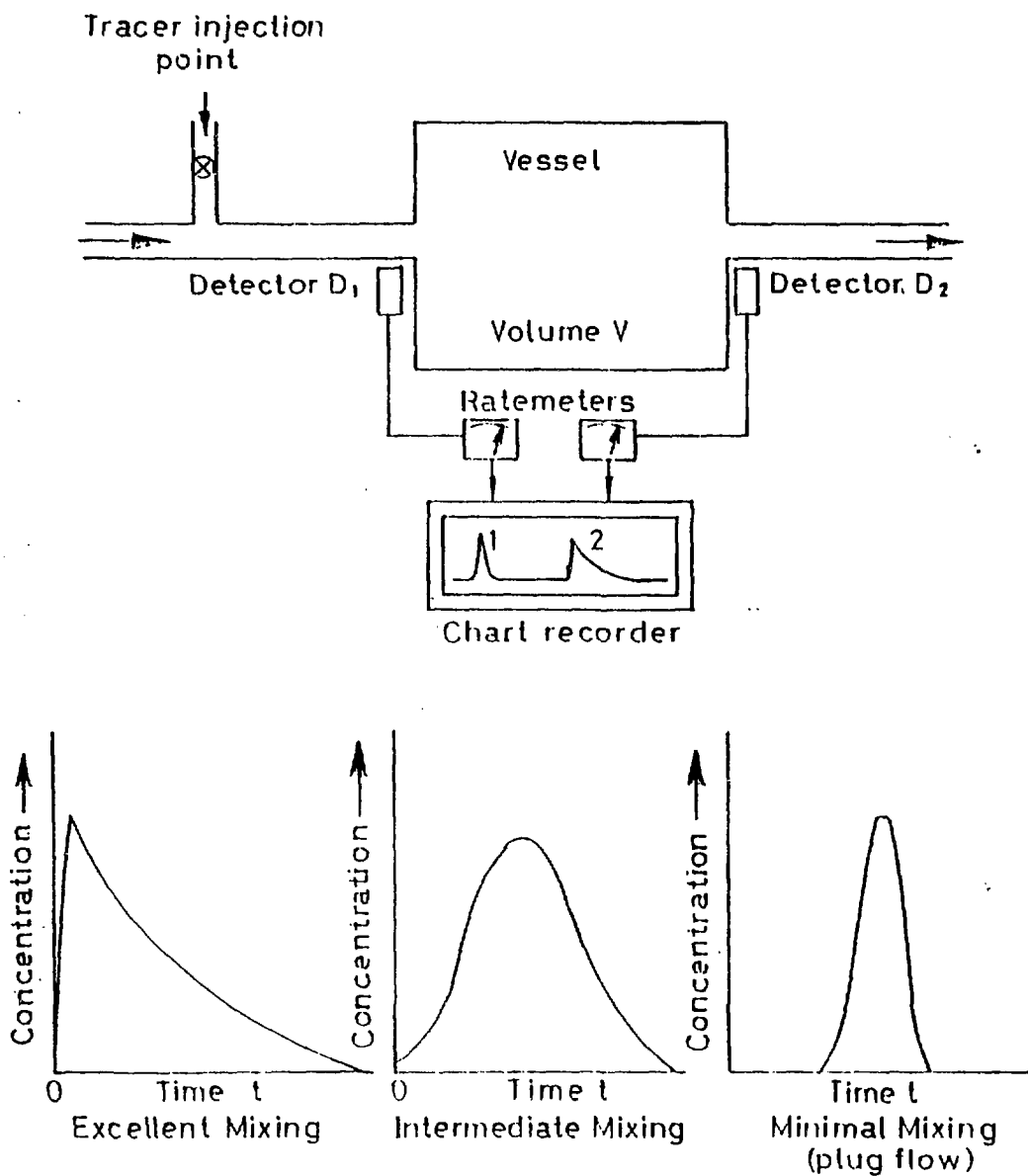


Figure 14. Typical concentration-time curves obtained in tracer studies

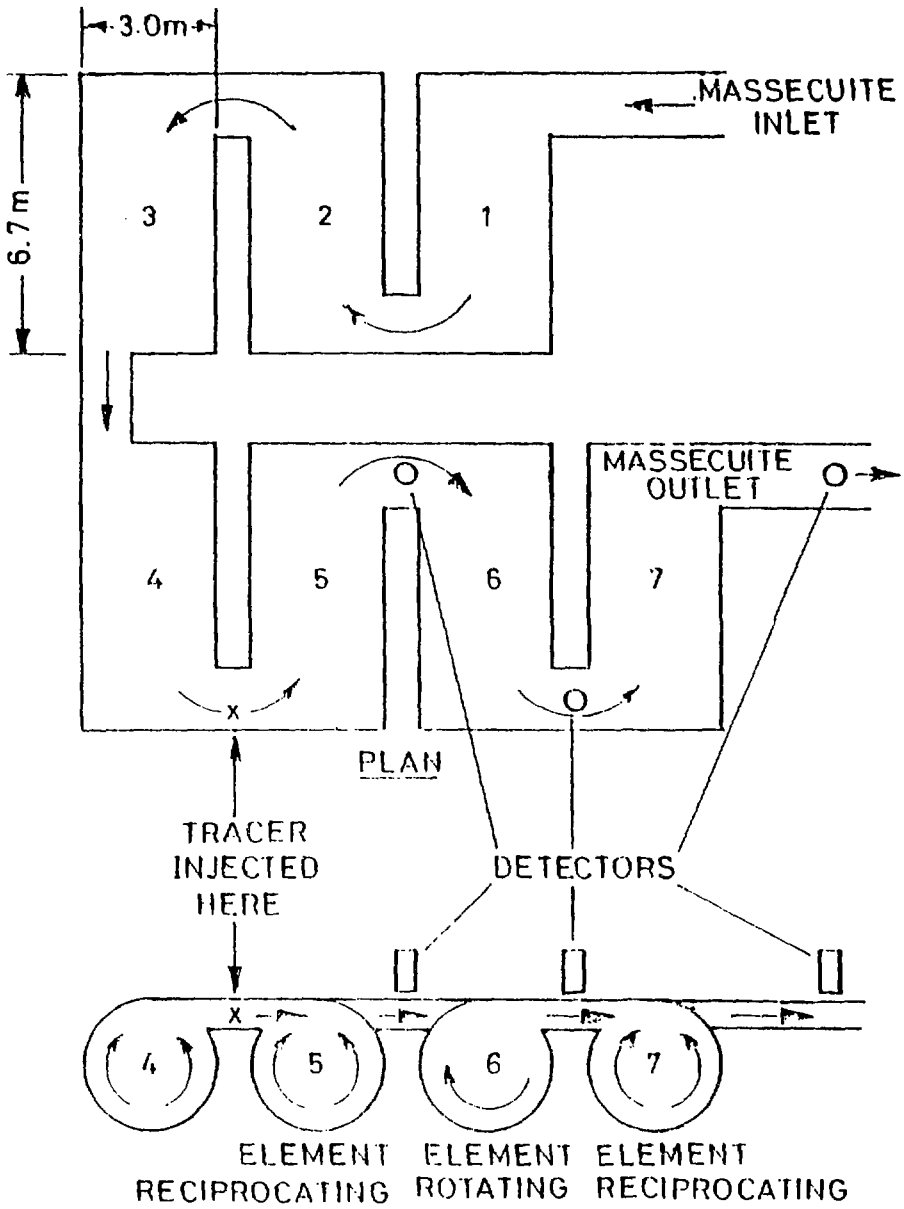


Figure 15. Lay out of the crystalliser system

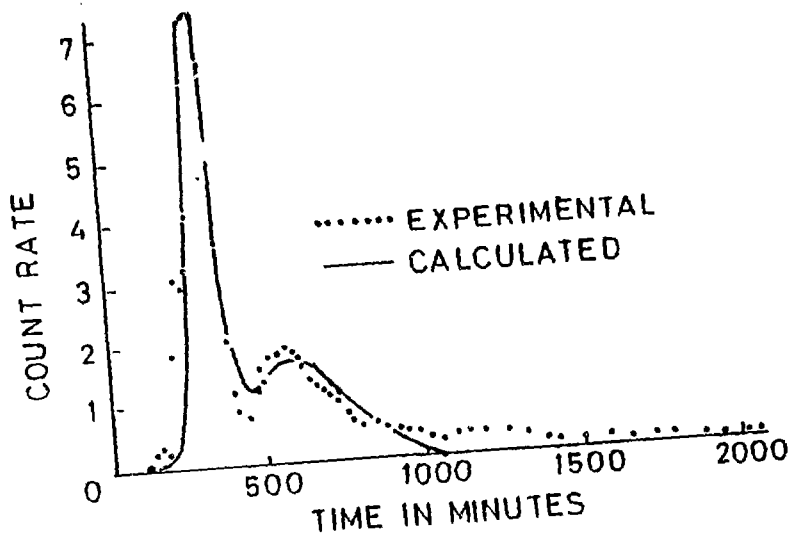


Figure 16a. Modified axial dispersion model applied to tracer response of crystalliser 5

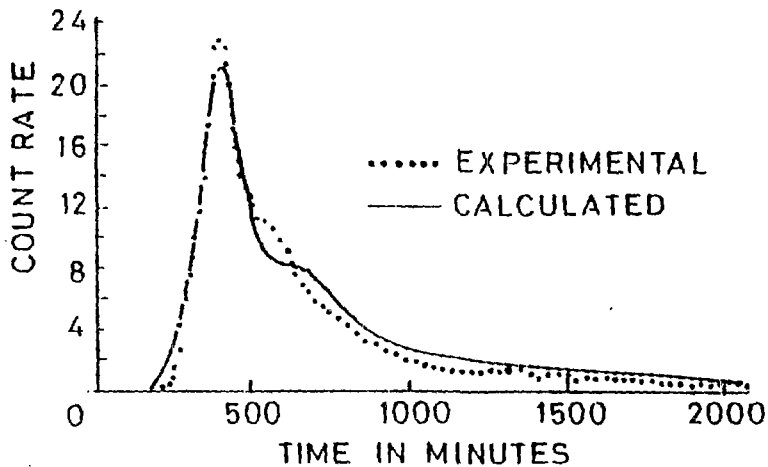


Figure 16b. Axial dispersion model applied to tracer response of crystalliser 6

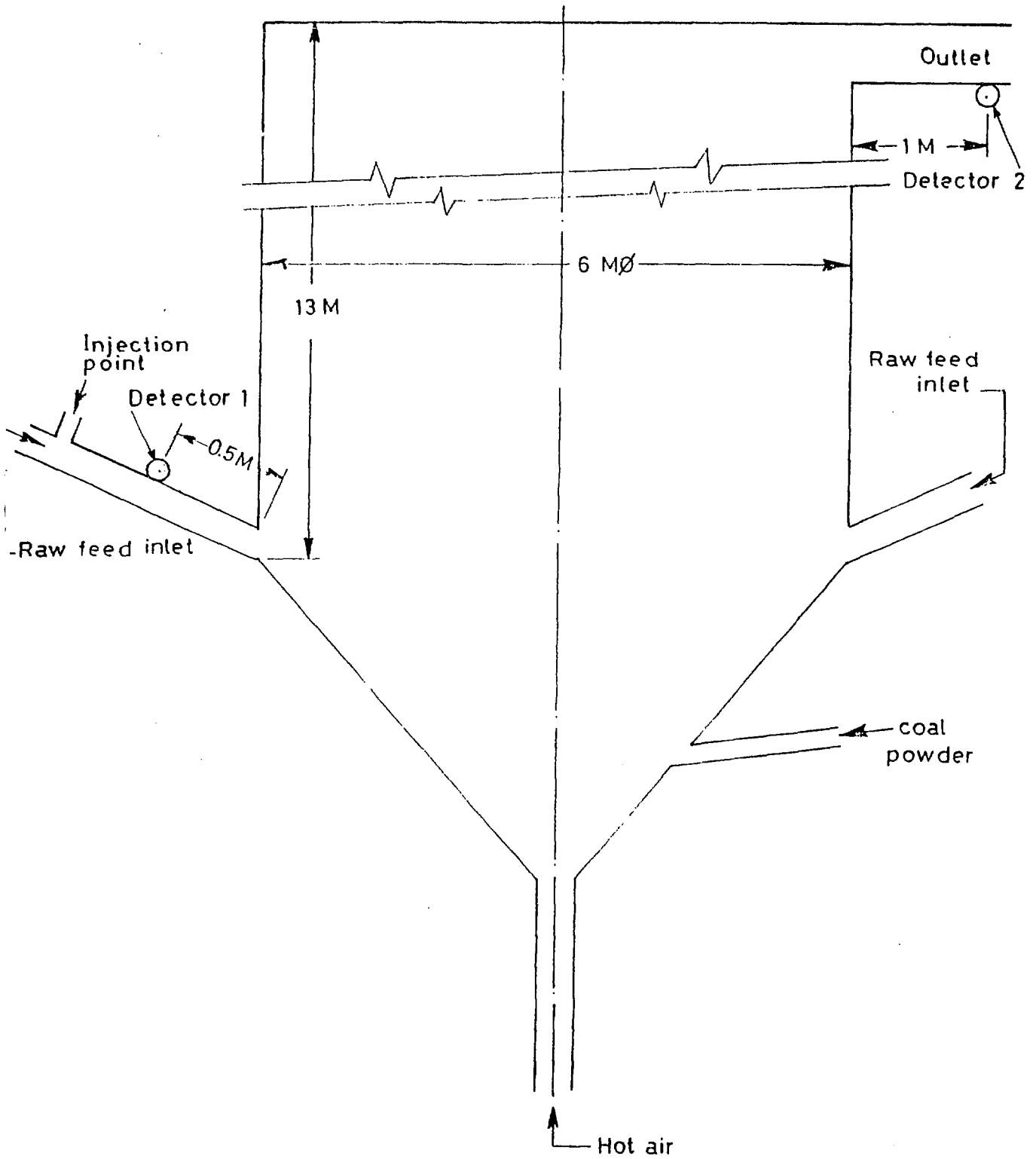


Figure 17. Sketch of precalcinator - injection and detection points

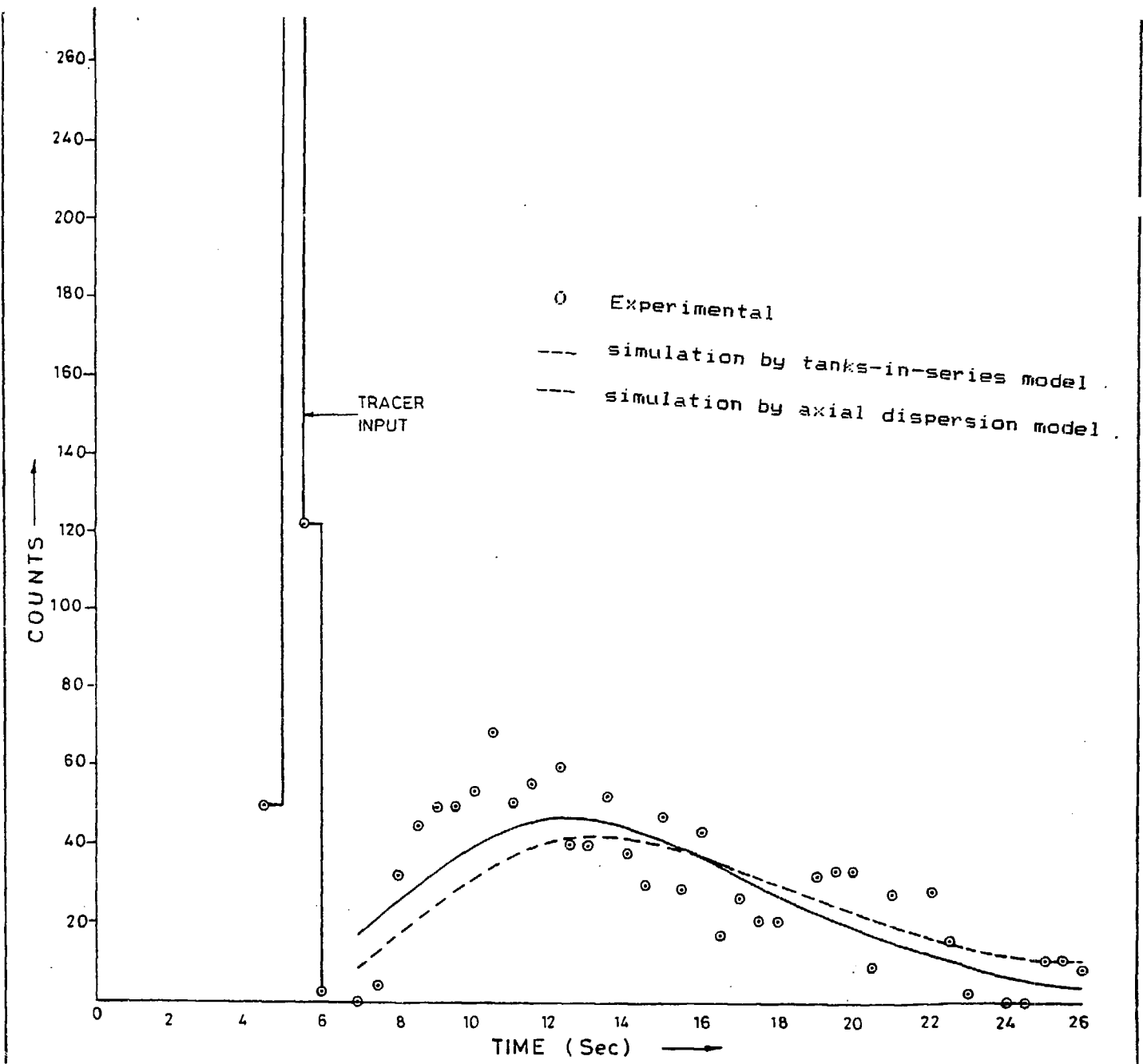


Figure 18. RTD in precalcinator - model simulations