THE INDUSTRIAL APPLICATION OF RADIOISOTOPES IN AUSTRALIA

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ABSTRACT

Over the past 10 years, the Australian Atomic Energy Commission has conducted a wide-ranging program of radioisotope applications to solve industrial problems of local, regional or national importance. Most of the investigations have been concerned with the behaviour of large complex systems. Broadly, the work covers such economically important fields as flow studies, environmental studies and coastal engineering studies.

INTRODUCTION

Since 1975, the Australian Atomic Energy Commission (AAEC) has had a group of scientists engaged in an on-going program on the development and extension of radioisotope techniques into industrial and related areas. Most of the radioisotopes used in this work are produced in the AAEC HIFAR materials testing reactor which is located at Lucas Heights.

Much of this effort has been in the field of radioisotope tracing techniques. Because the high sensitivity of modern nucleonic detection equipment allows very low levels of radioisotopes to be detected, it has long been recognised that they can be very effectively used to investigate large systems, i.e. those containing many tonnes or kilolitres of materials or material spread over areas of square kilometres. Radioisotopes can be used very effectively for large-scale investigations and there are few alternative techniques that can be as readily used, especially in a field situation.

Much of the AAEC work has been devoted to studying the behaviour of large and complex engineering or environmental systems. The information from such investigations may be required for many purposes; for example, it can be used to check whether systems are working in the way they were designed, or to develop mathematical models which are used to extend the applicability of specific observations. The development and verification of such models depends on measurements on the real system. Radioactive tracer investigations provide a vital step in this procedure.

Because radioisotopes are unstable and decay at known rates, they offer considerable advantages in a wide range of investigations requiring consecutive experiments where a suitably chosen radioisotope will effectively decay away before the next injection. Problems of radioactive contamination or build-up of tracer can thus be avoided.

Another consequence of the instability of radioisotopes is the absence of most of them in the environment. Rarely will any type of radioisotope, let alone the one chosen specifically for a particular investigation, be present in a system under investigation. Consequently, the radioisotope tracer will uniquely define material within the experiment.

If chemical species need to be monitored, the use of the appropriate radioisotope will ensure that the exact chemistry is followed.

In general, radioisotope tracers are required in only physically small amounts - milligrams of solids or millilitres of liquids - because of the large amounts of radioactivity that can be induced into many elements. The high sensitivity and stability of modern nuclear radiation detection equipment also ensures that relatively small amounts of radioactivity are required for most experiments. Radioisotope tracers can be added to most systems without affecting the material balance.

Many radioisotopes emit very energetic γ-rays that can pass through significant thicknesses of construction materials such as pipes and tanks. This radiation allows the system to be monitored externally, thus avoiding any perturbations that might arise from direct sampling. The γ-ray emissions also allow the monitoring of systems in real time which leads to maximum sampling efficiency and direct feedback on system behaviour.

For these reasons, radioisotopes are generally the most appropriate and cost-effective means of tracing large systems. Since the only indigenous supply of radioisotopes is in Sydney, a number of special logistics problems occur when work is carried out in other areas of Australia because of the large size of the country. Special techniques have been developed by the AAEC to overcome some of these problems, for example extensive use is made of ^{99m}Tc which is available from AAEC designed technetium generators with strengths up to 80 GBq.

The scope of radioisotope tracer work carried out by the AAEC has involved most sectors of Australian industry including iron and steel, coal, chemical, petrochemical, natural gas, metallurgical, mineral, power generation, liquefied air plant, as well as port authorities, water and sewerage instrumentalities, and environmental agencies. Generally, the type of work undertaken can be classified into three fields:

- Flow studies - involving flow rate measurements, residence times, leak-detection and wear.
- Environmental studies - examining the behaviour of existing pollution inputs into the environment or predicting the effect of new inputs.
- Coastal engineering studies - examining movement of sand in rivers, estuaries and near-shore areas, dredge spoil movement and sediment gauging.

Examples from each of these fields is discussed below.

FLOW STUDIES

A. Hearth Drainage in Blast Furnaces

The use of modern blast furnaces has revealed deficiencies in operating procedures developed for earlier types of furnace. Over the past 15 years, there has been a world-wide and vigorous program of research to understand the behaviour of the blast furnace, particularly the furnace hearth, in such areas as improvement of coke quality, and analysis of hearth drainage. Much of this effort has been devoted to developing mathematical models and the analysis of flow behaviour in laboratory scale models using glass bead/water systems.

Since 1979, the AAEC has carried out experiments at BHP Steel International on Nos 3 and 5 blast furnaces at Port Kembla (NSW) and on Nos 3 and 4 blast furnaces at Newcastle (NSW). The objectives are to measure the residence times of the molten iron and slag in the furnace, to determine whether there is a correlation between the furnace performance and residence times. The studies have also investigated the flow symmetry around the hearths.

Gold-198, silver-110m and cobalt-60, in the form of wire or foil, are used to label the molten iron, and lanthanum-140 and scandium-46, in the form of oxides, are used to label the molten slag. Because of the large number of variables operating in a blast furnace, a large number of experiments have had to be carried out to determine how sensitive flow parameters are to changes in blast-furnace behaviour and output.

Work to date has produced encouraging results. In experiments investigating the hearth drainage behaviour in the presence of two different cokes on two occasions the radioisotope tracer studies picked up a deteriorating flow behaviour over a three to four week period. Both preceded major production difficulties in the furnaces.

This work is being extended to investigate the behaviour of lead blast furnaces.

B. Waste-water Treatment Lagoons

Waste stabilisation ponds are used extensively throughout the world as the final stage in the treatment of sewage and other liquid wastes containing organic materials. In Australia, about 2900 hectares of ponds are in use in 320 country areas where the effluents are ultimately discharged into rivers. The correct functioning of these lagoons is, therefore, vital to the preservation of unpolluted surface waters.

In many cases, pond systems are providing effluents which do not meet the high pollution control standards set by the state, largely because of the poor understanding of pond function rather than limitations of the pond method. The processes that destroy pathogens and remove organic material are biological, biochemical and chemical, but the creation of favourable conditions under which these processes will proceed at the desired rates is dependent on physical conditions in the pond system. A number of workers have cited the hydraulic (flow) behaviour of ponds as one of the major areas in which more knowledge is needed. Although hydraulic behaviour, particularly, hydraulic retention time distribution (RTD), is one of the most commonly used parameters in the design and theoretical modelling of pond behaviour, it is one of the least researched.

Since 1979, the AAEC has used radioactive tracer techniques to study flow patterns and RTDs in sewage treatment plants in New South Wales.[1,2] The flows have been traced using bromine-82, tritium and technetium-99m but the two tracers mainly used are ^{99m}Tc (in the form of pertechnetate) and tritiated water (HTO). We have found that the pertechnetate ion is a conservative tracer for water over the day time scale (i.e. four half-lives). Technetium-

99m has the other advantages of short half-life (6 h), a weak (140 keV) γ-ray emission and is readily obtainable from AAEC designed generators at strengths up to 80 GBq for remote location work. The γ-ray has only a short range in water (half thickness 4.5 cm) making it an ideal tracer to study the fine structure of water flow. It has shown up stratified flow behaviour along thermoclines on hot summer days and has identified wind-induced preferential flow regimes on windy days. Long term flow behaviour is monitored using HTO, the limitation of the measurement of the T in the laboratory rather than the field environment, being more than offset by the known conservative properties of this tracer.

Experiments have been set up on operating pond systems which aim

- to look for variations due to seasonal and short-term weather factors,
- to improve the mathematical modelling of the RTDs,
- to correlate observed RTDs with the type of flow behaviour observed in *in situ* tracing experiments, and
- to study the efficacy and mechanism of baffling as a means of extending retention time.

These tracer studies have shown that the flow behaviour of individual ponds can deviate markedly from the design criteria. Weather has a marked influence on the flow processes.

C. Leak Studies in a Chemical Reactor

Identification of the absence or presence of leaks in industrial plant has traditionally been an important part of radioisotope tracing work. In a few cases, a yes/no answer is all that is required but most investigations determine the rate of leakage. The choice of radioisotope to be used depends on the nature of the industrial plant process. Typical of such work is an investigation carried out by the AAEC at a chemical plant in Sydney. One of the chemicals manufactured there is di-iso-octyl phthalate and the appearance of a discolouration in this material was thought to be due to a leakage of fluid from the 6500 L heat exchanger into the 20 000 L chemical reactor.

The reaction to produce the di-iso-octyl phthalate takes eight hours. During this time, the chemical reactor vessel is heated by a fluid which enters at 250-270°C and exits at 205°C. It was estimated by the company that the minimum leak rate to be detected would be 1 L of heat exchanger fluid into the reactor over the eight-hour process cycle. The high heat exchanger temperatures limited the use of a number of organic soluble radioisotope complexes. In this case, we used the 8-hydroxy quinoline complex of technetium-99m as the tracer material. This complex is readily synthesised in the laboratory and is thermally stable in the temperature range found in the heat exchanger.

A 40 GBq ^{99m}Tc complex was injected into the heat exchanger system at the beginning of the reaction cycle. One 18 L product sample was tapped from the chemical reactor each hour. These were cooled and the radioactivity level measured on site using a submersible scintillation detector with a 50 mm x 25 mm diameter NaI crystal. The 5 mL samples were also counted in a well counter at the AAEC Lucas Heights Research Laboratories within two hours of the end of the reaction. Eight 3 mL samples of the heat exchanger fluid were also counted in a well counter to monitor the level of ^{99m}Tc in the heat exchanger.

The results showed that any leakage in the chemical reactor was less than the minimum detectable level of 85 mL over the eight-hour process (at 99.7% confidence level). It was also shown that the tracer was stable over this time and did not plate out on the heating coils.

D. Wear Studies on a Jet Aircraft Fuel Pump

Jet fuels are highly refined and, in many cases, have to have added materials to lubricate the moving surfaces of the fuel pump. The selection of the best additive and its optimum concentration can be a long procedure. A radioisotope technique was used by the AAEC to establish these parameters more rapidly.

In the fuel pump studied, the main wear was on small piston rings. These rings were irradiated in the HIFAR reactor for 12 hours to produce significant amounts of tungsten-187 (half life 24.1 h). The specific radioactivity in each ring was 1.6 GBq/g. After decay of short-lived isotopes ^{52}V (half life 3.7 m) and ^{56}Mn (half life 2.56 h) the rings were reassembled on the pistons in the pump case. The feed for the pump was a 70 L storage tank which contained de-oxygenated fuel and the required level of additive. This fuel was passed through a 1.0 μm filter before the pump. The outlet from the fuel pump was connected to a 5 μm outlet filter which removed all wear debris. Two scintillation detectors with 25 mm by 36 mm NaI crystals were placed either side of the outlet filter to measure the build-up of radioactive wear debris. Heat exchangers were also present in the recirculating system to prevent the fuel heating up.

During the test, a number of other system variables was monitored; these included pump operating speed, the inlet and delivery pressure, the volume flow rate, the fuel temperature and torque. Each test lasted about two hours. Readings of radioactivity in the filter were taken every five minutes. These results were corrected for background contribution, for ^{187}W decay then plotted against time. The instantaneous wear rate was assumed to be proportional to the slope of the curve at a given time. Each test was run for 60 minutes once a steady wear rate had been established so that there could be an intercomparison between different test conditions.

The results from the experiments with the various base fuels and additives rapidly and clearly showed the comparative behaviour of the various components. The lubricating ability of the various additives could be compared and the optimum concentration determined. Additionally the persistence of the additive could also be investigated. Some additives were found to be rapidly washed off if a straight base fuel were substituted for the additive fuel whereas other additives were found to be quite persistent.

The utility of radioisotope techniques for assessment of wear in jet fuel pumps and the determination of optimum additive levels in the base fuel were readily shown. The radioisotope technique was at least ten times faster than the conventional assessment methods.

COASTAL ENGINEERING STUDIES

There has been a significant increase in interest in coastal engineering studies worldwide since continental shelf territorial rights were declared, and considerable research effort has been centred in the coastal, nearshore and continental shelf regions. In Australia, an average of \$760 million is spent annually on coastal work of which an average of \$220 million goes on capital works. Many of the problems are caused by either erosion or deposition of sediment, and quantitative field data (to prove existing sediment transport models or to provide an understanding of the basic morphological processes involved) are urgently required by engineers involved in oil and gas pipeline laying, sewage and industrial outfall design, harbour and port design and maintenance, coastal erosion control and estuarine siltation management.

A. Navigation Channel Infill Study

The operating economics of modern bulk carriers demand that vessels be able to enter and leave ports at full capacity under most tidal conditions and that maintenance dredging operations to keep these channels open must not be excessive. At present, the main shipping channel for deep draught vessels entering and leaving the Port of Brisbane follows a long and complex path through Moreton Bay to avoid the extensive sand shoals that block the northern approaches. The Port of Brisbane Authority has proposed that a new channel be dredged which could provide an alternative and more direct access to the port. The new route would involve upgrading an existing tidal channel and constructing a shorter (3 km) section through a sand bar to link it with the open sea. An understanding of sand migration in the vicinity of this section is critical to the success or failure of the proposal.

In this region of the east coast of Australia, there is a northward movement of sand; e.g. 5×10^5 t of sand moves past the neighbouring Gold Coast each year. The shoals across the bay entrance are the result of this long-term movement. If however, sand is still passing through the shoals en route to beaches further north, it will cut across the proposed channel and cause infilling. A preliminary two-month experiment traced the movement of chromium-51 labelled sand injected close to the channel mid point.[3] Movement of sand towards the northwest along the wave orthogonal was identified. This movement, driven by mass transport and wave asymmetry effects, is across both the tidal current direction and the channel alignment. It was calculated that the 3 km section would intercept 2.5×10^5t of sand annually. Investigations over a wider area and for 12-month periods were carried out using iridium-192 labelled sediment. The direction of sand movement, particle velocities and depth of incorporation were similar to the earlier study in the inner regions of the sand bar whereas the outer regions had higher particle velocities, more rapid rates of incorporation and the direction of movement was southeast rather than northwest.

B. Coastal Erosion Study

An investigation has been made of the movement of sand in and around sea-grass beds in Gulf St Vincent, near metropolitan Adelaide.[4] The area is experiencing beach erosion caused by die-back (~ 10%) of the stabilising weed beds. It is suspected that the introduction of sewage effluent into gulf waters 20 years ago initiated a die-back/erosion sequence in the sea-grass meadows whose matted rooted system stabilised the sea bed over hundreds of square kilometres of shallow (< 15 m) waters. Winter storms from the Great Australian Bight have extensively eroded the weakened beds and caused a deepening of some offshore zones. The resulting reduction in friction factors and damping effect has allowed waves of greater height to penetrate to the nearshore and beach zones causing erosion. In addition, increased sand mobility from an eroding area could be smothering an otherwise healthy bed elsewhere.

As well as implications for coastal management (predicted costs are \$1 million per annum) sea-grass loss, which is a world-wide problem, has important ecological consequences for Gulf St Vincent and elsewhere since sea-grasses provide a feeding ground and nursery for fish species and are the major primary producers within a detritus-based food chain.

Iridium-192 labelled glass was injected at four selected locations using an AAEC developed technique. The ^{192}Ir labelled glass was delivered to the experimental site packed in serum vials. These vials were loaded into brass holders fitted with two electric detonators. The brass holders were lowered to a predetermined depth in the ocean and the detonators exploded breaking the serum vials and releasing the tracer. This procedure gives a good initial dispersion of the ^{192}Ir glass and also provided an almost contamination free means of releasing the tracer.

The seabed monitoring of radioactivity is carried out using a submersible scintillation detector incorporating a 50 mm diameter by 25 mm long NaI crystal. The signal from the detector is fed to an AAEC designed field ratemeter and then recorded on a chart recorder. The radioactivity measurements are logged together with the boat position, which is got using state-of-the-art electronic distance fixing equipment. The areal dispersion of the tracer can be calculated using this data. For quantitative studies it is necessary to measure how deep the tracer has been incorporated into the seabed and this is achieved by monitoring the seabed cores. In this study each location was monitored seven times with excellent results.

The experiment was designed to investigate sea-grass tolerance to sedimentary processes (smothering), to compare rates of movement in heavily and weakly grassed areas, and to compare migration rate and direction as a function of water depth and wave climate. The areas included a nearshore erosion gully in 4 m of water, an eroded sand patch surrounded by healthy beds and a healthy bed surrounded by extensive erosion, both in 7 m of water; and an area of partial grass cover on the seaward limit of the meadows in 15 m of water.

Subsequent sand migration at each site was monitored and four complete scans of each area were carried out after storms. The centroids of the labelled patches have been calculated and the initial mean particle velocity and direction determined in each case.

Results showed an expected northeasterly movement in the nearshore zone and a surprising southerly trend in the deeper zones. Wave and wind data recorded during the test indicate that most sand transport occurs during winter storms from the south-west originating in the Great Australian Bight.

ENVIRONMENTAL STUDIES

A. Tracing Existing Pollution

A company located at Burnie, Tasmania was discharging sulphuric acid waste containing 27 g L^{-1} iron through a 3 km pipeline into the ocean at a depth of 30 m. Because ferric hydroxide contamination had been found on beaches adjacent to the plant, a study was commissioned to investigate whether the material discharged through the pipeline had given rise to this pollution.[5]

Ferric hydroxide on the seabed close to the pipeline was labelled with gold-198. Its dispersion was determined under calm and storm conditions. Tidal and wind-driven currents were measured and the oscillating wave generated currents were calculated.

It was found that the agglomerated floc remained trapped in a stable seabed layer which spread slowly over the seabed when wind velocities were less than 15 m s^{-1}. When wind velocities exceeded this level, the wave generated oscillating currents 30 m below the ocean surface were strong enough to raise the floc into suspension. The direction of wind-driven current determined the direction of movement of this suspended floc. Tidal currents in this area were found to be negligible.

B. Predicting Pollution Effects

Radioisotopes have been used to predict the behaviour of heavy metal components in effluents from proposed mining waste water releases. This work has provided valuable information on the behaviour of these metals which assists in the design of effluent release structures and hence helps to minimise the impact of the effluent on the environment.

In two systems studied in the Northern Territory of Australia by the AAEC, two significant components of the waste water were said to be zinc and manganese. Studies were undertaken on the transport of both these heavy metals.[6,7] The radioisotopes of metals have, suitably long-lived isotopes; zinc-65 has a half-life of 243 days and manganese-54 has a half-life of 312 days. Both isotopes can be readily measured at very low levels, using γ-ray spectrometry. Cost factors for the isotopes dictated a different approach in the experimental design. The experiments using the expensive ^{54}Mn were undertaken over a more restrictive area than those with ^{65}Zn.

In the studies with zinc-65, the isotope was injected into the river flood-plain system at the proposed effluent release site. The injection was made at a steady rate over a period of 36 hours. This time enabled a quasi-steady state condition to be established for the dispersion study and also minimised the tidal effects. Tritium, in the form of tritiated water (HTO) was injected with the zinc-65 so that the dispersion could be related to a conservative tracer. In the experiments 3700 GBq (100 Ci) HTO and 30 GBq (800 mCi) ^{65}Zn were used.

Water, vegetation and sediment samples were taken at known locations across the river and associated flood-plain. These results yielded data on the longitudinal and horizontal diffusion of the tracers. Water samples were separated into 'dissolved' and 'particulate matter' using a 0.45 μm membrane filter. The dissolved tracer was precipitated as ammonium phosphate salt. All samples were analysed using γ-ray spectrometry. The sampling program covered the passage of the main tracer pulse and was continued to allow monitoring of any held-up and released tracers.

Samples of soil and vegetation were taken for analysis in the dry season (six months later) and also at the start of the following wet season.

From these experiments it was possible to follow the passage of the tracers and determine retention times on sediments and vegetation. The dry season distribution of tracer could be assessed as could the tracer levels in new season growth in vegetation during the following wet season.

In studies with manganese-54, which is an expensive cyclotron-produced radioisotope, a different approach was used. The tracer was released as an instantaneous point source injection. Three additional tracers were also released. Tritium was again used as the conservative tracer reference; zinc-65 was added for direct comparative assessment; technetium-99m (half life 6 hours) was added to assist in direct monitoring of the pulse. The quantities of tracers used were: ^{54}Mn, 0.74 GBq (20 mCi); ^{65}Zn, 2 GBq (55 mCi); HTO, 37 GBq (1 Ci) and ^{99m}Tc, 37 GBq (1 Ci).

Samples of water, sediment and vegetation were taken at known locations during and after the passage of the pulse. Analyses of these materials yielded details on retention times of the heavy metals on sediments and vegetation.

These experiments yielded valuable data on the behaviour and dispersion of the heavy metals zinc and manganese in a flood-plain environment and the interactions of the metals with sediments and vegetation over a period of one year.

This technique has significant advantages over tracing with materials such as dyes, because of the direct relation between the heavy metal species under study and the tracer, the long time-scale over which the study can be made, and the direct reference to a known conservative material (HTO).

SUMMARY

Radioisotope tracing techniques are powerful tools for analysing the behaviour of large systems and investigating industrially, economically important processes. The results of radioisotope experiments can yield important information, for example, on parameters such as flow rates, mixing phenomena, flow abnormalities and leaks, pollution problems (current or predicted) and offshore sediment movement. A wide range of Australian organisations have experienced the beneficial uses of radioisotope tracer technology.

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