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# RADIO-ISOTOPES AS A TOOL IN STUDYING MIXING METHODS

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## 1. Introduction

Although naturally occurring radio active isotopes were first used in tracer experiments some forty years ago, the limitation in availibility and number and the high cost of these isotopes restricted the application of the tracer technique until the discovery that the radio active isotopes of the commoner elements could be prepared. In recent years the production of a wide range of radio active isotopes in the atomic pile "BEPO" of the United Kingdom Atomic Energy Establishment at Harwell has stimulated the uses of these isotopes in industry. About one hundred different radio isotopes are available, and statistics show that deliveries of these isotopes to industry have increased from 19 in 1948 to 1,180 in 1954, the latter figure including deliveries from the Radiochemical Centre at Amersham. If we include deliveries to hospitals, universities, etc., and export, the deliveries have increased from 135 in 1947, to 16,425 in 1954. The American figures for radio isotope deliveries are very similar, since, over a period of eight years from 1946-1953, nearly 50,000 deliveries were made from the Oak Ridge National Laboratories to hospitals, universities, research laboratories and factories.

Numerous examples of the uses of radio isotopes in industry have appeared in the literature. It will suffice to mention here one example of radio isotope application. The rate of wear in the lining of a blast furnace has been measured (1) by embedding small pellets of cobalt 60 in the refractory lining when the furnace is being built. By measuring the radiation from the outside of the furnace during operation and from small samples of steel from each batch, the rate of wear of the lining can be closely followed. This example suggests possible application in the fertiliser industry for use in studying the life of brick-lined reaction vessels and acid storage tanks, etc.

When we came to consider the branch of industry concerned with agricultural chemicals, we find that there has been during the last decade a wide application of radio isotope research, mainly concerned with the fields of soil science and plant metabolism, and not with manufacturing technique. It is true that uses have been made o..

radioisotopes for the determination of transit times of fertiliser materials through driers and coolers, etc., but apart from this there is little published information on applications of radioisotopes to the manufacturing techniques of the fertiliser and allied industries. We have, however, had occasion to use the technique of radioisotopes in studying mixing efficiencies, and the purpose of this paper is to report the development of our methods and the results obtained, in a way which it is hoped will be of value to those interested in the application of radioisotopes to the study of mixing and other problems in the fertiliser industry.

## 2. Objectives

We are not concerned in this paper with the efficiency of mixing of the major components of any mixture of chemicals since the efficiency of distribution throughout the mass can generally be satisfactorily determined by chemical analyses of samples. Rather do we wish to consider the distribution of materials present in small quantities where ordinary methods of chemical analysis become either insensitive or unduly time-consuming. In order to make the treatment general we shall refer to the major body of the mixture as "raw material" and the small quantities whose efficiency of distribution we wish to determine as "minor constituents".

## 3. Experimental

3.1. Preliminary experiments. One gram of $Na_2CO_3$, which had been irradiated with neutrons in the atomic pile "BEPO" for 1 week at File Factor 0.6, was obtained from the Atomic Energy Research Establishment at Harwell. From irradiation data it was calculated that the specific activity of this material was 8.3 mc/g. on despatch. Sodium$^{24}$ in the form of $Na_2CO_3$ was chosen, since it is a $\beta$ emitter and, therefore, gives a suitably high accuracy in counting, and has a half-life of fifteen hours. The short half-life of $Na^{24}$ was chosen as an important asset in these investigations, since it minimises the storage time necessary before the final product becomes safe for subsequent handling. Although activated $Na_2CO_3$ may not be a normal constituent of the mix and may not therefore give a completely true picture of the mixing efficiency, the alternative procedure of irradiating the minor constituent in the atomic pile might give rise to unsuitable types of activity which could not so easily be controlled. Sodium carbonate was chosen rather than the chloride because of the presence of $Cl^{36}$, $S^{35}$, $P^{32}$ and $K^{42}$ associated with irradiated NaCl.

From initial counting experiments in which radioactive $Na^{24}$ was dispersed in a 1 lb. sample of raw material, it was established that a total activity of 1.5 mc of $Na^{24}$ would be required per ton of final product, so that the number of counts from a 1 lb. sample would be sufficiently high to give at least 5% accuracy in the counting experiments.

3.2. Apparatus. After the preliminary studies it was decided to obtain for our counting experiments a cold cathode scaling unit developed jointly by Ericsson Ltd. and A.E.R.E. Harwell, and now available commercially as Unit Type 1221 C from Ericsson Ltd. This was used in conjunction with a high efficiency gamma counter tube, Type G 10 Pb, supplied by 20th Century Electronics and a Probe Unit Type 1014 A supplied by Fleming Radio Ltd. The E.H.T. voltage for the counter tube was obtained by using a stabilised Power Unit Type 1082 supplied by A.E.R.E. Harwell. There are many types of equipment now on the market, but the above were chosen as being most suitable for our own applications.

Owing to the very high sensitivity of the tube, and in order to minimise background radiation, it was necessary to make a lead container to screen the tube. This container which had lead walls of 1" thickness was designed to hold each sample in the same relative position to the tube during counting.

3.3. Final technique. It was decided that, in view of the large number of tests necessary in our experiments, we should restrict the counting time to 1 minute for each sample and do three counts on each sample. In order to obtain a sufficiently high number of counts from each sample, it was necessary to add $Na^{24}$ of activity 1.5 mc to each ton intimately mixed with the minor additive. These conditions were found to give a sample count of 900-950 per minute, a figure which agreed closely with our preliminary calculations, and which was sufficiently greater than the background count of 60/min. for statistical accuracy.

The procedure then, on receipt of the radioactive $Na_2CO_3$ from Harwell, was to dissolve it in water and spray it on to the minor additive while the latter was being agitated in a horizontal paddle mixer. The precautions from the health aspect mentioned later were strictly adhered to and close contact with the isotope was avoided as far as possible. The treated minor additive was then added to the one ton batch of raw material and, after mixing, a 1 lb. sample was drawn from each 1 cwt. during bagging. The samples were then placed in 1 lb. cylindrical snap-closure tins, which were each measured for radioactivity by counting for three one minute periods.

## 4. Treatment of results

The mixing efficiency of minor additives can be assessed by calculating the variance between the counts of the 20 samples from each one ton batch. The 20 groups of three counts obtained were examined in all cases by the method of Analysis of Variance. This gives an estimate of the variance due to error which includes the variance due to fluctuating background interference, errors in counting and in timing, and an estimate of the variance between samples which is the variance due to mixing.

## 5. Safety precautions

We were informed by the Industrial Advisory Group at Harwell that we should have to satisfy the local Factory Inspector and the Radiological Protection Service regarding the procedure we proposed to adopt. At a meeting with H.M. Inspector of Factories for Edinburgh and H.M. Medical Inspector for Scotland, it was agreed that the following precautions should be adopted:-

a) Overalls and some form of hair covering should be issued to all operators in the immediate vicinity during the experiments. These articles should be withdrawn and washed at the conclusion of the experiment.

b) Wearing of micro-filter masks should be enforced where there is danger of radioactive dust being present.

c) Radiation dosimeters should be used during the experiment to measure both the dosages received by personnel and the amounts of radiation present in various sections of the plant.

d) Personnel should be advised to wash thoroughly after the experiments are completed.

e) The "active" product should be kept in a section of the store which is sufficiently isolated from personnel until the activity has decayed to a safe level.

The Radiological Protection Service were quite satisfied that we were not presented with any serious hazard from either external or internal radiation at the levels of radioactivity proposed and did not consider it necessary that they should visit us.

In addition to the above precautions, we decided that radiation dosage tests would have to be done, and this involved all personnel in wearing small film badges supplied by the National Physical Laboratory. All stages in the mixing and sampling procedure were carefully worked out in advance. It is difficult to work out the safe exposure times for personnel in the factory, since the material is dispersed in the various mixing and conveying units, but a safe rule is to work at a maximum distance from the active material, since the activity falls off with the square of the distance. The results of the radiation film tests, which were assessed for us by the National Physical Laboratory, fully confirmed that our precautions from the health aspect were adequate, and that the exposure to harmful radiations had been negligible.

## 6. Example.

One example of the technique outlined above is the measurement of dispersion of minor additives in feedingstuff mixtures. Although this is not an example of application to fertiliser manufacture, it is chosen as a difficult and extreme case because here we have to consider the question of the product being safe for subsequent sale and for consumption by animals.

The problem of minor additives in feedingstuffs has arisen because of the significance now attached to the scientific feeding of livestock and the increasing use of antibiotics, vitamin supplements, and potent drugs such as Nitrofurazone, and Phenyl-arsonic acid which are present in the proportion of only a few pounds per ton. The dispersion of these materials cannot be adequately and quickly measured by chemical methods, but the use of radio-isotopes has enabled us to examine the efficiency of our mixing techniques and to suggest ways by which they can be improved.

In order to calculate the minimum storage time for feedingstuff containing $Na^{24}$ before consumption by animals, it was necessary to have some close idea of the safe dosage level for animals. The National Institute for Medical Research advised that a dose of 0.03R per week should not be exceeded and that this would be equivalent to an activity of 0.06 µc per day per kg. of animal weight. Considering pig feed, where the feeding rate is 50 g./day per kg. of body weight, then there should not be more than 0.06 µc per 50 g. of feed, i.e. 0.56 µc per lb. of feed. It might be noted here that his value includes a safety factor of 10. The level of activity which could be tolerated by humans is actually 0.3R per week. We were able to hold the few tons of feedingstuffs used in our experiments for periods of 14 days, but in actual fact, at a level of activity equal to 1.5 mc per ton, a storage time of one day would bring the activity well below the safe dosage level for animals.

## 7. Conclusions.

We have tried to show that radioisotopes constitue an efficient tool which may well be applied more widely in the fertiliser and allied industries. The potential medical hazards which might at first sight seem to outweigh the simplicity of the method can be efficiently controlled, and the extreme rapidity of

determining the outcome of trials compares very favourably with the time required for chemical analyses of equivalent accuracy.

## 8. Acknowledgments.

Grateful acknowledgments are due to Dr. N. Miller of the Physics Department of Edinburgh University for advice and help received in the preliminary radioisotope measurements, and to Mr. J.A. Mackenzie and Mr. H. Borland of our Production Department who carried out the factory trials and the statistical evaluation of the results.

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## APPENDIX

As an example of the results which can be obtained by the radioisotope technique in the study of a mixing problem, the counting results for addition of a minor constituent at the rate of 3 lb. per ton batch are compared in Table I with similar results for an improved mixing method. Twenty random 1 lb. samples are taken from each batch and the radioactivity of each sample is examined by three separate counts.

### TABLE I.

### Counting results.

| Mixing Method A | | | | Mixing Method B | | | |
|---|---|---|---|---|---|---|---|
| Sample No. | Counts per minute | | | Sample No. | Counts per minute | | |
| | (a) | (b) | (c) | | (a) | (b) | (c) |
| 1 | 856 | 884 | 880 | 1 | 979 | 985 | 977 |
| 2 | 797 | 777 | 775 | 2 | 922 | 965 | 936 |
| 3 | 843 | 845 | 808 | 3 | 948 | 1007 | 918 |
| 4 | 873 | 860 | 882 | 4 | 1056 | 1016 | 1059 |
| 5 | 948 | 946 | 958 | 5 | 944 | 990 | 957 |
| 6 | 990 | 957 | 995 | 6 | 981 | 1027 | 980 |
| 7 | 996 | 1010 | 1005 | 7 | 1011 | 993 | 1068 |
| 8 | 1011 | 1015 | 1040 | 8 | 1039 | 1016 | 1084 |
| 9 | 918 | 892 | 926 | 9 | 991 | 914 | 911 |
| 10 | 934 | 969 | 968 | 10 | 996 | 963 | 968 |
| 11 | 992 | 956 | 992 | 11 | 1054 | 1049 | 1033 |
| 12 | 848 | 875 | 850 | 12 | 1019 | 1025 | 1070 |
| 13 | 905 | 930 | 961 | 13 | 960 | 1020 | 1067 |
| 14 | 999 | 955 | 983 | 14 | 966 | 1015 | 985 |
| 15 | 1021 | 1008 | 987 | 15 | 940 | 1047 | 1029 |
| 16 | 942 | 963 | 960 | 16 | 900 | 930 | 951 |
| 17 | 956 | 1006 | 974 | 17 | 1082 | 1045 | 1083 |
| 18 | 877 | 846 | 858 | 18 | 929 | 949 | 976 |
| 19 | 933 | 895 | 868 | 19 | 966 | 1034 | 1010 |
| 20 | 942 | 895 | 903 | 20 | 1079 | 1102 | 1078 |

The interpretation of these results is shown in Table II.

TABLE II.

Mixing efficiencies.

| Mixing Method | % Probability of being within stated limits | | | |
|---|---|---|---|---|
| | 1 lb. sample | | 5 lb. sample | |
| | 95%-105% | 90-110% | 95-105% | 90-110% |
| A | 52.2 | 84.5 | 89.0 | 99.9 |
| B | 76.20 | 98.17 | 99.07 | 100.00 |

Although the results for 95-105% limits are given, it is believed that these conditions are too severe for a constituent present in such small amounts, and accordingly the wider limits of 90-110% are generally adopted. The results show that mixing method A would be less satisfactory for general use than the improved method B.