

APPLICATION OF RADIOISOTOPES IN SUGAR TECHNOLOGICAL PRACTICE AND RESEARCH

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Abstract

The use in sugar technology of various radioisotopes for measuring, analysing and research purposes is reviewed and discussed.

Introduction

Four years ago, J.K. Basson², a specialist of the South African Atomic Energy Board, presented to this association an excellent paper that discussed the special characteristics of the most important radioisotopes, the various kinds of radiation, the types of detectors and counters, and some examples of application to factory control. The present paper does not originate from a nuclear institution but from one of the sugar laboratories which only make use of the commercial radioactive substances and measuring devices for their special sugar technological research work. Therefore this paper will not enlarge upon the different radiators and detectors but will only give a general picture of how they can be applied for practical or scientific aims in sugar technology, including the culture of sugar plants and the utilization of sugar products.

Absorption of Radiation

The most simple application of radioactivity which is described in sugar literature, is a Soviet¹⁹ installation that is attached to a belt conveyor, in order to count the number of sugar bags which travel to the warehouse. It consists of a radioactive source on one side of the conveyor, and of an ionizing chamber on the opposite side. The latter is connected to an electronic device that registers the number of interruptions the radiation beam is subjected to by the shadow of the bags. Now the authors do not mention why this is done by means of radioactivity, as an ordinary lamp and a photo-electric cell would render the same service. Radioactivity is normally only then employed, when sources of light or actual light measuring are not applicable or at least not opportune. This situation arises, when such radiation is required that penetrates into non-transparent substances. Although X-ray lamps could also do this, the radioactive sources have some advantages. One of these is the closer contact to the objects that are being tested. The British Sugar Corporation developed a method in order to determine the evaporator tube corrosion with ³²P- sodium phosphate. A solution of this radioactive salt is filled into the steam chamber after the campaign and a Geiger counter is then inserted tube by tube into the calandria. Thus corroded tubes are indicated by a relay and sound signal. It is obvious that this experiment could not be carried out by use of an X-ray tube as a source of radiation.

A large field is open to the useful application of radioactivity, when it comes to level gauging in

non-transparent containers, if their solid, scummy or aggressive charge does not permit that other indicators are made use of.

A typical example is the lime kiln. The Ukrainian Sugar Factories¹³ have been fitted with more than forty installations, in order to control the charging of their lime kilns automatically. In order to achieve this aim, two radioactive sources and two detectors are mounted on opposite sides of the lime kiln, at the desired maximum and minimum levels. They give off siren signals as soon as the kiln commences to be too full or too empty. The Polish Sugar Industry¹⁴ has developed a similar apparatus, which consists of three radioactive sources and three detectors at a height of 3,6; 3,8 and 4m from the bottom.

A similar application exists for the beet or cane feeding tanks of continuous extractors. Already in the nineteen fifties, the German Sugar Factory Neu-Offstein introduced into its beet tank a steel tube, which contained a long wire of the radioactive cobalt isotope ⁶⁰Co, and fixed a Geiger counter to the outer wall of the tank. The more the tank is filled, the more the length of the wire is covered by beets and the more radiation is absorbed by them. Thus the apparatus does not only give "yes—no" signals at certain levels, but above that shows the actual level quantitatively. Furthermore, it is able to control the stream of beets from the flumes automatically. Devices of this type were used as well in the level gauging of beet pulps and of molasses¹⁰.

As far as level gauging is concerned, we only regard the two possibilities: That either the radiation is absorbed by the material or not. But sufficiently strong radioactive sources offer further possibilities. With these one may measure a partial grade of transparency, if the layer of the material is thin enough. The absorption of gamma or beta radiation increases with the mass of the material that has been penetrated, i.e. it depends on the density and the thickness of the material. The function is similar to Lambert's and Beer's law for visible light. We are therefore given the opportunity to weigh the mass of the material by measuring its absorption of radiation. Generally, there is no need to do this, as mechanical balances are less complicated and cheaper. Yet for the continuous weighing of a stream of material, the isotope method does possess certain advantages when compared with conventional belt weighing. For instance, the weighing element does not possess moving parts and can easily be installed on existing conveyors. The pioneers who introduced this technique to the sugar industry were in Hawaii⁴. They employed it in order to weigh the raw sugar mass which passed the main conveyor, in the bulk loading terminal of Kauai.

The uncertainty of the rate of movement of the belt was eliminated by measuring the absorption of rays in a free falling stream of the sugar. In this device, gamma rays emitted from $^{137}\text{cesium}$ are partially absorbed by the falling stream of sugar and their diminished intensity is measured by a Geiger counter and compared with the original intensity either by an occasional interruption of the sugar stream or by a second Geiger counter in a null-type circuit. This method did not prove to be as successful for raw sugar as it was for bagasse. Numerous bagasse weighers of diverse design have been set up in Hawaiian sugar factories. The initial experiments were also carried out in a free falling curtain of bagasse. Later on however, it was preferred to measure the absorption of the passing bagasse directly on the conveyor. Normally, the source of cesium is attached to one side and the Geiger counter to the opposite side of the conveyor. Subsequently the absorption is registered by a recorder and the total weight is obtained by a planimeter or by electronic integrators. The different installations have been closely observed in Hawaii for four years. The result gained thereby was that the actual weighed mass of bagasse can deviate from the calculated inferential mass of bagasse by + 10 to -22,5%. These deviations correspond to an effect on Mill Extraction of approximately -5 to + 1,5%.

Not only weight can be gauged by such methods, but also density, when the thickness of the material is constant as in the case of liquids in a pipe. This fact was used in the Soviet Union¹⁷ for the automatic measurement and the control of density of juice in evaporating stations. In order to achieve this, the radiation of an isotopic source passes across the juice pipe to a detector, which is empirically calibrated in density units.

A method for the rapid determination of lime in milk of lime, by means of backscattering of beta rays, was developed in the Hungarian Sugar Industry²⁶. A probe contained a source of ^{90}Sr and ^{90}Y , mounted in a lead ring which was covered with a thin plastic foil. The backscattered radiation was detected by a Geiger counter; this radiation varies linearly with the content of CaO in the solution.

Tracing by radiation

A further range for the application of radioisotopes is to trace the pattern of particles in a stream of material. As far as possible, such experiments are made by dyeing the particles in parts of the stream and then observing the movement of the colour. However, very often this simple method is not applicable e.g. in non-transparent liquids or slurries. When such cases arise, then it might be helpful to use a radioactive instead of a dyed marking and to observe the movement by suitable detectors. In Australia the Mackay Sugar Research Institute¹ has studied the flow pattern and the speed in vacuum crystallizers, by recording the movement of a radioisotope, which was enclosed in a capsule of the same density as the massecuite. The results

obtained showed, that only for one-third of the time does the circulation of the boiling massecuite follow the simple circulation pattern. It was found that local circulation did occur in spots for periods ranging up to five minutes. The movements were often sluggish and indefinite.

The Swedish Sugar Research Laboratory²⁷ carried out tests in order to determine the retention time of the liquid phase and the mud in subsiders. The average retention time can be calculated quite easily. Nevertheless, it was interesting to study the time and the pattern of particles which find an opportune or not so opportune way through the labyrinth of the subsider. At a certain moment therefore, a quantity of sodium bicarbonate, with the radioactive isotope ^{24}Na , was injected into the slurry at the top of the subsider. Then the time was measured until the first and last appearance of radioactivity in the different tubes of the subsider. The experiment was repeated once again later on, but with the addition of radioactive ^{140}La , in form of lanthanum hydroxide, thus making it possible to follow the pattern of the solid particles as well.

As these experiments were not carried out in a laboratory but in a sugar factory, it was essential that such radioisotopes were selected that possess a very short half-life. So they lose their activity fast enough for the end product to be sufficiently inactive for human consumption. This is indeed a disadvantage for the food industry for all applications of radioisotopes which cannot be strictly isolated from the products as it was possible to do in the gauging devices. In order to make use of short half-life isotopes, local equipment for the isotope production by irradiation of the inactive compounds is required. The half-life time of ^{24}Na is 15 hours, and that of ^{140}La 44 hours. Later on, similar experiments were made with other types of subsiders in Australia⁸, for which ^{82}Br (half-life being 36 hours) in form of ammonium bromide, for the liquid phase was used. The detection was effected in Sweden and in Australia by means of scintillation counters.

Polish scientists¹² made use of radioactive tracers in order to carry out tests with continuous extractors of the DDS type. They distributed 10 scintillation counters, 5 on each side, along the trough of a DDS-extractor. Then they marked some individual beet cossettes with small glass needles which contained ^{14}Na . The cossettes possessed a mean speed of 0,25m/min. of net longitudinal movement, but they did not move along the trough linearly and were subjected to considerable mixing. In order to study the movement of the liquid phase also, experiments were made with a solution of ^{140}La , in form of a complex which the cossettes do not absorb. This marker was introduced at approximately $\frac{1}{4}$ of the length of the trough from the water inlet end. Thus the movement of the liquid phase could be observed not only in the normal but also in the upward direction. Indeed, the juice tended to be carried upwards with the cossettes to a considerable extent.

The mechanical movement of material can only then be correctly traced with markers, if these possess the same solubility, density etc. as the material in question. Beyond that, the radioisotopes offer us the unique possibility of carrying out tests with such markers that are completely identical with the material, not only in the physical but also in the chemical respect. This enormous advantage over all other marking methods is attained through the addition of small quantities of chemical compounds which are wholly identical with those of the material, but contain radioactive isotopes as one of their constituents, e.g. sucrose with the radioactive carbon isotope ^{14}C . There is no need that all the carbon atoms in the molecules are ^{14}C and it is even not necessary that all the molecules of such a "labelled" compound contain ^{14}C . It is only important that there are sufficient ^{14}C atoms in the substance to be detected by the means available, and that these atoms are well distributed through the labelled substance and that this substance has to be carefully mixed with the material in question. In this case, the fate of the original substance in the tested material and the radioactivity is inseparable through any possible physical or chemical change, which might even be the total destruction of the molecule.

As regards the radioactive carbon isotope ^{14}C , this is produced from nitrogen by way of neutron bombardment. Hundreds of ^{14}C compounds are commercially available, amongst them sucrose, glucose, fructose, and other significant carbohydrates, also amino acids and numerous other organic compounds which are important to sugar technology. The half-life of ^{14}C is 5 500 years. Therefore it is only suitable for laboratory work and must not be included in products which are intended for human consumption. For experimental aims, carbohydrates can also be labelled with radioactive hydrogen ^3H ("Tritium").

The Research Laboratory of the British Sugar Corporation⁵ made a simple and typical use of ^{14}C -labelled compounds as tracers. They had in mind to test the efficiency of a new method which separated the lactic acid from the juice by ion exchangers. Therefore they added a small quantity of ^{14}C -labelled lactic acid to the juice and applied the juice to the ion exchanger and examined the radioactivity of the remainder. They found that about 10% of the added activity — i.e. 10% of the added lactic acid — was still in the remainder. Consequently also 10% of the total lactic acid was still in the remainder, as the mixture of the original nonradioactive and the added radioactive lactic acid is not to be separated under any circumstances. So the tested ion exchange method had to be improved until the radioactive tracer proved it efficient enough.

The case may arise, that when a chemical compound is isolated for analytical purposes, no reaction is to be obtained that is sufficiently quantitative. When faced with that problem, then the radioactive tracer may nevertheless help in the receiving of a correct, analytical result. If for

instance the above mentioned ion exchange separation is used for the determination of lactic acid, and 10% of the added activity is still in the remainder, then it is obvious that the weight of the separated lactic acid is 10% too low and consequently has to be corrected by adding 10% in order to acquire the true result.

This is the principle of the so-called isotope dilution methods, which enable us to correct analytical results when confronted with non-quantitative separation methods. The first isotope dilution method for the determination of sucrose was evolved in the Berlin Sugar Institute¹¹. In order to avoid the errors of the polarimetric determination of sucrose by other optically rotating substances, it was tried to develop a gravimetric method of determination. The precipitation of sucrose with copper sulphate was found to be outstandingly specific, but unfortunately not quantitative. Therefore a small quantity of ^{14}C -labelled sucrose was added before precipitation. Subsequently the gravimetric result was corrected in accordance with the ratio of the total radioactivity which had been added to the radioactivity recovered in the weighed precipitate.

This method is used especially in order to check the conventional methods of sucrose determination in sugar beets. In this case the polarimetric methods are not only influenced by other optically rotating substances but were also found to possess systematic errors which are caused by the uncertainty of the real volume of the beet marc. There again the isotope dilution method has practically no systematic errors. So it was also applied for the determination of sucrose in sugar cane, in molasses and other sucrose containing material. Later on, another isotope dilution method was developed in the United States²⁴, which used the precipitation of the sucrose as barium sucrate which also is specific but not quantitative. Both methods are too complicated for routine work in sugar factories, but they are undoubtedly the most accurate methods for the determination of sucrose.

Still more interesting is the use of labelled compounds as tracers, in order to investigate the mechanism of complicated reactions. The most remarkable field of application is the formation of sucrose in plants. The procedure which is commonly employed in order to investigate photosynthesis, is to expose foliage to labelled carbon dioxide for a short period of time. Then the plant is harvested and the radioactive products which the plant has meanwhile synthesized are examined. In experiments carried out on sugar beets, Russian scientists¹⁶ realized that after an exposure of five minutes, the largest part of the radioactive carbon was found in the leaves in form of sucrose. The formation of radioactive glucose and fructose took a longer period of time. That means that sucrose is the first free carbohydrate which is formed by photosynthesis in plants. Glucose and fructose are formed later by an enzymatic cleaving of sucrose or by dephosphorilization of hexosephosphates.

Notwithstanding, the foliage also possesses the ability to re-form sucrose from glucose and fructose. Although the leaves are able to form sucrose directly by photosynthesis as well as by condensation of glucose and fructose, the sucrose content of the leaves is low. The answer to this lies in the fast transport of sucrose to the beet root. The entire sucrose which is in the root was formed in the leaves, as the root of the adult beet cannot form sucrose from its components. These results were obtained by pursuing the labelled sucrose and glucose respectively, during their transport from the leaves down to the root. As regards sugar cane, there the conditions for the formation and transport of sucrose from the leaves to the stalks are comparable. However, the velocity of the transport of the assimilate in the beet is 40 to 120cm/h, in cane only 4 to 6cm.

With the aid of labelled compounds, numerous other experiments³ were carried out on beet and cane, e.g. formation of plant acids, fatty acids, amino acids and proteins, aspiration and fertilization. Fertilization experiments were chiefly made with phosphate, as phosphate compounds can easily be labelled with the isotope³²P. The velocity of acceptance by the roots, the transport to the different parts of the plants and the chemical transformations in the plant of various phosphate fertilizers were investigated. Some experiments were also made with radioactive calcium (⁴⁵Ca).

The above mentioned investigations provided us with further knowledge about the complicated process of sucrose formation in sugarcane and sugar beet. In the same way research work has already been done, which concerns the fate of sucrose and other significant constituents of cane and beet during the technological process of sugar production. Many experiments were carried out in the Berlin Institute²¹, in order to prove that the speed of the extraction in beet or cane extractors ("diffusers") is not caused so much by diffusion of the sucrose molecules in the liquid, but mainly by the exchange of the liquid itself. In one of these experiments, sugar beets with radioactive marked sucrose were sliced and the cossettes treated with warm water of 78°C or with press juice of 78°C. When using water for the treatment, a rapid extraction of the sugar was obtained while a slow extraction could be observed when using press juice (after an initial rapid separation of radioactive marked sucrose which was caused by physiological qualities of the plants). These test data furthermore indicate that the extraction of sugar from beet cossettes is obtained for a minor part by diffusion (caused by thermal molecular movement) and that, instead, it is almost completely caused by another physical process which must be caused by a difference of the sugar concentration in the beet cells and of the surrounding solution. As the exchange of liquidity previously proposed is caused by such a concentration difference, the test data might furthermore indicate that the sugar extraction from beet cossettes in an industrial scale is

effected by an exchange of liquidity.

The British Sugar Corporation⁵ made an extremely interesting investigation. The origin of the oxalate scale, which is deposited in evaporator tubes has since been controversial, although the solubility of calcium oxalate is extremely low, the possibility might arise that raw juice oxalate is not completely eliminated in the process of juice purification. Alternatively, there is the possibility that oxalate may be formed by thermal degradation of saccharides during the evaporation. In order to solve this problem, the scientists made two experiments. In the first one they added labelled oxalic acid to raw juice, purified it in the usual manner by liming and two carbonatations and determined the residual activity in the purified juice to be 3,3% of the original value. They concluded that a remarkable part of the raw juice oxalate is able to pass through the usual juice purification. In the second experiment, they added labelled fructose to a solution of fructose and a suitable buffer substance. This solution was heated vigorously in order to degrade fructose. Then they precipitated the formed oxalic acid and determined its radioactivity to be only 0,1% of the original value. Therefore they deduced that the oxalate scale in evaporators is not caused by degradation of monosaccharides but by raw juice oxalate which has not been removed during the process of juice purification. The same British team⁵ accomplished a radioactive study of the degradation products of fructose on liming and they found primarily saccharinic acids, lactic and glycolic acid.

Several co-operating American sugar institutions²⁰ have developed techniques in pilot plant experiments which enable them to observe the fate of cane constituents during the technological process and molasses formation, with the aid of either labelled glucose, fructose or sucrose. With the aid of these techniques, the New York Sugar Trade Laboratory carried out a series of investigations. They realized that the action of heat on limed cane juice led to the formation of hexose interconversion, condensation and polymerization products. D-glucose and D-fructose interconversions were established by chromatographic and radio-tracer studies. Residues from both hexoses were present in the molasses melanoidins. D-fructose yielded several simple condensation compounds. In another investigation the probable formation of D-mannose in small amounts during the heating of limed cane juice was established by carbon-14 tracer studies and paper electrophoresis. More D-mannose was produced from D-fructose than from D-glucose. A continuation of the research work showed that the probable formation of D-psicose in trace amounts by the epimerization of D-fructose during the heating of limed cane juice was established by carbon-14 and paper chromatographic techniques. In other experiments the fate of the principal amino acids of cane during the formation of molasses was studied utilizing ¹⁴C and ³H-labelled amino acids. These were added to solutions of lyophilized cane juice solids which

were then heated under circumstances similar to those of molasses formation in factories. The browning polymers of hexoses and amino acids consisted of units that contained 52 to 53 asparagine, 5 gammaaminobutyric acid, 2 to 3 proline, 2 aspartic acid, 2 glutamic acid, 2 alanine and 1 valine residue.

Labelled sucrose was also used as a tracer for research work about sugar consumption. The vast range of possibilities that exist in this respect may best be illustrated by a test about the fate of sucrose during breadmaking¹⁸. The bread was made whilst labelled sucrose was added to the dough. As much as 42% of the activity remained in the finished bread and nearly 20% could be recovered in the form of hydroxymethylfurfural in the condensate of the oven vapour. The rest was undetectable, that means probably volatile. The main part of the residual activity which was found in the crumb and crust of the bread, consisted of neutral compounds. The paper chromatography of this neutral fraction showed some unidentified components as well as glucose and fructose in the ratio 1 : 3,5.

An investigation which was made on tobacco additives and cigarette smoke⁷ shows what great changes can still be pursued by radioactive tracing. Labelled sucrose was added to tobacco. The cigarettes which were produced from this tobacco were smoked mechanically. Now 60% of the activity was transferred to the smoke in the form of benzene, acetonitrile, carbonyl compounds, furans and others. In the cigarette butt 40% of hydroxymethylfurfural, 1,6 anhydro- β -D-glucopyranose etc. was found.

Japanese scientists⁹ incubated human dental plaque for two hours in solutions of labelled sucrose at a temperature of 35°C. 4% of the polysaccharides was synthesized, one eighth of them was levan. A conversion of fructose into glucose did not occur during the synthesis.

In addition to such applied research, fundamental research work was also done on transformations of labelled sucrose. The Berlin Sugar Institute²² studied the mechanism of the biochemical transformation of sucrose into a similar disaccharide 6-(alpha-D-glucopyranosyl)-D-fructose, which also consists of one glucose and one fructose moiety. However, this disaccharide, so-called palatinose, is linked in another way than sucrose. By employing active sucrose, glucose and fructose, we could ascertain that the reaction is not an intermolecular movement of the bond but a dimolecular reaction between one sucrose molecule and one cleaved glucose moiety.

An interesting question concerning the inversion of sucrose is which of the two components, glucose and fructose, possesses the original oxygen atom of the glycosyl bond or which of them has to take the oxygen from the hydrolyzing water. The question was answered by the inversion of sucrose in water enriched by ¹⁸O : ¹⁸O was incorporated by glucose as well as by fructose, the ratio being 1 : 3. We may therefore deduce that the bond is able to cleave in both

positions⁶.

A vast region for still further research is disclosed as it is possible not only to label molecules but also certain parts of these. Whilst the radioactive saccharides which have been obtained through photosynthesis in radioactive carbon dioxide are uniformly labelled, one may produce carbohydrates synthetically, in which only certain carbon atoms are radioactive. If e.g. sucrose is synthesized by one of the known reactions of active glucose but inactive fructose, only its glucose component will actually be labelled. Sucrose can also be synthesized in an analogue reaction, in which only the fructose component is labelled. It is quite obvious that this opens the door to numerous physiological experiments about the varying fate of the two components of sugar in the animal organism. One can derive from the blood or even from the expired air of an animal, how much of the blood glucose or of the expired carbon dioxide respectively are formed from the fructose component of the consumed sucrose etc. Even single C-atoms can be marked by a suitable synthesis of saccharides. Some monosaccharides which have only been partially marked are commercially available.

Effect of irradiation

Now in all the cases at which we have looked up till now, the radioactivity has been used in order to indicate something. We still have to deal briefly with another group of application and that is the one which alters²³ materials by way of irradiation. The gamma rays which are released by radioisotopes may be utilized for the purposes of sterilization, for the discharge of mutations etc. They offer the advantage of a closer contact to the material than X-ray tubes do. The cobalt isotope ⁶⁰Co is mainly employed in this respect.

So it is understood that the breaking of bonds in the sucrose molecule yields a number of compounds as a result of quantum absorption of gamma rays. After an intense and long irradiation, specially inversion is observed. Inulin is hydrolyzed into fructose by irradiation. Monosaccharides are subjected to further degradation. Also gas is formed, primarily hydrogen. Free radicals come into being and are the cause for further chemical reactions.

Irradiation by radioisotopes was also applied to plants, including sugar cane and sugar beet¹⁵. The application is made possible by irradiation of the seed through an exterior source or by soaking the seed in a radioactive solution. It is further possible by the irradiation of the plant during the vegetation period, either through an exterior source or through a radioactive fertilizer.

It is well-known that the irradiation of plants can cause mutations which may be exploited for the object of breeding. The success of such attempts is only small, as mutation is a random effect. The influence of the irradiation on non-hereditary alterations of sugar plants appears to be much more remarkable. Stimulation of the ger-

minating capacity, of yield and of sucrose content was observed by different authors. It seems to be evident, that after the irradiation with appropriate doses of gamma rays, less sugar losses are visible in the beets after long storing period²⁵. This is, however, still far from the possibility of a practical application.

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