

REMOVING RADIOACTIVITY FROM MILK

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Abstract—The need for measures to reduce public exposure to environmental radiological contamination from accidents and fallout has led to a program of research, development, field testing, and operations by the Public Health Service in the United States over a period of several years. This effort has now been focused to form a consistent pattern under the guidance of the Federal Radiation Council. Laboratory, field sampling, and evaluation experiments related to radioactive iodine will be presented. The data from various research investigations useful to operating agencies in the benefit-risk decisions will be described. The research, development, and large-scale testing of methods for concurrently removing anions and cations from milk during processing will be described including presentation of data from both laboratory and large-scale experiments. Cost data related to some large-scale experiences will be given where it would be useful for comparative purposes. Presently, for example, the work indicates that a ^{90}Sr removal process alone will add approximately 2 cents per quart to the cost of milk. Indicators of possible ways to reduce this cost by combining control procedures, such as for ^{131}I and ^{90}Sr , will be given. The administrative procedures used to initiate each phase of the laboratory research, development, and field evaluations of these systems will be described.

INTRODUCTION

Historically, the Public Health Service has had the principal Federal responsibility for protecting the public from health hazards in the United States. In the radiological health field, the Public Health Service has adopted a two-prong approach; first, to determine where the radiation health hazards exist, and second, to do something to eliminate them. This approach might be termed one of "surveillance plus action". This program is carried out by the National Center for Radiological Health of the Public Health Service.

To demonstrate how this approach works, let me refer to Fig. 1 which is derived from the U.S. Federal Radiation Council chart showing several important paths of ingestion of radioactive material in man. It is not the intention to cover the entire National Center for Radiological Health program, but to briefly describe the range of our surveillance-plus-action approach and then focus on how this approach was carried out in the development of practical methods for

protection of the public from radiation exposure through milk.

You will note in Fig. 1 that tobacco and water have been added to the Federal Radiation Council chart as being part of the chain of human ingestion. Under the "surveillance" portion of our program, the Center now monitors the amount of radioactive traffic along several of these paths through extensive networks. The Center monitors the amount of radioactivity in the atmosphere and the extent to which it is deposited on the earth's surface. The amounts of radioactivity in water, milk, and food are also regularly determined. This information is summarized and made available to the profession and the public regularly through Radiological Health Data and Reports, a publication of the National Center for Radiological Health. ⁽¹⁾

This surveillance is essential in that it indicates existing potential problem areas of environmental radiation exposure and guides the direction of our "action" program. Some of our

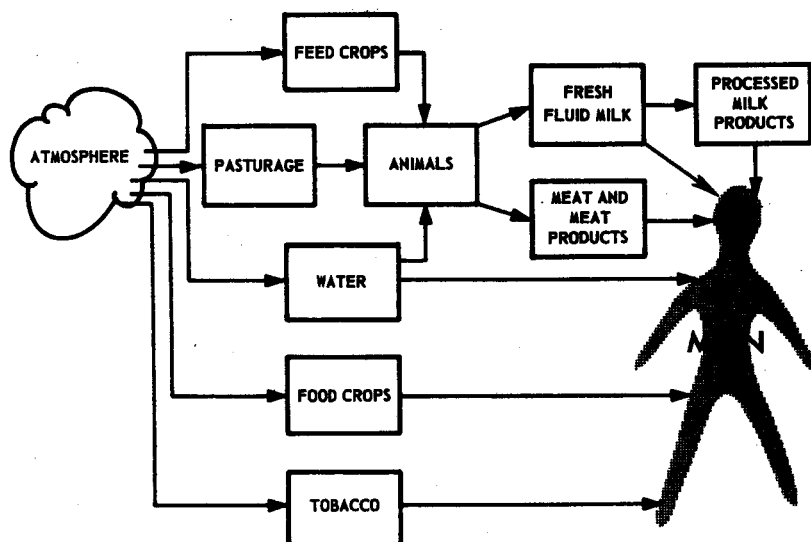


FIG. 1. Important paths of ingestion of radioactive materials in man.

action consists of notification of states or industry of the data observed. A system through which surveillance networks can be used to implement the Protective Action Guide applicable to ^{131}I recommended by the Federal Radiation Council has been described earlier.⁽²⁾ This system can be applied on a regional, state, milk shed or individual farm basis. Other actions consist of research on ways to break this transmission chain at several points. For example, the Center is now doing research aimed at controlling uptake of radionuclides from the soil into feed crops. It is also investigating methods for removal of radioisotopes from tobacco, and has two projects under way to determine practical methods for reducing the uptake of radionuclides in meat and milk from animals which eat feed containing high levels of radioactivity.

One of the major efforts over the years has been directed at the problem of radioactivity in fresh milk as a result of fallout. This paper will describe this research and development in some detail. In the United States, the persons responsible for weapons development began in the early 1950's to be concerned about the projected increase in strontium-calcium ratios in the environment and to a degree based their testing plans on learning more about this factor

with the aim of reducing the amount of strontium released to the environment. The Public Health Service began in 1954 to analyze selected U.S. milk supplies for certain radioisotopes. By 1958, the levels of ^{90}Sr observed in this milk indicated that the maximum permissible concentration (MPC) which existed at that time for milk might be exceeded in some locales in the United States. At this time, the consensus of testimony before Congressional hearings on fallout in milk was that the MPC should be between 33 and 150 PCi/l. , with the most commonly discussed figure at 73 PCi/l.

Because of the long half-life of ^{90}Sr and its biological similarity to calcium, it is generally regarded to be one of the principal fission products which may affect the health of humans. Since milk is such a basic food and is the main source of calcium for most western countries, it therefore has received principal attention as a health hazard.

Several laboratories have done work which indicated that an ion-exchange process could be developed for removing ^{90}Sr from milk. As early as 1954 the University of California investigated radiostrontium removal from milk by ion-exchange under contract with the U.S. Atomic Energy Commission. There were also the studies of Migicovsky⁽³⁾ of the Canadian

Department of Agriculture, studies by the British Atomic Energy Research Establishment,⁽⁴⁾ the work sponsored by the Atomic Energy Commission of the University of Tennessee^(5, 6) and studies by the Public Health Service in Cincinnati.

ORGANIZATION OF RESEARCH AND DEVELOPMENT

There was thus at this time (1958) strong indications that a practical process for removing ⁹⁰Sr from milk could be developed. Since no private milk producer could have been expected to underwrite the necessary research, the three government agencies most vitally concerned (the Atomic Energy Commission, the Department of Agriculture and the Public Health Service) entered into a cooperative agreement to underwrite jointly the cost of a program to develop a feasible process for removing ⁹⁰Sr from milk. The program, financed equally by the three agencies, was carried out in the Agricultural Research Service's Eastern Utilization Research and Development Division in Beltsville, Maryland. Representatives of the cooperating agencies established two criteria for the process: first, it had to remove substantial quantities of ⁹⁰Sr from fresh milk, and second, it had to do so without impairing the milk. Laboratory scale research was carried out by the Agricultural Research Service and the Public Health Service, and on the basis of these tests a pilot plant was constructed to test the practicability of the process before taking the final step upward to the construction of a commercial scale milk plant.

In September 1959, at the suggestion of the Public Health Service, a joint advisory committee for overall planning was established with membership from the three U.S. agencies and advice from the Canadian Department of Agriculture. This group acted as a steering committee guiding the direction of the research work.

COMMERCIAL SCALE FIXED-BED CATION REMOVAL

In September 1962 this committee recommended that the agencies develop a commercial scale plant for radionuclide removal from milk. This recommendation was based on the labora-

tory and pilot plant work to date⁽⁷⁻⁹⁾ which indicated success in the treatment of milk in the pilot plant without impairment of nutritional or organoleptic qualities.

In October 1962 the Public Health Service and the Department of Agriculture jointly agreed to finance investigations on the commercial feasibility of the pilot plant process. As a result of this agreement, a contract was let in June 1963, equally financed by both agencies, with the Producers Creamery Company of Springfield, Missouri for the construction, operation, and evaluation of a full-scale ion-exchange plant capable of treating 100,000 lb of milk per 8-hr day. It should be noted that the Atomic Energy Commission did not participate in this phase of the project, as the Commission believed it was not within its province to carry their support past the research into the practical application stage.

As an aside, I would like to point out that at this time, mid-1962, the ⁹⁰Sr levels in the U.S. milk supply were near the low end of the range considered by the Federal Radiation Council to be an acceptable intake for a lifetime. Although these levels were expected to remain low for the foreseeable future, from a long-range viewpoint it appeared, and still appears, that we should be in a position to safeguard such a vital food as milk in the event of any emergency. The possible seriousness of widespread contamination of the U.S. milk supply is indicated by the fact that over four million liters of fresh milk are consumed daily by infants less than two years old, the age group which is considered to be critical from the standpoint of radionuclide intake. This is based on a population of eight million in this age group with an average consumption of 500 ml of fresh milk per day.⁽¹⁰⁾ It should be noted that fresh milk consumption by individual infants may approach 1000 ml per day, but because a large number consume no fresh milk, the average intake is lower.

In the contract with Producers Creamery Company, the development of the mechanical design was left in the hands of the Company. The ultimate objective of this contract was to produce practical working drawings which could be duplicated by the milk industry in the construction of other large scale plants for removal of radionuclides from milk if necessary. The

contract included requirements for the performance of tests to determine the percent removal of ^{90}Sr and the effect of the treatment on the milk from the standpoint of sanitary, organoleptic, and nutritional quality.

Within a year after letting of this contract, the full-scale plant (shown in Fig. 3) had been completed and on June 24, 1964, the first full-scale treatment of 107,000 lb of fresh whole milk was successfully carried out in 12 hr and 10 min.

The environmental level of ^{90}Sr in the raw milk was 38.4 PCi/l. Samples of the treated milk taken at intervals indicated that the treatment process reduced the ^{90}Sr levels to between 1 and 3.2 PCi/l, a removal of greater than 90%. Subsequently, on September 16, 1964, and on February 25, 1965, full-scale milk treatment operations were carried out with similar results.

This series of tests successfully demonstrated the commercial feasibility of the process. The plant removed more than 90% of the environmental levels of ^{90}Sr with no increase in microbial population, a very slight decrease in flavor score, and only minor compositional changes.^(11, 12) The cost of processing was estimated to be 1.7 to 2.3 cents per quart of milk.⁽¹¹⁾

The first full-scale test (June 24, 1964) was observed by the joint advisory committee, with representatives of the Public Health Service, the Agricultural Research Service and the Canadian Department of Agriculture present. A meeting of this group immediately following the tests laid the groundwork for expanding this commercial process to include a unit for the removal of radioactive anions, particularly ^{131}I , from milk.

LABORATORY SCALE FIXED-BED ANION-CATION REMOVAL

In the early days of nuclear testing, radioiodine had been overlooked as a hazard in milk, because of the difficulty in measuring it and also because of the short half-life. As a result of weapons testing in 1961 and 1962 it had been observed that the quantities of ^{131}I in milk in the United States reached relatively high levels, as shown in Fig. 2. When these results were associated with the doses of radioiodine considered safe for lifetime intake, it

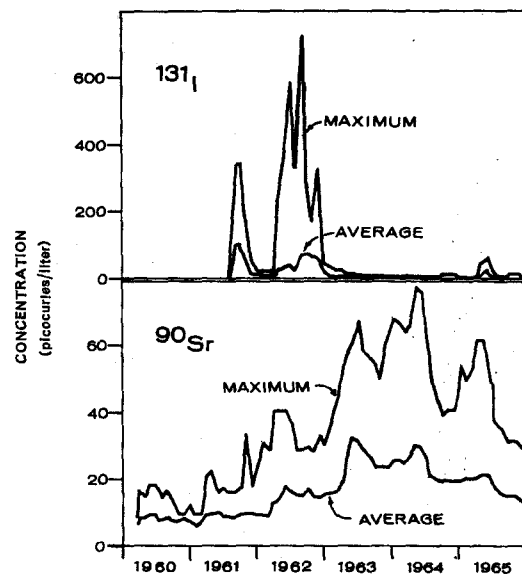


FIG. 2. ^{131}I and ^{90}Sr concentrations in milk. From U.S.P.H.S. pasteurized milk network.

became apparent that the dose of radioiodine to the susceptible population group might, in some geographic areas, exceed the radiation protection guide which had been established by the Federal Radiation Council for "normal peacetime operations". Thereafter, radioiodine was recognized to be a nuclide of major concern in milk. Public Health Service action included development of techniques for rapid measurement of radioiodine and investigation of ways to break the transmission chain (again referring to Fig. 1) between the contaminated feed and the milk-producing cattle. The Division of Radiological Health established a dairy farm near the nuclear test site in the State of Nevada to evaluate whether iodine levels in milk, following radioiodine release, could be kept low by various means, including substituting uncontaminated feed for pasturage on which local dairy cattle ordinarily grazed. As a result of these studies, whenever this technique has been used, the results were much as the earlier studies had predicted. It is therefore considered to be a practical method in localized areas for preventing radioiodine contamination of milk in a

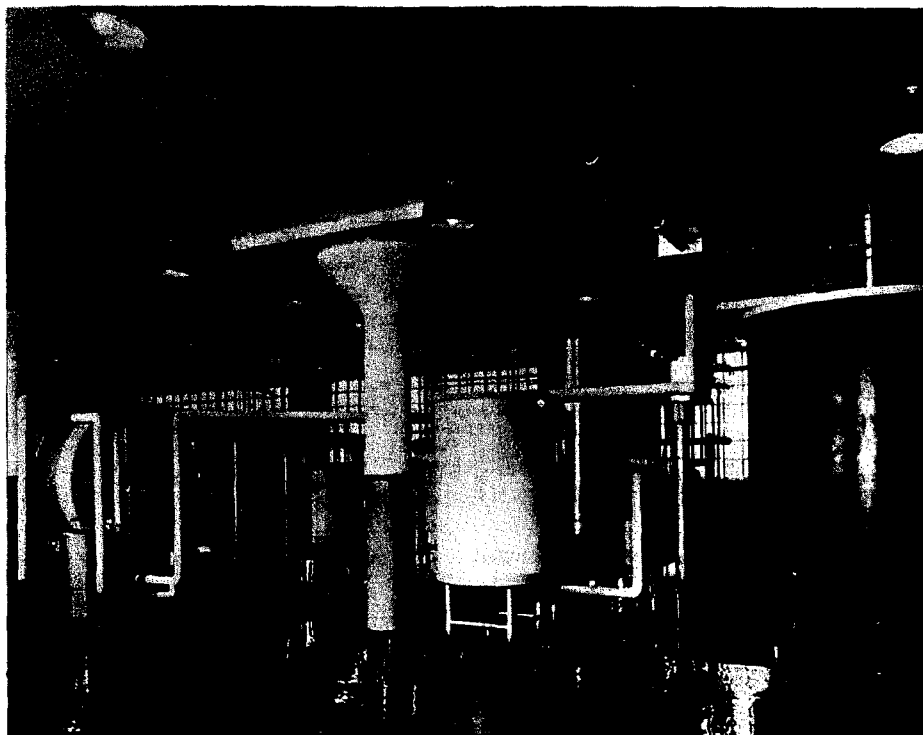


FIG. 3. Commercial scale fixed-bed plant for removing ^{90}Sr from fresh milk.

country such as the United States, where a rapid monitoring system for radioiodine exists.

Even with the development of this method for reducing transmission of radioiodine from atmosphere to man, the Public Health Service had encouraged laboratory scale studies to develop methods for removal of anionic radionuclides, principally ^{131}I , from milk by ion-exchange. Murthy *et al.* (^{13, 14}) had obtained 90% ^{131}I removal from fluid whole milk labeled *in vivo*, and removal of over 95% of ^{131}I from milk labeled *in vitro* by passage through Dowex 2-X8 resin. The flavor of the treated milk was comparable to that of untreated milk, and the anionic composition of the milk was not altered significantly.

By 1964, it was felt that a practical method for removing ^{131}I from milk could be developed on the basis of the laboratory work. Under sponsorship of the Public Health Service and the Department of Agriculture, a contract was let with the same commercial milk processor

to design and build a full-scale system to remove ^{131}I from milk which would be compatible with the cation removal process, and to incorporate it into the existing full-scale plant. The contract also included full-scale testing of the enlarged plant to confirm the feasibility of the combined strontium-iodine removal process. The progress and results of this endeavor are reported here in some detail, as the final tests were only recently completed.

Prior to design and construction of the full-scale anion removal unit, further laboratory tests were performed by the contractor. The laboratory-scale system consisted of an anion-exchange column of 200 ml Dowex 2-X8 resin followed by a cation-exchange column of 920 ml amberlite IR 120 resin. The flow diagram of this system is shown in Fig. 4. The treatment is similar to that previously described (^{8, 11}) except that treatment for anion removal precedes the cation removal process. Following passage through the anion column, milk was acidified

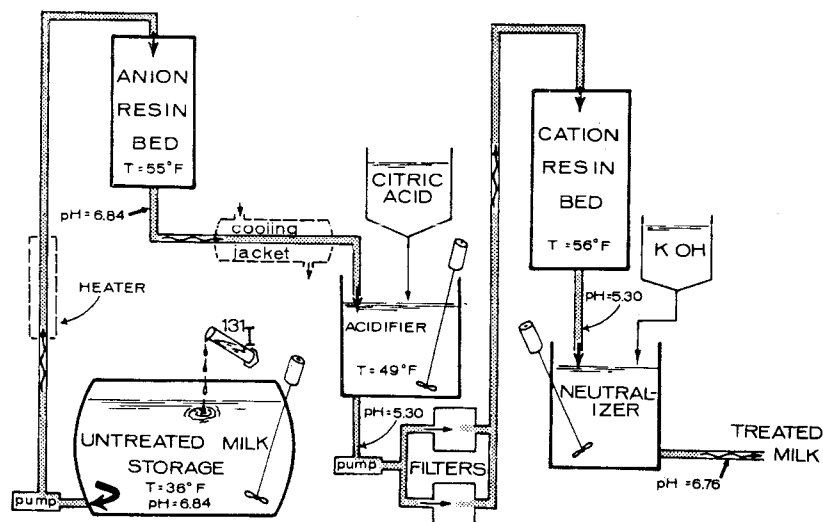


Fig. 4. Laboratory scale cation-anion bed system, with typical pH and temperature.

to pH 5.3 with 20% citric acid solution and filtered before passage through the cation removal column. The pH was then adjusted to 6.8 with 2M KOH (about 15 ml/l. of milk).

Anion removal preceded cation removal because the regenerant salts for the anion resin bed had been proportioned on the basis of the ionic content of normal raw milk. Since cation treatment increases the potassium and citrate content in the milk, treatment for anion removal subsequent to cation removal would have required further laboratory development on regenerant salt proportioning and delayed the program. It may be practical to have cation treatment precede anion treatment.

Four trial runs were made with this system. In each run, 35 l. of grade A whole raw milk were processed at a flow rate equivalent to 16,200 lb/hr in the full-scale cation-removal plant.

The milk was labeled with from 4000 to 48,000 PCi/l. of ^{131}I , as the environmental level was too low to be readily detectable. A small quantity of sodium sulfite ($50\text{ }\mu\text{g}$) was added to the ^{131}I tracer solution to prevent iodide oxidation. The environmental level of ^{90}Sr in the milk was approximately 30 PCi/l.

The laboratory-scale plant performed satisfactorily. In the final trial run, ^{131}I was reduced

97.1% (from 3970 to 115 PCi/l.), and ^{90}Sr was reduced 84.5% (from 30.7 to 4.7 PCi/l.). ^{90}Sr removal was less than expected on the basis of past experience. However, because of the limited accuracy in ^{90}Sr analysis at such low levels and the demonstrated ability of the full-scale plant to remove over 90% of ^{90}Sr , it was decided that further laboratory testing of cation removal was not necessary.

The organoleptic quality of the processed milk (which had been pasteurized) was compared with that of the raw milk and also with pasteurized homogenized milk purchased locally. The processed milk showed a slight but insignificant change in flavor.

Chemical analyses of the raw and processed milk showed no significant changes due to processing other than the expected increase in the citrate and potassium from the acidification and neutralization processes.

Of some concern was the formation of a visible precipitate in samples of processed milk after standing overnight. Solubility index determinations were made on various fractions of processed milk from one trial in an effort to determine the seriousness of the problem. On the basis of these tests, it was concluded that this was due to inadequate mixing of the acid and neutralizer with the milk, and that rapid mixing

of both citric acid and potassium hydroxide is necessary to insure stability of the milk.

From the results of the laboratory tests, it was concluded that, assuming good control of acid-base mixing, the commercial scale combined process would produce a commercially acceptable homogenized milk. Design of the anion resin unit therefore proceeded on the basis of the laboratory findings.

COMMERCIAL SCALE FIXED-BED ANION-CATION REMOVAL

The full-scale ^{90}Sr removal plant has been described previously.^(11, 12) The flow diagram in Fig. 5 shows where the anion resin column was inserted in the system. The anion column (Fig. 6) is a mild carbon steel cylinder 26 in. in diameter and 100 in. long with an inner lining

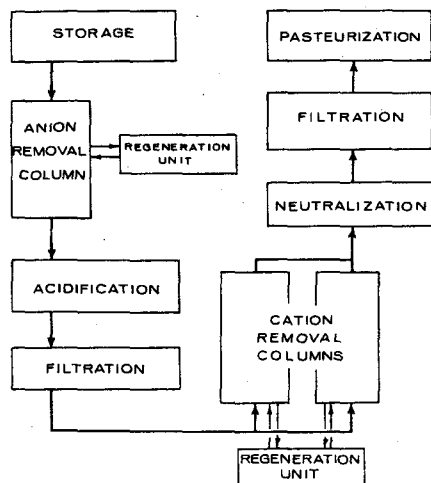


FIG. 5. Full-scale system for removal of radio-nuclides from milk.

of $\frac{1}{4}$ in. thick, high density polyethylene. The cylinder contains 284 l. of Dowex 2-X8 resin. The resin is supported on a sixty-mesh woven nylon screen resting on a 5 in. bed of graded glass beads $\frac{1}{2}$ to 1 in. in diameter. As in the remainder of the plant, all materials, chemicals and resins used in the anion unit in contact with milk had been previously approved by the U.S. Food and Drug Administration for use in food processing.

Three full-scale plant tests of the system were made in May and June 1966, with 100,000 lb of fresh milk processed in each test.⁽¹³⁾ In the first test (May 18, 1966) the milk was labeled *in vitro* with 1880 PCi/l. of ^{131}I . Processing reduced this to 87 PCi/l., a removal of 95.4%. Radiation levels outside the anion column reached 0.1 mr/hr on the surface of the column at a point opposite the surface of the resin. The treated milk lost some flavor; however, this was noticed principally in the first 15,000 lb of milk, and thus the flavor loss was related to the start up of the process.

The second test run was made June 8, 1966. The milk was labeled *in vitro* with 2000 PCi/l. of ^{131}I and 3000 PCi/l. of ^{85}Sr , as environmental levels of both ^{131}I and ^{90}Sr in the raw milk were too low for accurate determination of the efficiency of radionuclide removal. The milk was processed at the rate of 19,300 lb/hr, approximately 50% above the design flow rate of the cation column and 20% above the design flow rate of the anion column.

The final test run was made June 22, 1966. Milk was labeled *in vitro* with 4760 PCi/l. of ^{131}I and 4225 PCi/l. of ^{85}Sr , and processed at approximately 20,000 lb/hr. The results of the last two test runs are shown in Tables 1 through 4.

As shown in Table 1, the process removed over 99% of the ^{131}I and over 91% of the ^{85}Sr . The strontium removal efficiency decreased toward the end of each run, falling below 90% removal after treatment of 25 resin bed-volumes (rbv) of milk at a flow rate of 0.11 rbv/min. This decrease is thought to be due to the high flow rate as reported by Walter.⁽¹⁴⁾ It may be necessary to use a larger amount of cation resin to maintain satisfactory strontium removal at high flow rates.

Analysis of the milk processed on June 8, 1966, by gamma spectroscopy also showed that treatment reduced the environmental (*in vivo*) levels of ^{140}Ba by 88% and ^{137}Cs by 71%.

Chemical analyses of the milk processed on June 8, 1966, are given in Table 2. The increases in citrate and potassium are due to chemicals added in the acidification and neutralization processes. The differences in the other chemical ions in the raw and processed milk are inherent in the process, as the quantities of regenerant

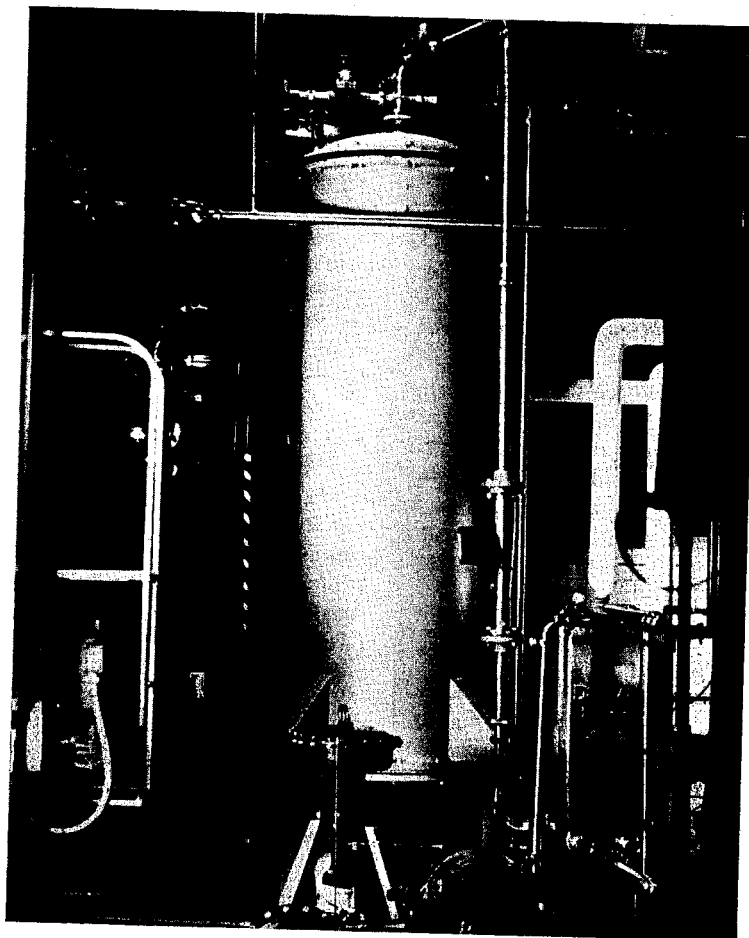


FIG. 6. Commercial scale resin bed for removing ^{131}I from milk.

salts are based on an "average" milk, which may differ from the milk processed on any single day. These differences between the raw and processed milk are not considered significant.

The dilution of the milk due to processing was calculated on the basis of the change in total solids and fat content of the milk during processing. These values are shown in Table 2. Approximately 2.5 to 3% of this is due to water added in the acidification and neutralization processes. The remainder, particularly the excessive dilution in the first 3000 lb of milk processed, is due to the fact that the system is full of water when processing begins and there is considerable mixing at the water-milk interface. As a result, the first portion of milk processed is quite heavily diluted. Chemical

analyses of the milk treated on June 22, 1966, were comparable to those of the milk treated on June 8, 1966.

Flavor tests were made on the raw and processed milk to determine the effects of treatment on the organoleptic quality of the milk. Results are shown in Table 3. The first 1000 lb of milk treated on June 8, 1966, was not acceptable in flavor, and contained a taste described as "astringent". This taste, associated with starting plant operation, had been observed before and is tentatively attributed to the anion resin regeneration and storage procedure. Laboratory tests have shown that storage of the anion resin in 2 M HCl deteriorates the resin. Storage at a higher pH (2.0) is indicated to minimize resin deterioration.

Table 1. Removal of ¹³¹I and ⁸⁸Sr from Fresh Milk

Pounds of milk processed	¹³¹ I				⁸⁸ Sr			
	June 8, 1966		June 22, 1966		June 8, 1966		June 22, 1966	
	pCi/l.	% removed	pCi/l.	% removed	pCi/l.	% removed	pCi/l.	% removed
Tank No. 1* 0	2220	—	4760	—	3030	—	4225	—
Tank No. 2* 0	2070	—	—	—	2680	—	—	—
15,000	34	98.5	43	99.1	20	99.3	378	99.2
30,000	21	99.0	38	99.2	25	99.2	356	91.6
45,000	28	98.7	56	98.8	95	96.9	337	92.0
60,000	32	98.5	52	98.9	108	96.0	298	93.0
75,000	30	98.6	59	98.8	269	90.0	412	90.3
90,000	40	98.0	85	98.2	414	84.6	568	86.6
Composite sample of 100,000 lb of processed milk	15	99.3	43	99.1	70	97.5	357	91.6

* The first 50,000 lb of milk treated was from Tank No. 1 and the remainder was from Tank No. 2.

Table 2. Composition of Milk Processed June 8, 1966

Pounds of milk processed	Solids %	Fats %	Ash %	Na ppm	K ppm	Ca ppm	Mg ppm	Citrate ppm	Chloride ppm	Phos. ppm	Sol. Index
(Raw)											
0	12.43	3.59	0.73	457	1725	1217	112	1840	984	653	0.04
3000	10.84	3.04	0.73	488	2488	994	241	4000	465	533	—
9000	12.02	3.33	0.81	431	2625	1108	245	4140	666	586	—
15,000	12.12	3.41	0.78	401	2275	1157	263	3400	858	583	—
45,000	12.18	3.38	0.84	451	3075	1117	147	3600	954	595	0.04
60,000	12.25	3.40	0.89	487	3200	1163	99	3840	954	600	—
75,000	12.21	3.40	0.87	438	2825	1124	134	3520	968	598	—
90,000	12.22	3.41	0.86	443	2850	1185	119	3320	982	595	0.04
(Composite)	12.06	3.37	0.83	490	2725	1155	145	3640	908	550	0.05

% Dilution of milk from processing calculated from change in	Solids %	Fats %
First 3000 lb	17.8	18.1
Next 6000 lb	6.3	7.8
Next 80,000 lb (Composite)	4.3	5.6
100,000 lb	5.5	6.5

Table 3. Evaluation of Flavor in Processed Milk

Pounds of milk processed	Flavor score*					
	Raw milk		Effluent from anion column		Fully processed pasteurized milk	
	June 8	June 22	June 8	June 22	June 8	June 22
0	37.4	soured	—	—	—	—
1,000			—	37.2	33.6	35.3
9,000			36.1	36.7	—	36.8
15,000			—	36.6	35.3	36.7
22,000			36.1	—	—	—
26,000			—	—	35.6	—
30,000			—	36.8	36.2	35.8
38,000			36.4	—	—	—
60,000			—	36.3	36.0	36.3
61,000			36.4	—	—	—
90,000			—	36.7	36.2	36.4
95,000			36.3	—	—	—
Composite of 100,000			—	—	35.9	36.3

* American Dairy Science Association standard flavor test; perfect score is 40; acceptable score for pasteurized commercial market milk is 35.

The flavor score of the last 70,000 lb of processed milk was higher than that of the composite sample, indicating that the initial volume of unacceptable milk affected the quality of the entire 100,000 lb of milk.

To minimize the taste problem, the anion resin was stored at pH 2.0 between June 8 and June 22, 1966. This procedure, plus discarding the first 1000 lb of milk treated on June 22, 1966, was believed to be responsible for the improved flavor scores in the milk treated on June 22 (see Table 3).

The results of microbiological examination of the milk processed on May 18 and June 8, 1966, are shown in Table 4. The high coliform counts are attributed to contamination after pasteurization due to immediate storage of the finished milk in the raw milk tank which had not been adequately cleaned. The microbiological examination indicates no significant defects in the processing system itself from the sanitation viewpoint. Bacteriological examination of the milk processed on June 22, 1966, confirm these results.

The use of the woven mesh screen and the glass bead bed in the anion column has caused some concern from the sanitation viewpoint, but no bacteriological growth in this space could be detected from the test results.

In summary, the full-scale plant effectively reduced the levels of ^{131}I and ^{90}Sr in fresh fluid milk without any serious changes in organoleptic, chemical or microbial quality of the milk, and without significant operational problems.

The cost of chemicals for regenerating the anion unit in the full-scale tests was approximately 1.5 cents per quart of milk (¢/qt). Cost of plant operation for strontium removal was 1.7 to 2.3 ¢/qt, including approximately 0.6 ¢/qt for chemicals.⁽¹¹⁾ Since the addition of the anion column did not increase labor or other costs appreciably, it appears that the combined process adds approximately 3.2 to 3.8 ¢/qt to the cost of milk.

The use of processes involving additives to food shipped interstate commerce requires approval by the U.S. Food and Drug Administration. Therefore, extensive testing of the

Table 4. Microbiological Analysis of Raw and Processed Milk

Sample	Micro-organisms per ml			
	Std. plate count	Psychrophilic	Coliform	Staph.
<i>Test of May 18, 1966</i>				
Raw	137,000	51,000	—	—
Processed and pasteurized	3 500	—	980	0
<i>Test of June 8, 1966</i>				
Raw	9,100,000	10,900,000	—	—
Processed and pasteurized	15,800	—	2260	0
Public Health Service Recommended Standards*				
Raw	100,000	—	—	—
Pasteurized	20,000	—	10	—

* Grade A Pasteurized Milk Ordinance—1965 recommendations of the United States Public Health Service, PHS Publication No. 229, pp. 39-40, Washington, D.C. (1965).

wholesomeness and nutritional and toxicological quality of the treated milk was necessary to provide the FDA with sufficient data for evaluation of the process as a basis for approval.

In addition to the tests reported above, further chemical and vitamin analysis is being done on the raw and processed milk. Chemical determinations include analysis for crude protein, acidity, ash, iron, copper, iodine, silicone, cobalt, zinc, aluminum, manganese, and molybdenum. Vitamin assays include determination of thiamine (bound and unbound), riboflavin, vitamins A, B₆ and B₁₂, folic acid, ascorbic acid, pantothenic acid, niacin, biotin, and a wide spectrum of fatty acids. Biological and chemical analysis of protein quality will also be carried out.

To evaluate the nutritional quality of the processed milk, baby rats will be maintained 10-12 weeks in seven groups which will receive diets prepared with dried processed or raw (control) milk. Baby pigs (one day old and ten days old) will be maintained until 8 weeks of age on diets prepared from dried processed milk. Feeding studies may also be done with other species of animals.

To provide the materials for this test program, 3000 lb each of the processed and raw milk from the June 8, 1966, test run have been dried and sealed in containers in a nitrogen atmosphere. The testing will be conducted at the PHS Radiological Research Laboratory in Rockville, Maryland.

LABORATORY SCALE MOVING-BED ANION-CATION REMOVAL

The Department of Agriculture and the Public Health Service have been encouraged to conduct research and development for practical methods for removing radionuclides from milk using processes developed in the scientific and industrial community at large, as well as those developed in their own laboratories. One of these which has shown promise is the Higgins continuous ion-exchange contactor.* In this process (see Fig. 7) an ion-exchange resin bed moves counter-current to the flow of milk

* Manufactured by Chemical Separations Corporation, Oak Ridge, Tennessee.

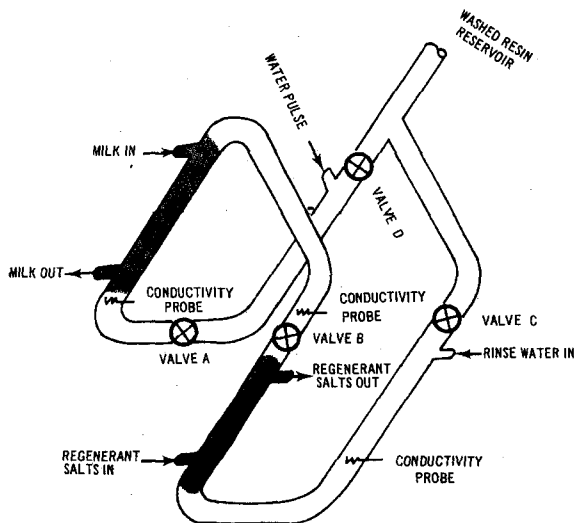


FIG. 7. Flow diagram of moving bed ion-exchange system.

through a closed system, and is regenerated in another section of the system.

Edmondson,⁽¹⁷⁾ using an early model of the system, obtained 90% removal of ^{85}Sr from milk acidified to pH 5.3 and over 90% removal of ^{134}Cs . Edmondson also treated milk for concurrent cation-anion removal by passing acidified milk through a fixed cation resin bed and then through an anion resin in the contactor. The anion resin was charged for simultaneous ^{131}I removal and neutralization of the milk. Removal of 86 to 91% of the ^{131}I was achieved.

Although problems were encountered in these investigations with pH control, milk flavor, and regeneration of the anion resin, the results obtained were sufficiently encouraging to warrant further study of the process.

PILOT SCALE MOVING-BED ANION-CATION REMOVAL

In 1964, the Public Health Service and the Department of Agriculture jointly sponsored construction of a moving bed ion-exchange milk treatment plant, based on the Higgins patent.⁽¹⁸⁾ The public's rights to patents resulting from work sponsored by these agencies have been protected under the provisions of the Atomic Energy Act.

Since the process had not been used to treat milk on a large scale, extensive laboratory work was necessary before the plant was designed. Investigations by the Public Health Service⁽¹⁹⁾ formed the basis for the design of the pilot plant. These studies established flow rate, the depth/diameter ratio of the ion-exchange unit necessary for adequate treatment, and identified the need for continual cooling of the milk and adjacent sections of the loop during processing to prevent microbial growth.

The plant (shown in Fig. 8) has a capacity of 7000 lb of milk per 8 hr and consists of two continuous ion-exchange contactors, one for removal of radioactive cations and one for removal of radioactive anions. The resins are the same type used in the full-scale fixed-bed plant. The plant is self-contained (except for power supply) and is designed for mounting on a

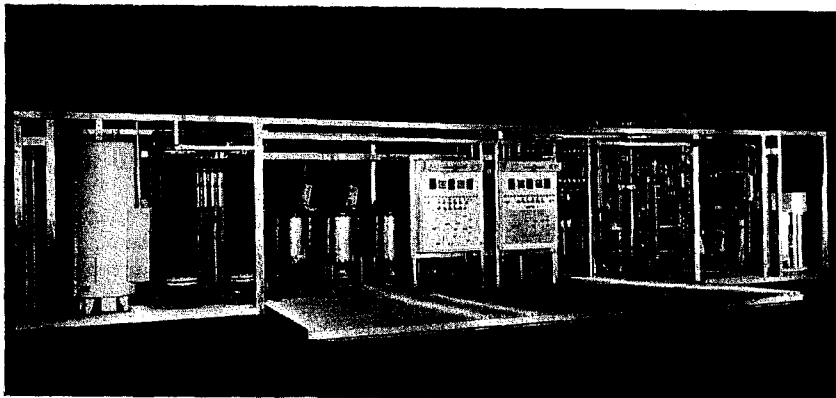


FIG. 8. Pilot plant moving bed ion-exchange system.

standard 40 ft \times 8 ft truck trailer to make it readily transportable.

This process is still in the experimental stage, and has a large degree of flexibility designed into it, particularly in the operation of the anion bed. The anion removal portion was originally conceived to be a moving resin bed. However, system testing will be done initially with a small anion fixed-bed column, since the high efficiency and large capacity of the anion resin indicates this to be the most simple and adequate operating design for anion removal. Dual anion columns are provided to achieve continuous operation.

The pilot plant is scheduled to be operated at the Public Health Service laboratory in Montgomery, Alabama to evaluate its effectiveness. The plant testing program will include evaluation for efficiency of radionuclide removal, product acceptability (in terms of taste, odor and appearance), product alteration, and sanitary quality. As the operation becomes refined, nutritional testing and/or chemical analyses of the treated milk are planned, using methods similar to those used in evaluating the product from the full-scale fixed-bed plant.

SUMMARY

Although the environmental levels of ^{90}Sr and ^{131}I and other radionuclides in milk are presently well within acceptable limits,⁽²⁰⁾ it is prudent to have a system capable of treating milk for the removal of these radionuclides in the event of gross environmental contamination to a milk producing area.

In the case of acute contamination of milk with radioiodine, action must be taken promptly to effectively reduce the radiation dose to humans. If a week elapses before controls are initiated, more than half of the human dose will already have been received. With this in mind, the moving-bed unit has been designed and constructed for placement on a standard truck trailer, to determine whether such a mobile unit would be practical. If so, it is estimated that the strategic location of six to twelve full-scale standby mobile treatment units would make it possible to place a unit in operation in any milk shed in the United States within 24 hr should an emergency arise. Because even the large-scale units are limited in

capacity, they would be adequate only in the event of radiological contamination covering a limited area. Obviously, an effective surveillance system is required in order to know when to initiate and terminate these control actions.

A major use of the two existing plants in the near future is in the development and refinement of lower-cost treatment techniques or treatment of milk massively contaminated. They may also be used for training and for demonstrating the processes.

It is emphasized, however, that the objective in developing these processing plants is to provide the capability to treat fresh whole milk for immediate human consumption. To this end, formal approval of the U.S. Food and Drug Administration has been requested to use the full-scale combined anion-cation fixed-bed unit for this purpose on an emergency basis, and extensive testing has been undertaken to insure the wholesomeness of the product. Similar approval will be sought for the moving-bed plant at the appropriate stage in its development.

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