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The Effect of Fruit Acids on Aluminum

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THE EFFECT OF FRUIT ACIDS ON ALUMINUM

By

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PREFACE

Aluminum has been used in cooking utensils for about thirty years and is still the favorite material employed. There is no disagreeable taste, odor or discoloration discernible from its use.

There have been many experiments to prove that aluminum does not depreciate the quality of the food cooked in it. It does, however, add itself to the acidic and basic foods cooked therein. The amount taken up by neutral foods is negligible

For experimental purposes, fruit juices were chosen as the attacking substances and the strips of aluminum were of the quality used in ordinary cooking utensils, not cast aluminum.

The writer wishes to thank Dr. John R. Koch for his assistance in directing the work and to acknowledge the authorities quoted.

INTRODUCTION

CHAPTER 1.

Aluminum utensils discolor in some cases and brighten in other cases when certain foods are cooked and allowed to stand therein. Why is this so and what is the amount of aluminum that a piece will gain or lose when heated and allowed to stand in contact with food juices?

This, our problem, has been confined to fruit juices and sauces. Most vegetables are usually cooked in water, which would thus be added as a complicating factor, since water itself exerts its own influence on aluminum.

This problem was brought to the writer's attention when tomatoes which had been cooked and allowed to stand in an aluminum kettle for twenty-four to forty-eight hours caused the kettle to become punctured with little holes, some as large as the head of a pin.

Our difficulty could have been solved in two ways; one, by analyzing the food, and, secondly, by weighing the loss in weight of the strip of metal.

Recently Beal, Unangst, Wigman, and Cox (1) of the Mellon Institute of Industrial Research, have conducted an

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(1) George D. Beal, Richard B. Unangst, Helen B. Wigman, and Gerald J. Cox, "Aluminum Content of Foodstuffs Cooked in Aluminum", Industrial and Engineering Chemistry, Vol. 24, No. 4, April 1932, p. 405.

experiment, using the first method, to study the amount of aluminum which enters the food by contact with aluminum. Attention has been directed to the food rather than to the utensils. The same foods were cooked by the same recipe in glass and in aluminum. The difference in the amount of aluminum present in the aluminum vessel and the glass is then taken as the amount which was introduced by the aluminum utensil. Table I. shows a portion of their results dealing with fruits.

TABLE I

Aluminum Content of Foodstuffs

Cooked in Glass and in Aluminum

Food	Duration of Cooking Minutes	Cooked in Glass P.p.m.	Cooked in Aluminum P.p.m.	Average Increase in Aluminum P.p.m.	Remarks
Stewed Tomatoes	20	.12	4.28	4.16	Bright Pan
Stewed Tomatoes	20	.12	15.42	15.3	Dark Pan
Rhubarb	5	.95	13.4	12.5	Bright Pan
Rhubarb	5	.94	41.8	40.9	Dark Pan
Apricots	40	24.6	73.3	48.7	
Apple Sauce	10	.28	1.4	1.12	
Apple Butter	390	5.28	118.0	113.0	Includes Time to Concentrate Cider.
Orange Marmalade	90	.30	3.06	2.76	
Cranberry Sauce	10	.54	7.9	7.36	Bright Pan
Cranberry Sauce	10	.52	28.0	27.5	Dark Pan

These results, however, have not allowed for the water used. The same tap water was used all through the experiment.

This amount of aluminum is found to be far below 1400 P.p.m. which is necessary to produce symptoms of phosphorus starvation in animals on a low phosphorous diet.

Lung and Schmid (2) and Mrak and Cruess (3) exposed aluminum strips to various food acids and obtained small and varied amounts of corrosion.

Other experiments have been done which concern pitting, polishing, discoloration, precipitates formed, and alleged changes of taste.

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(2) G. Lung and E. Schmid, Z. Angew. Chem. 5,7 (1892)

(3) E. Mrak and W.V. Cruess, Food Ind. 1, 559 (1929)

CHAPTER II

Manufacturers of aluminum cooking utensils admit that a discoloration does appear and add immediately, "but this is not harmful".

Investigations of this sort resolve themselves into two parts; the study of acids and the study of bases.

The Aluminum Wares Association in a little bulletin "Aluminum and Aluminum Wares" issued by them tell of a study made by Alberton S. Cushman, the director of the Institute of Industrial Research, Washington D.C., states that a one-half percent acetic acid solution in distilled water after one hour's boiling attacked the aluminum very little. If an equal percentage of common table salt is added to this the attack is greater. The same results occurred if this same solution of vinegar and salt was allowed to stand in a vessel cold for two days. To this experimentor there was no apparent attack on the metal surface. The pamphlet hastens to add that this was a much greater acid strength than is used in most cooking operations.

Whittaker (4) in speaking of the corrosive effect of acetic acid on aluminum says, "Concentrations up to one percent corrode aluminum with the formation of adherent protective coatings. At ordinary temperatures (20° C)

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(4) H.F. Whittaker, Research Information Surveys on Corrosion of Metals, No. 2 Corrosion of Aluminum, p. 2.

the concentrations of acetic acid do not seriously corrode aluminum, although the corrosion at about one percent concentration is about three times as great as at five percent. As the concentration rises, the corrosion rate gradually diminishes to zero at about ninety-nine percent. Boiling one percent acetic acid attacks the metal appreciably, the corrosion rate being about four times that of the cold acid".

Seligman and Williams (4) have found that aluminum is vigorously attacked by boiling acetic acid after the last traces of water have been removed from the acid.

Calcott and Whetzel (4), however, have made tests with one hundred percent acetic acid and found the corrosion rate very low. They did find, however, that there was a serious attack with mixtures of glacial acetic acid and acetic anbydride, particularly when the mixture contained mostly the glacial acid.

Whittaker (4) claims that the affect of commercial acetic acid upon aluminum is caused by the presence of small amounts of formic acid.

Anderson (5) says that aluminum is attacked slowly by cold acetic acid but that the rate of attack

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(5) R.J. Anderson, The Metallurgy of Aluminum and Aluminum Alloys, p. 139.

increases with increasing temperature and with increasing dilution of the acid.

The following table taken from Anderson shows the rate of solution of aluminum of different purity in boiling acetic acid.

Rate of Solution of Aluminum
in Boiling Acetic Acid. (5)

Concentration of the acid percent	Solution rate, mgs. Aluminum dissolved per 24 hours, per sq. cm. exposed.		
	Sample 1. 99.73% Al.	Sample 2. 99.6% Al.	Sample 3. 99.1% Al.
50	315	340	415
60	285	340	405
70	240	280	330
80	210	220	295
90	100	120	165
98	33	28	35
99.9	3	3	7

According to Whittaker (6) the corrosion rate of Boric Acid on aluminum is very low.

Butyric acid, a constituent of butter, has about the same effect on aluminum as acetic acid with the boiling acid. However, in the cold the attack is very slight.

Whittaker (7) says that citric acid does not affect aluminum and E.E. Smith (8) confirms this but says

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(6.) H.F. Whittaker, op. cit., p. 4.
(7.) Ibid., p. 5.
(8.) E.E. Smith, Aluminum Compounds in Foods, p. 29.

that in actual cooking experiments by Dr. John Glaister and Dr. Andrew Allison of Glasgow University, small amounts of dissolved aluminum were found in marmalade made from oranges and lemons. In two and one-half pounds of marmalade, they found 1.018 grams of aluminum hydroxide. This was the largest amount found in their experiments and they concluded that the ordinary use of aluminum cooking utensils for culinary purposes is not attended with any risk to the health of the consumers of food cooked therein.

Both Smith (8) and Whittaker (9) in speaking of the effect of Lactic Acid on aluminum quote Utz as the authority. He observed that Lactic Acid up to one percent concentrations at room temperature has no affect on aluminum, but that at higher temperatures it does dissolve a small amount of aluminum which is harmless physiologically; and he concluded that aluminum vessels were suitable for milk products.

Trillat (9) and Droully reached about the same conclusions.

Anderson (10) says, "Lactic acid attacks aluminum very slowly and both aluminum and certain of its alloys are suitable for milk cans and containers for buttermilk".

Nitric, Sulphuric, Oxalic and Hydrochloric acid all attack aluminum appreciably. The Aluminum Wares Association

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(9) H.F. Whittaker, op. cit., p. 7.
(10) R.J. Anderson, The Metallurgy of Aluminum and Aluminum Alloys, p. 139.

warns against the use of oxalic acid which is contained in some cleaning solutions.

Phenol, or carbolic acid, has no action on aluminum so long as it is in aqueous solution but anhydrous samples attack the metal vigorously. (11)

Oleic acid has no effect at all on aluminum nor have soaps or fats.

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(11) H.F. Whittaker, op. cit., p. 10.

CHAPTER III

Alkalis used in kitchens are more harmful to aluminum than the acids used. All caustic alkalis attack aluminum vigorously.

R.J. Anderson (12) speaking of the action of alkalis on aluminum says, "Ammonium hydroxide attacks aluminum slowly, forming aluminum hydroxide; on first exposure of the metal the attack proceeds at once, but a protective coating is formed that prevents further action. Potassium and sodium hydroxides attack aluminum rapidly, giving hydrogen and aluminum hydroxide, which passes into solution as an alkali aluminate. While aluminum is not appreciably affected by ordinary waters, it is attacked by alkaline waters, and by water to which alkalies or soaps have been added. The blackening and corrosion of aluminum kitchen cooking utensils is often traced to alkalies which come in contact with them".

And Evans (13) says that alkaline liquids have a distinct action on aluminum, since the oxide is soluble in alkalis. Also that it is well known that aluminum vessels must not be cleaned with ordinary washing soda; and special cleaning preparations manufactured for

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(12) R.J. Anderson, The Metallurgy of Aluminum and Aluminum Alloys. p. 141.
(13) U.R. Evans, The Corrosion of Metals, p. 111.

use with that metal mostly contain sodium silicate.

Again Allerton S. Cushman (14) found that on boiling ordinary cooking soda for one hour in an aluminum vessel, the attack was four times that produced by the acid solution.

Whittaker (15) says that sodium carbonate in aqueous solution is corrosive to aluminum.

E.E. Smith (16), quoting from "Lancet", says that carbonate of soda certainly attacks aluminum freely and that it would be well to exclude it from an aluminum cooking utensil.

According to the Aluminum Wares Association (14), it is the alkali present in the water supplies that causes the discoloration of aluminum cooking utensils.

Likewise, Evans (17) says that many ordinary tap waters are sufficiently alkaline to cause a dark stain on commercial cooking vessels. This stain, he continues, is connected with the presence of iron in the material and does not occur with acidic waters, which would dissolve the iron as well as the aluminum. Protective films tend to fail most easily in the presence of chlorides.

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(14) Aluminum and Aluminum Ware, p. 18-19
(15) H.F. Whittaker p. 12
(16) E.E. Smith, Aluminum Compounds in Foods, p. 27
(17) U.R. Evans, The Corrosion of Metals, p. 111

Seliganan and Williams (18) did numerous experiments dealing with the action of hard industrial waters on aluminum. They feel convinced that ordinary "tap water" invariably attacks aluminum "unless special means be taken to prevent it".

They placed a strip of hard rolled aluminum sheet in ordinary tap waters and observed what took place. The first visible sign of attack was the appearance of gas bubbles on the surface of the metal. The bubbles on examination seemed to be encased in tenuous clouds of $Al(OH)_3$ which if left undisturbed adhered lightly to the metals for a considerable time. In the tap waters used by the experimentors these gas bubbles appeared in fifteen minutes after the strip was immersed. This form of corrosion is purely superficial and was termed "etching". When the strip was washed and dried, its surface was seen to be mottled where the gas bubbles were but no deep seated corrosion was apparent.

As the experiment continued, it was found that the etching went on for about twenty-four hours with more and more of the surface becoming involved. Then this type slowly ceased. The unattached portions showed light brown

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(18) R. Seligman and P. Williams, "The Action on Aluminum of Hard Industrial Waters", Engineering, Vol. 109 pp. 362-364. March 12, 1920.

stains and were rough to the touch owing to a crystalline deposit of calcium carbonate. After a day or two "pitting" began to show.

They described this form of corrosion as intensely local and deep seated, characterized by the growth at certain spots on the surface of the metal of white gelatinous tufts or nodules each of which is associated with one or more relatively large bubbles of hydrogen. Pitting showed no tendency to diminish but proceeded as long as the experiment continued. After a strip had been immersed one week each tuft or nodule was found to be connected with a pit, group of pits, or a blister on the surface of metal. Each pit or center of attack was surrounded by a zone of unattacked metal which retained its original lustre.

They have come to the conclusion that the pitting of Aluminum by ordinary water is dependent upon the simultaneous presence of chloride and bicarbonates; the pitting seems then to arise at places where half-closed-up cavities are present in the metal, a state of affairs clearly favorable to the production of non-aerated (anodic) points.

Etching is superficial, ceases after a time, and is of little consequence from a practical viewpoint. Pitting may penetrate deeply and continue indefinitely and, therefore, presents a difficulty to industry.

CHAPTER IV

Experimental Results

The method used in this experiment was the latter one. Strips of aluminum were cleaned, rubbed with emery paper, washed, dried, and placed in different juices, dilute acids and bases, heated to boiling, and then allowed to stand for two weeks except for the time necessary for weighing each day.

We have sought to measure the amount of aluminum that a strip of the metal will lose, when placed in different fruit juices, dilute acids and alkalis for varying lenghts of time, over a period of two weeks.

We experimented with this method on tomatoes first to determine whether or not it was a feasable way of detecting the loss of aluminum since it was certain that tomatoes had caused the perforations that were first noticed in an aluminum kettle.

The juices used were strawberry, logenberry, peach, apricot, cranberry, pineapple, orange, lemon, grapefruit, sauces from apples, cranberries rhubarb and tomatoes; also dilute solutions of acetic, tartaric, citric, hydrochloric acids, sodium carbonate and sodium chloride which were used for comparative results.

A weighed sample was placed in the liquid and heated to boiling. Then the strips were let stand over night since samples showed no loss in weight after three or four hours. Table II shows the loss in aluminum of a strip exposed to the designated juices for varying lenghts of time. Table III is the loss in weight of aluminum strips after standing in contact with weak acids and alkalis.

TABLE II

Loss in Weight of Aluminum Strips

Exposed to Fruit Juices

Foods	Sample of Aluminum Grams	Amount Lost by Sample Grams	Time of Contact Days	Area of Sample Square Cm.
Stewed Tomatoes	5.4196	.0021	2/3	66
Tomatoes (Soup)	5.3060	.0077	7	25
Strawberry (Juice)	2.0103	.0000	1	25
	2.0103	.0003	7	25
	2.0103	.0102	14	25
Loganberry (Juice)	2.0923	.0000	1	25
	2.0923	.0043	7	25
	2.0923	.0081	14	25
Rhubarb	2.1244	.0000	1	25
	2.1244	.0082	7	25
	2.1244	.0186	14	25
Peach (Juice)	2.0672	.0002	1	25
	2.0672	.0016	7	25
	2.0672	.0042	14	25
Apricot (Juice)	2.0612	.0019	1	25
	2.0612	.0025	7	25
	2.0612	.0028	14	25
Pineapple (Juice)	2.0784	.0018	1	25
	2.0784	.0150	7	25
	2.0784	.0170	14	25

Table II (Continued)

Foods	Sample of Aluminum Grams	Amount Lost by Sample Grams	Time of Contact Days	Area of Sample Square Cm.
Orange (Juice)	5.4914	.0000	1	66
	5.4914	.0044	7	66
	5.4914	.0044	14	66
Cranberry (Sauce)	3.3601	.0031	1	42
	3.3601	.0085	7	42
	3.3601	.0105	14	42
Lemon (Juice)	2.0652	.0012	1	25
	2.0652	.0032	7	25
	2.0652	.0052	14	25
Grapefruit (Juice)	2.0630	.0010	1	25
	2.0630	.0030	7	25
	2.0630	.0048	14	25
Apple (Sauce)	2.0912	.0012	1	25
	2.0912	.0030	7	25
	2.0912	.0040	14	25

TABLE III

Loss in Weight of Aluminum Strips Exposed to Acids and Alkalis.

Solution	Sample of Aluminum Grams	Amount Lost by Sample Grams	Time of Contact Days	Area of Sample Square Cm.
Acetic Acid - 1%	3.7002	.0022	1	42
	3.7002	.0073	7	42
- 5%	3.6929	.0004	1	42
	3.6929	.0029	7	42
Tartaric Acid - 1%	5.1378	.0040	1	66
	5.1378	.0096	7	66
-5%	5.1397	.0019	1	66
	5.1397	.0077	7	66
Citric Acid - 1%	5.3300	.0006	1	66
	5.3300	.0112	7	66
- 5%	5.3188	.0024	1	66
	5.3188	.0086	7	66
Hydrochloric Acid - 1%	5.4483	.0184	1	66
	5.4483	.0615	7	66
- 5%	5.3868	.0052	1	66
	5.3868	.0370	7	66
Sodium Carbonate - 1%	5.3694	.0488	1	66
	5.3694	.0398	7	66

Table III (Continued)

Solution	Sample of Aluminum Grams	Amount Lost by Sample Grams	Time of Contact Days	Area of Sample Square Cm.
Sodium Chloride				
-1%	5.1324	.0010	1	66
	5.1324	.0082	7	66
	5.1324	.0123	14	66
-5%	2.0576	.0000	1	66
	2.0576	.0018	7	66
	2.0576	.0046	14	66
-25%	2.0546	.0006	1	66
	2.0546	.0021	7	66
	2.0546	.0026	14	66

Where the liquids were clear, the bubbles were seen to form on the surface just as was observed with the tap waters. After standing three or four hours the strips showed no loss in weight. After twenty-four hours, the loss could be detected in most cases. Beyond two days the signs of attack on the aluminum strip became more apparent and the loss in weight larger. The strip was seen to be traced by spots rougher and more worn than the rest of the strip.

Strawberry, logenberry, peach, rhubarb and tomato caused an attack deeper than the other edibles. It was evidently an example of what Selignan and Williams called "pitting". One sample of tomatoes caused an attack through the strip and holes were formed. There was, however, a small amount of baking soda present which some housewives use when cooking tomatoes and this might have been the cause of the holes.

Here, however, we must allow for the strips of aluminum used. Although these were of the same grade there may have been a flaw in one part of the sheet and not in another. For instance the peaches, although they caused an attack that looked deeper than some of the other fruits, actually caused the strip to lose less than orange or pineapple which showed no apparent attack.

Thus we see that there is, of course, no uniformity of attack but if the loss in weight of a strip is plotted against time in days in which the aluminum has been immersed in the liquid, the curve will take the same general shape for all of the juices and the dilute one-half percent acids and bases. The initial attack may be steeper in some cases than in others but then there is uniformly a gradual rise for about a week; then there is a leveling off and a constant weight for a day or two followed by a sharp attack and a rise in the curve during fermentation. Fig. 1 and 2. exemplify this.

In figure 1 the loss in weight of the aluminum strip immersed in peach juice is plotted against the time during which it was exposed to the attack.

Figure 2 shows the result of the attack of one-half percent sodium chloride on an aluminum strip exposed for a definite time in it.

Figure 3, a constantly rising curve from the continued attack, shows the affect of a one percent acid solution.

Figure 1.

Peach Juice

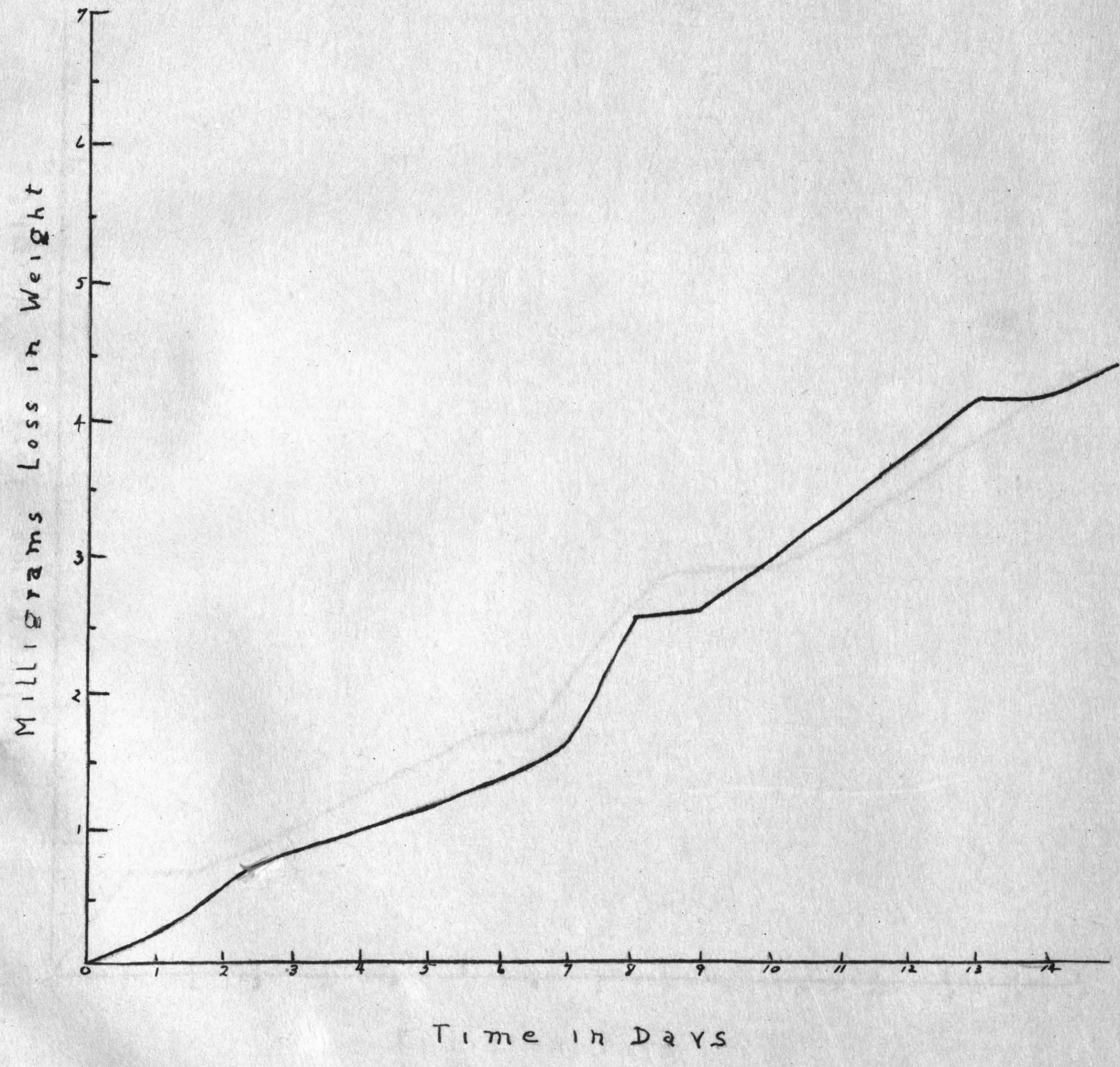


Figure 2

One-half Percent Sodium Chloride

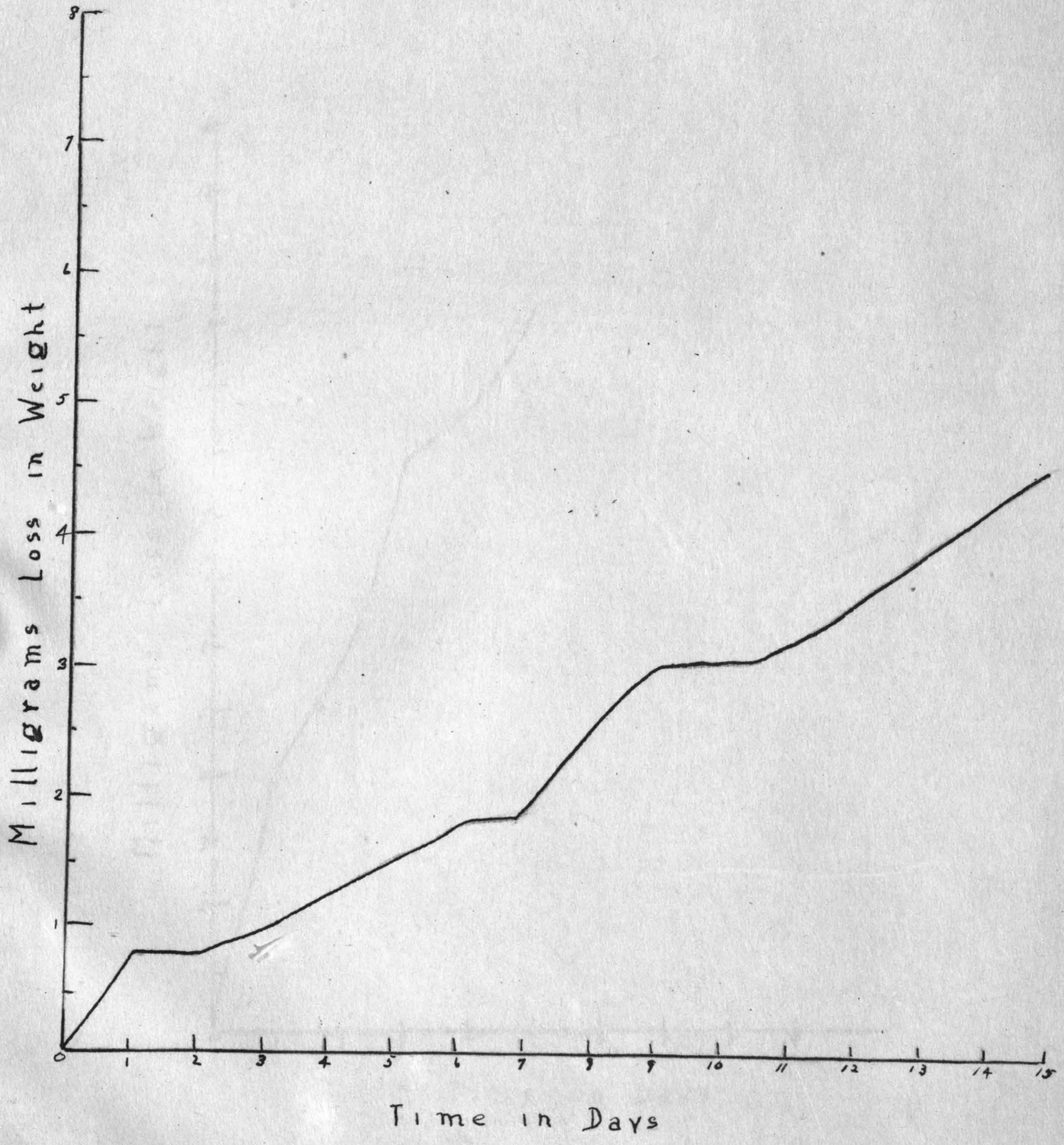
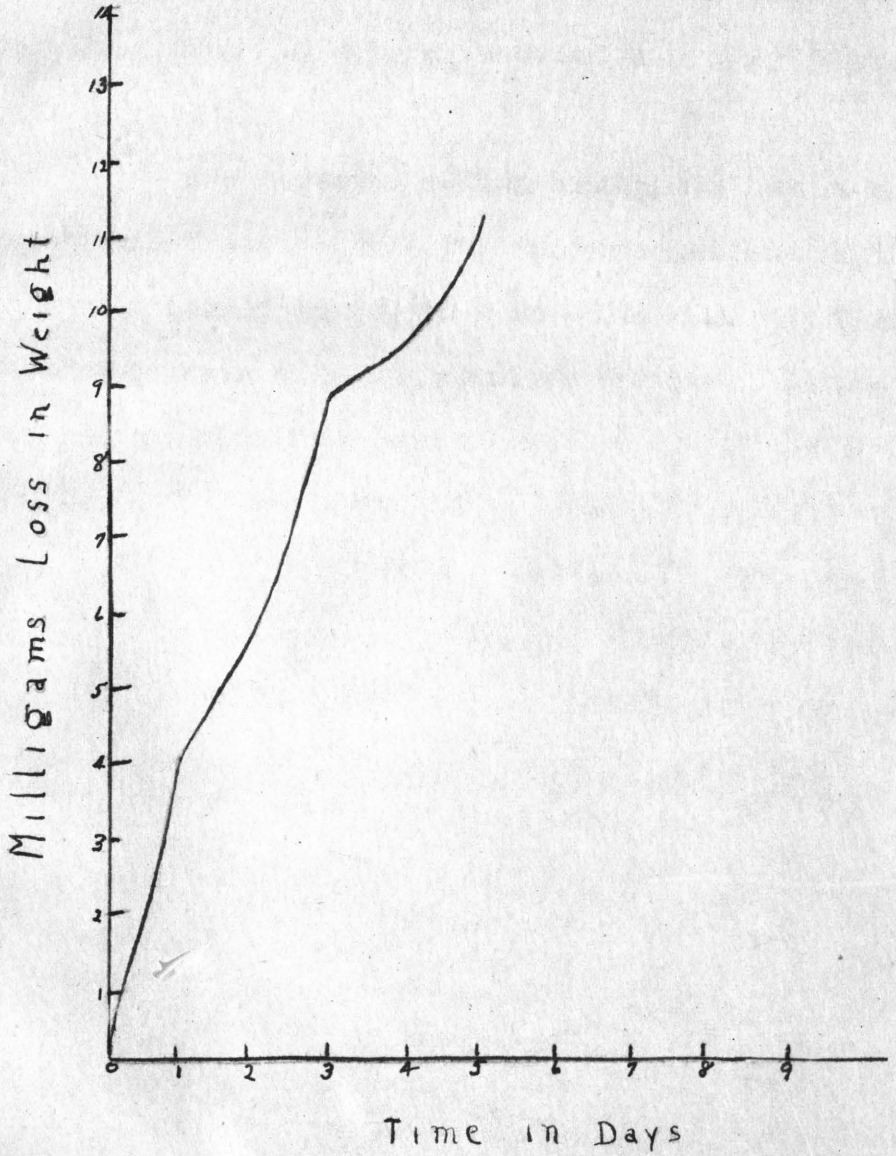


Figure 3

One Percent Tartaric Acid



Tartaric, hydrochloric, citric and acetic acids were used. The first three showed themselves to be much stronger than the fruit acids since it was possible to detect a loss of weight after only four hours of immersion. The acetic acid showed a sharp attack at first also, but after a week, the attack lessened and no loss was apparent after the strip was again immersed for four hours at this point.

One percent sodium carbonate has a more disastrous effect initially than one percent hydrochloric acid which itself is harmful to aluminum. A teaspoonful can quickly ruin a large aluminum kettle. While weak solutions of sodium chloride have very little effect.

The one percent or one-half solutions of acetic tartaric, or citric acids can be compared quite well with the juices. Pineapple, tomato, rhubarb, and cranberry being compared to the one percent acid solutions and the rest to the one-half percent solutions.

CHAPTER V

CONCLUSION

Our first conclusion is that alkalis darken aluminum very much, They put what looks like a tan coat on the strip of aluminum but nevertheless the strip has lost weight and becomes smooth. Acids, on the other hand, make the aluminum strip bright and smooth, removes the shine, and the strip loses weight.

If you examine an aluminum utensil that has been used about a month you can easily see the dark stain that is caused by the alkali in the food itself or from the water in which it was cooked and on examining closer, the little round spots which are the seat of attack becomes apparent.

An older utensil will be darker and there will be spots where the aluminum has been attacked and eaten away.

This loss is what we have attempted to measure. The fruit juices have only remained in contact with the aluminum about two weeks. After that time they start to ferment and the aluminum strip loses weight more rapidly. Tomatoes ferment in less time than this and attack the aluminum more rapidly than do the fruit juices.

In fact it seems to be true that those juices which attack the aluminum the most in a given length of time are those which do not keep well and ferment easily. From this we could say, then, that material which is old will attack aluminum more quickly than fresh material which is farther away from fermentation.

The amount of aluminum lost by an aluminum utensil one day or even two or three days is very small and according to Dr. E.E. Smith, who is a widely quoted authority, one hundred to two hundred miligrams a day could be taken with no harm being done.

This attack probably will vary with different kinds and grades of aluminum utensils.

It also seems quite probable that once the attack has been started, more and more metal is lost and that older utensils will show more attack in a given length of time than a new one will although there is no experimental evidence for this.

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