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Dissolution of Metal Foils in Common Beverages

Bev Christian†, Nancy Wang[2], Mark Pritzker[3], Daniella Gillanders[4], Gyubok Baik[4], Weiyi Zhang[4], Joanna Litingtun[4] and Brian Kim[4]
Blackberry[1,2,4] and the University of Waterloo[3,4]
Waterloo, Ontario, CANADA

Abstract
How susceptible are the metals used in modern electronics manufacturing to corrosion by common beverages? This is a question of interest, especially to manufacturers, retailers and to a certain extent end customers. In this study the dissolution of aluminum, copper, gold, iron, lead, nickel, SAC305 solder, silver, tin and zinc was examined. Individual foils of these materials were fully immersed in one of sixteen chosen beverages and heated for 3 days at 40°C. The resulting solutions were analyzed using ICP-OES. The data were examined in light of the known pH, conductivity and ionic contents of the beverages, determined in previous work. Conclusions about the relative susceptibility to corrosion of the various metals and the corrosive power of the different beverages are made.

Introduction
Corrosion of one form or another affects all matter. It is one manifestation of entropy at work. For modern, portable electronics, a main cause of corrosion is contact with liquids due to spills caused by equipment owners. This particular study was centered on spilled beverages, so no mention is made of tap water, salt water or toilet water - three other very common liquids spilled onto and/or into electronics. In some instances the data presented here may help manufacturers shy away from certain metals in particular circumstances/locations.

Previous work in this series has already looked at the inorganic ionic content of beverages, the measurement of the concentrations of common carboxylic acids in beverages, beverage pH and conductivity, and their effect on the specific metals listed in the abstract and the electrochemical interactions with three of the elements: aluminum, copper and zinc. [1-4].

Copper corrosion rates do increase in increasing concentrations of citric acid. [5] Reference 5 goes on to look at the additional effect on copper when oxalic acid is added to the citric acid, but it will not be described here, as the present focus is on palatable beverages. Another study has shown that low pH in sodium chloride solutions has the highest effect on copper and its alloys at pH 1 and the lowest at pH 7. [6] Chloride is a corrosion promoter in acidic media and an inhibitor in neutral conditions for copper. The combination of different phosphate concentrations and pH levels have a non-linear effect on copper dissolution. In fact different ratios can actually act as corrosion inhibitors. [7]

Another set of experiments looked at the effect on aluminum corrosion of adding small amounts of fluoride ions to varying concentrations of citric acid. The results were fairly dramatic with some increases in dissolution increasing by a factor of about a thousand. [8] Further work has been done using potentiodynamic polarization techniques, but as these methods were not the focus of this work, one is referred to the thesis of one of the authors for further references of this type. [3]

But beverages are even more complex mixtures. For example, aluminum-bronze dental alloy showed no simple linear correlation between corrosion rate and beverage pH. [9] Although the work did show an increasing rate of corrosion with

† Corresponding author, BlackBerry; [2] BlackBerry; [3] Professor, Department of Chemical Engineering, University of Waterloo; [4] Co-op students at BlackBerry from the University of Waterloo

decreasing pH for artificial saliva, sodium chloride, red wine and artificial orange juice; white wine, vinegar and lemon juice showed the opposite pH effect on the alloy.

Another study looked at the corrosion of aluminum by beverages using potentiodynamic polarization measurements. The results showed that the corrosion of the aluminum was slow, time-dependent and complex. It depended on passivation, complexation and adsorption processes. Phosphoric acid was more corrosive to aluminum than citric acid. [10]

Many more references to the interaction of consumables and metals can be found in the book "Mineral Components in Foods". [11]

Method

The beverages chosen were juices, alcoholic drinks, carbonated choices (some overlap) and others. Here is the full list:

Minute Maid™ Apple Juice	Coca-Cola™
Alexander Keith's™ India Pale Ale	Dr. Pepper™
Lipton™ Red Rose Orange Pekoe Tea	Sprite™
Gatorade™ Sports Drink (Fruit Punch)	Canada Dry™ Ginger Ale
Carnation™ Rich Chocolate Hot Chocolate	Pepsi™ Cola
Neilson™ 2% Partly Skimmed Milk	Barq's™ Root Beer
Heinz™ Canada Fancy Tomato Juice	
Starbucks™ Breakfast Blend Ground Coffee	
Wolf Blass™ Yellow Label 2006 Cabernet Sauvignon	
Tropicana™ Pure Premium 100% Pure & Natural Orange Juice (No Pulp)	

For samples which required additional preparation – namely coffee, hot chocolate and tea – manufacturer's instructions printed on the packaging, or close approximations thereof, were followed. In the case of tea, which did not include brewing instructions, one tea bag was allowed to steep for 3 minutes in 250 mL.

Metal sample purities, sizes and the milliliters of beverages the metal samples were immersed in are listed in **Table 1**.
The metals were all prepared in a similar manner. First, they were measured and cut to the sizes listed in Table 1. Most metals were placed in a solution of deionized water and CR-2 low particulate cleaner; each strip was scrubbed on both sides with a soft brush to ensure all dirt and particulates were removed. The strips were rinsed multiple times with deionized water. Following this, they were rinsed with undiluted ASC grade isopropyl alcohol (IPA), and air dried. In order to minimize the effects of surface impurities, each metal was cleaned the day that it was introduced to the sample beverage. As the tin foil was wrapped in plastic wrap prior to its use, it was not washed with CR-2 cleaner. However, it was rinsed with undiluted IPA and air dried to ensure that no surface impurities were present.

Each cleaned strip of metal was lightly folded in half length-ways and inserted into a 75 ml Nalgene vial. A known amount of beverage was then measured using a graduated cylinder and poured into the container such that no metal was left directly exposed to air. The vials were then transported to the oven. A lid was placed on each sample in order to limit the amount of evaporation from the sample. However, it was noted that many beverages tended to explode out of the vial when placed in the oven with a sealed lid, due to increased pressure from de-carbonation or bio-reactions. Therefore, each lid was left partly open so that pressure could escape.

Table 1 Metal sample purities, sizes (mm^2) and the milliliters of beverages the metal samples were immersed in

Metal	Length	Width	Surface Area	mL of Beverage	% Purity
Gold	25	25	1250	20	99.9
Silver	25	25	1250	20	99.9
SAC305	25	25	1250	20	??
Aluminum	80	25	4000	50	*
	80	25	4000	50	99
Copper	99.8	29.9	5968	50	99.9
	100	30	6000	50	99.9
	80	25	4000	50	99.9
Tin	110.5	27	5967	50	99.99
	120	25	6000	50	99.99
Zinc	100	30	6000	50	99.8
	80	25	4000	50	99.95
Nickel	100	30	6000	50	99.9
Lead	100	30	6000	50	99.9
Iron	100	30	6000	50	99.9

*Commercial aluminum foil

Results

The main ionic and ionizable contents of the beverages were measured and reported previously [1,3], so the methods of obtaining the results are not delineated here. **Table 2** lists the concentration of the most common organic acids that were found in the beverages used in the present study. Benzoic, fumaric and ascorbic acids were only found in relatively small amounts in the beverages tested. Wine had the largest number of different acids: lactic, acetic, succinic and tartaric acids. Apple and orange juices had the most malic acid. The most common acid found in the largest quantities was citric acid. Gatorade, ginger ale, milk, orange juice, Sprite and tomato juice had the most of this acid.

Table 3 lists the inorganic anions. Of the six anions listed; chloride, phosphate and sulfate were the most prevalent and show up with the largest concentrations. Chloride was in all 16 beverages and overall has a combined concentration about five times that of sulfate. Phosphate overall had a combined concentration about three times that of sulfate. Sulfate was often present in the 10 to 40 ppm range, except in apple juice (107 ppm), beer (638 ppm), milk (86 ppm), orange juice (84 ppm), tomato juice (140 ppm) and wine (310 pm). Nitrate was only a minor constituent in three drinks and nitrite was only significant in one drink, wine at 933 ppm. Fluoride showed up in moderate amounts with coffee containing a surprising amount.

Table 4 lists the inorganic cations found in the beverages. Iron was only found in small quantities in a few beverages and will not be discussed further. The sum of the amount of potassium in the 16 beverages (~10,000 ppm) was more than the combined total (~6,000 ppm) of the other three main cations; sodium, magnesium and calcium. Not surprisingly, tomato juice and Gatorade had the most sodium. Tomato juice also had the most potassium and magnesium. Milk had by far the most calcium, eight times its nearest rival, beer.

Table 5 contains the information about the pH, conductivity and titratable acids in the beverages. There is no simple, linear correlation between any two of pH, conductivity and titratable acids. However, comparing a sum of the total number of millimoles of titratable organic acids as a function of the sum of the concentrations of the acids detected by IC or HPLC-MS gave a correlation coefficient of 0.88. And it appears that once the concentration of titratable acids rise above 20 millimoles/L or the sum of the concentrations of the acids detected by IC or HPLC-MS is greater than 2000 ppm, the pH settles out between 3 and 4. This means that at the higher concentrations the large amount of carboxylic acids swamp the effects of other dissolved materials with regards to pH. Also, **Figure 1** does show an interesting fact regarding the pH and titratable acids. The beverages that had either low pH (Coke, Pepsi, ginger ale and Dr. Pepper) or high pH (root beer, tea, coffee, milk and hot chocolate) had less than 30 millimoles of titratable acids. This was somewhat unexpected and one might think the low pH beverages would also have high levels of titratable acids in them. **Figure 2** shows a very good correlation between the sum of the inorganic cation concentrations in each beverage and the corresponding measured conductivity of the beverages. This gives more confidence to the actual cation concentration results. The corresponding correlation using anions is not nearly as good.

Table 2 Concentrations of Organic Acids in the 16 Beverages (ppm)

	Lactic	Acetic	Succinic	Malic	Tartaric	Benzoic	Fumaric	Citric	Ascorbic
Apple juice	42	220	6.7	4125				53	11.8
Beer	81	56	143	143				150	
Coffee	108	299	3.6	104			3.8	329	
Coke									
Dr Pepper						253	1.8		
Gatorade								3289	
Ginger Ale						190		1234	
Hot chocolate	161	112	5.7	18.7				335	
Milk				18.4				1270	
Orange juice	121	82	6.1	2135				7415	
Pepsi								55	
Root beer						502		152	
Sprite						61		1135	
Tea				22.6					
Tomato juice	77		4.6	480				2964	8.3
Wine	1080	563	1023	35.4	1345			46	15.4

Table 3 Inorganic Anions in the Beverages (ppm)

	F^-	Cl^-	SO_4^{-2}	PO_4^{-3}	NO_2^-	NO_3^-
Apple juice	199	1251	107	197		
Beer	97	86	638	222	98	
Coffee	149	107	20	293		
Coke	15	33	41	670		
Dr Pepper	22	19	32	470		
Gatorade		456		418		
Ginger Ale		35	27			
Hot chocolate	50	864	28	333		
Milk		991	86	1330		
Orange juice	69	601	84	401		
Pepsi	13	17	29	533		
Root beer	18	30	25			3
Sprite	7	14	22			
Tea	22	14	12	14		2
Tomato juice		3373	140	234		
Wine	398	260	310	859	933	7

Table 4 Inorganic Cations in the Beverages (ppm)

	Na^+	K^+	Mg^{+2}	Ca^{+2}	$Fe^{+2/+3}$
Apple juice	27.6	1120	42.9	55.1	0.737
Beer	24.3	303	76.4	88.6	
Coffee	117	854	36.2	94.2	
Coke	16.2	1.29	2.98	12.5	1.06
Dr Pepper	42.8	2.17	0.302	1.68	
Gatorade	458	149	0.886	2.45	0.58
Ginger Ale	41.3	36.8	12.9	26.1	
Hot chocolate	669	901	25.7	22.1	0.019
Milk	266	994	69.8	716	
Orange juice	5.03	1630	110	83.2	0.54
Pepsi	2.85	24.2	0.253	1.55	156
Root beer	92.7	1.5	4.09	15.8	
Sprite	86.3	1.23	4.25	17.2	
Tea	8.27	112	6.24	0.464	
Tomato juice	2120	2590	96.6	42.1	9.94
Wine	37	943	130	70.5	2.46

Table 5 pH, Conductivity and Titratable Acidity of the Beverages, Ordered by pH

	pH	Conductivity	Titratable Acidity
		(µs cm^{-1})	mmole/L
Coke	2.47	1255	13.67
Pepsi	2.48	1124	16.58
Ginger Ale	2.82	598	24.83
Dr. Pepper	2.86	657	10.58
Gatorade	2.92	2200	46.12
Sprite	3.26	499	18.50
Wine	3.46	2445	82.00
Apple juice	3.57	2375	56.75
Orange juice	3.80	4295	120.42
Beer	3.98	1497	14.50
Tomato juice	4.00	15060	72.00
Root beer	4.24	433	8.00
Tea	5.33	506	4.25
Coffee	6.04	2555	5.37
Milk	6.63	5210	13.17
Hot chocolate	7.01	4760	3.67

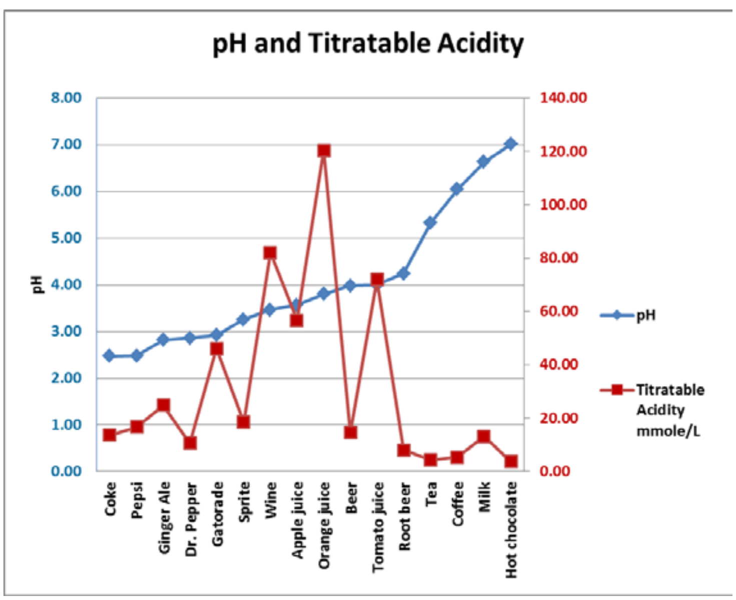

Figure 1 pH and Titratable Acids of the Beverages

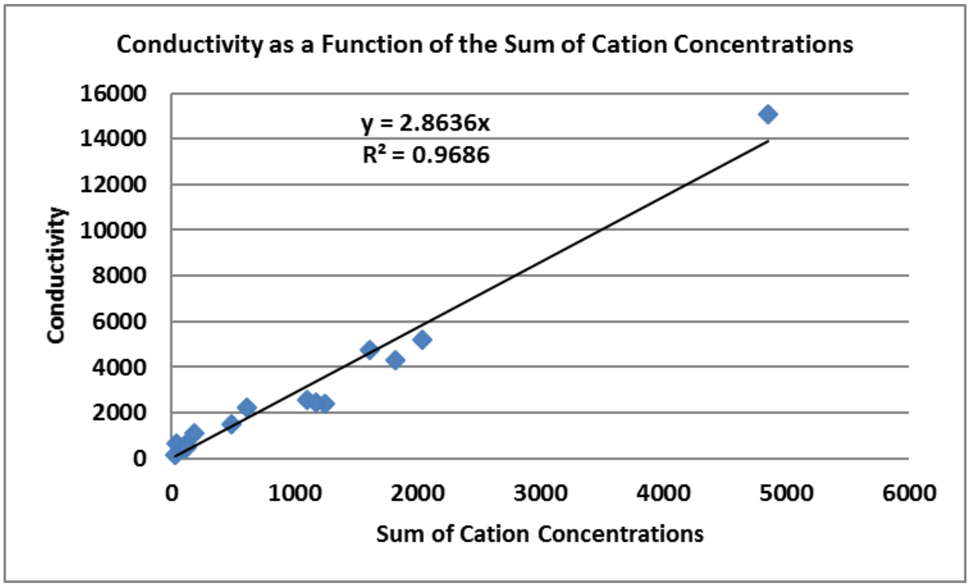

Figure 2 Comparing the Sum of Cation Concentrations of Each Beverage to the Measured Conductivity of the Same Beverages

Figure 3 shows the results for dissolving aluminum in the beverages. The substances are listed left to right in increasing pH. These sets of experiments were carried out by two people separated by a period of two years. Both samples sets were of the same surface in size. The first person used regular aluminum foil purchased from a grocery store and only sonicated the carbonated beverages to remove dissolved carbon dioxide, while the second person used foil specifically purchased from a lab supply house and used a dichloromethane (DCM) extraction carried out three times to remove sugars before subjecting the strips of aluminum to dissolution in the beverages. The results are very similar in most cases, if not statistically the same, except in the cases of Gatorade, ginger ale, root beer and Sprite. Yet the differences for Coke, Pepsi and Dr. Pepper are small; ruling out that sonication or DCM extraction are the cause of the differences.

The question that immediately comes to mind, is there a relationship between the concentrations of dissolved aluminum and different properties of the beverages? **Figure 4** shows a moderate relationship between pH and the amount of dissolved metal.

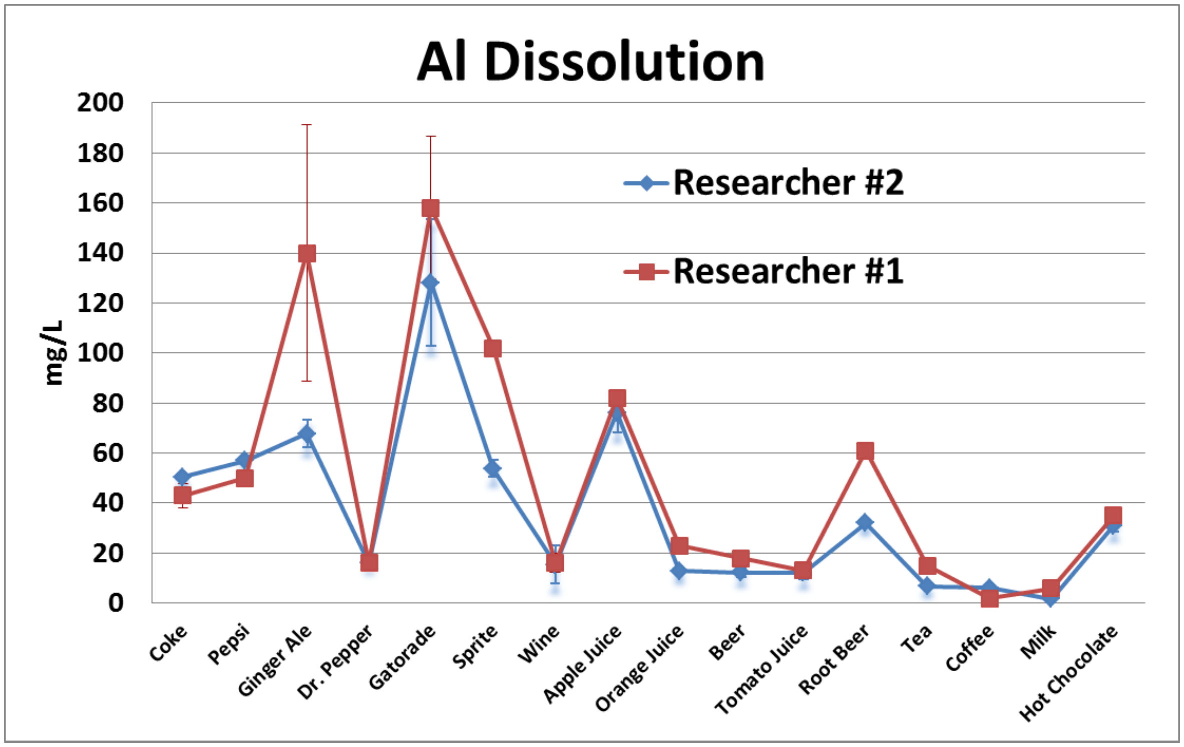

Figure 3 Results for Al Dissolution Carried out by Two Different People

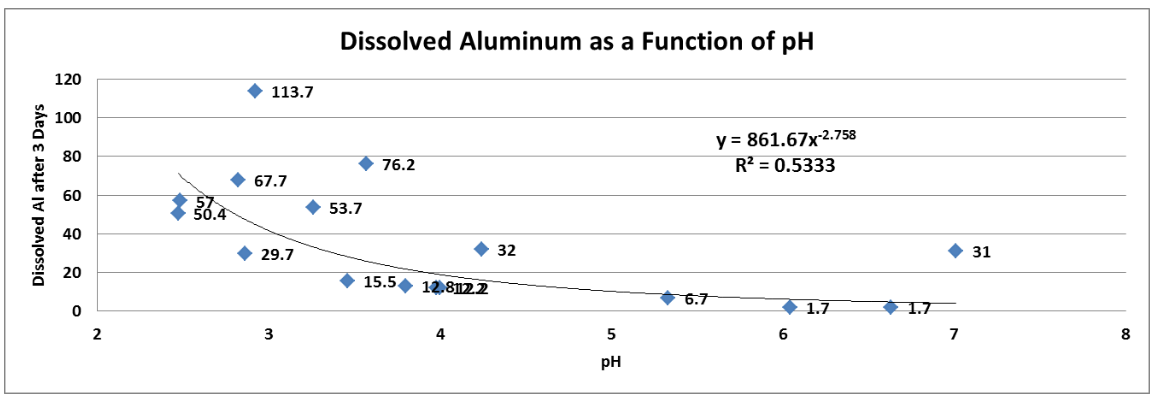

Figure 4 Amount of Dissolved Aluminum as a Function of pH

Concocting an arbitrary relationship using pH, [K^+], [Ca^{+2}], [Cl^-], [PO_4^{-3}], [citrate] and total titratable acid appears to show a better correlation (**Figure 5**) but there are still outliers, like apple juice (76 ppm). Obviously something is still missing. It could be the result of a variable or variables not measured.

Zinc is the fourth most commonly used industrial metal; after iron, aluminum and copper. It is used to coat metals as a sacrificial anode in order to improve the corrosion resistance of the metals on which it is coated. **Figure 6** shows the dissolution results for zinc carried out by two researchers. The data is shown with increasing amounts of titratable acids moving from left to right in the figure. Both used foil from the same supplier, one having foil of 99.8% and the other using 99.95% purity. The standard deviations were smaller than the size of the data points in the figure for most of the beverages and thus did not show up on the chart. The sizes of the pieces of foil used by researcher #2 were only two thirds of the size of the blue data set, thus the raw data values were multiplied by 1.5 for data parity.

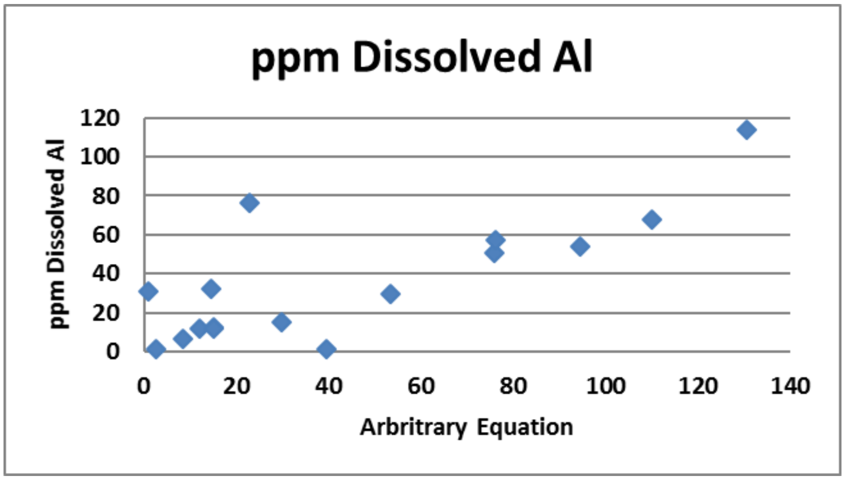

Figure 5 Amount of Dissolved Aluminum as a Function of an Arbitrary Equation Involving pH, [K^+], [Ca^{+2}], [Cl^-], [PO_4^{-3}], [citrate] and total titratable acid

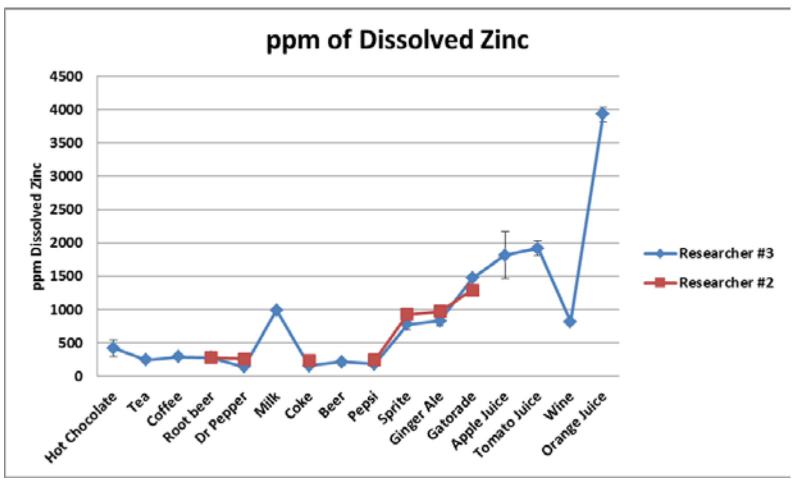

Figure 6 Amount of Dissolved Zinc in Beverages Determined by Two Different Researchers

There is no correlation with pH for zinc. However, there is for the concentration of citrate and dissolved zinc, with a correlation coefficient of 0.80. Two outliers are apple juice (1813 ppm Zn) and wine (813 ppm Zn), which only had around 50 ppm of citrate, but did have large amounts of other carboxylic acids. Apple juice was found to have 4125 ppm of malic acid; while wine had 1080 ppm of lactic acid, 1023 ppm of succinic acid and 1345 ppm tartaric acid. Plotting the sum of the concentrations of the carboxylic acids with the concentration of dissolved zinc shows a stronger correlation. See **Figure 7**. For this pairing the correlation coefficient is 0.91, with the value pair for apple juice falling on the trend line and the result for wine at least much closer, although still the one the farthest away from the trend line.

The study of the dissolution of copper in the 16 beverages is covered in much more detail in references 3 and 4. There is a very general trend of more copper dissolution with decreasing pH and also decreasing chloride. However, no method has been found date to link these two variables or in fact any other of the many possible variables to give a completely consistent trend. See **Figure 8** for the results by two of the study's participants. The two data sets track very well when the data of researcher #2 is multiplied by 1.5 to account for a surface area of 4000 mm^2 as opposed to the 6000 mm^2 for the foils of researcher #4.

The results for tin are shown in **Figure 9**. Pure tin foil and SAC305 foil, which is 96.5% tin, have very similar results. Standard deviation results are available for the pure tin set of data and they have been applied to the data in the figure. The standard deviations for apple juice (33 ppm) and Sprite (37 ppm) were large. For most of the other beverages the standard deviation is smaller than the size of the red data point squares in the figure and thus do not show up on the chart. The result for the dissolution of tin from SAC305 foil in orange juice is anomalous and no explanation can be provided at this time. What is surprising is that the two data sets give very similar results, even though the size of the foils for the SAC305 data set are only about a fifth of the size of those for the pure tin.

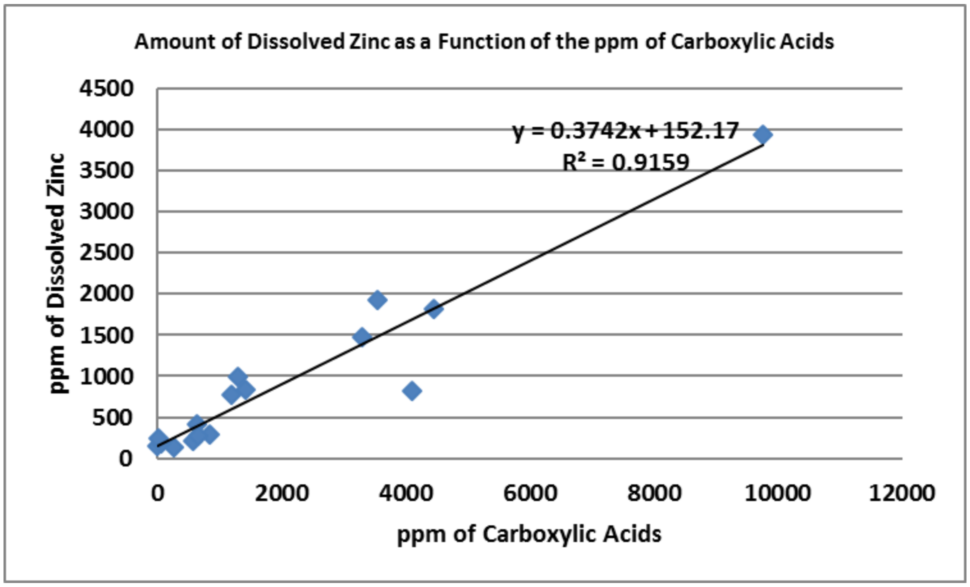

Figure 7 Correlation of Concentration of Dissolved Zn and Summed ppm of Carboxylic Acids in the Beverages

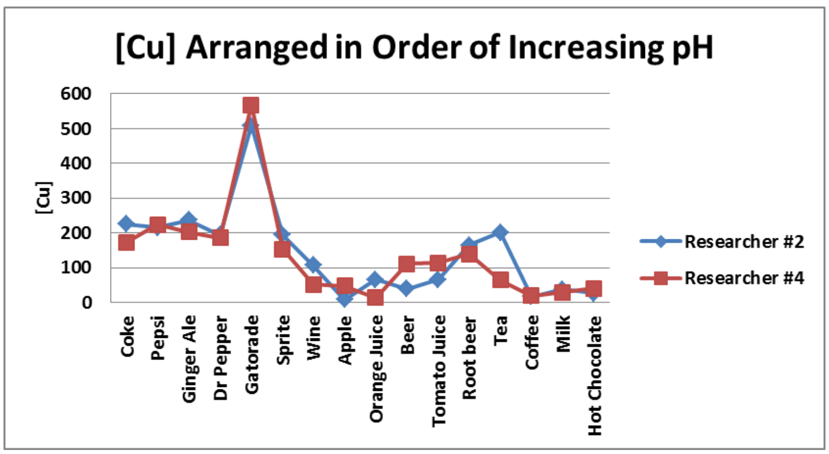

Figure 8 Copper Concentrations as a Function of pH

Figure 10 shows the dissolution results for foils of iron, lead and nickel. Iron was certainly the most soluble of the three and lead and nickel have fairly similar average solubilities in the beverages. The trends for the three metals are different. Nickel shows the previously seen trend of higher solubilities in the carbonated pops. Interestingly, iron shows high solubilities in ginger ale and Sprite, but low solubility in Coke, Dr. Pepper and Pepsi.

Very little gold dissolved in any of the beverages, except in Gatorade and then only to the tune of 0.015 ppm. The silver dissolution was more typical, with the most dissolving in the carbonated pops and the fruit juices. Surprisingly little dissolved in the Gatorade and none was found in the beer, coffee, milk, root beer or tea. See **Figure 11**. **Figure 12** shows that for silver there is a linear, dramatic relative rise in solubility once decreasing pH reaches 4. Orange juice and tomato juice are the two "outlier" orange data points in the figure

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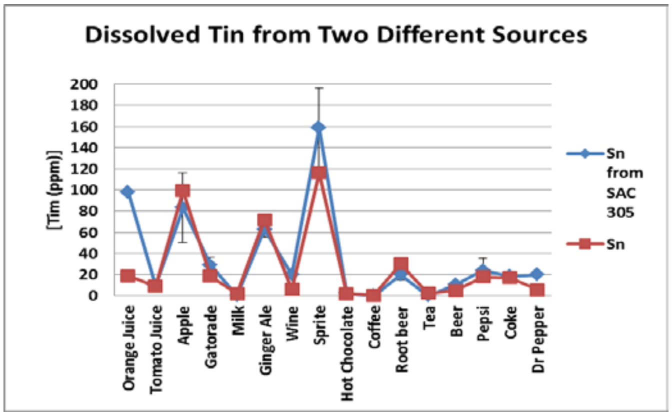

Figure 9 Tin Dissolved from Pure Tin Foil & SAC 305 Foil

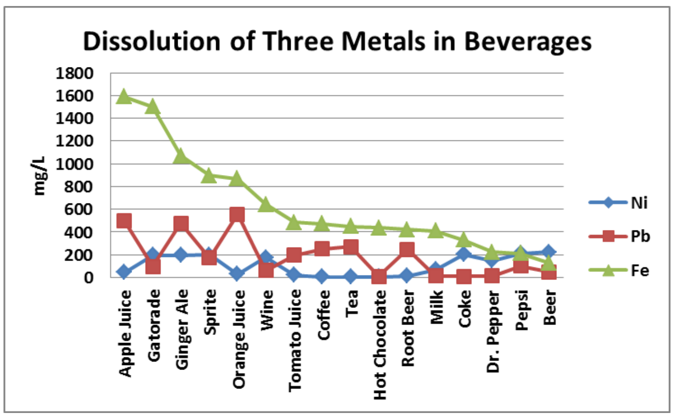

Figure 10 Dissolution of Iron, Nickel and Copper in Beverages

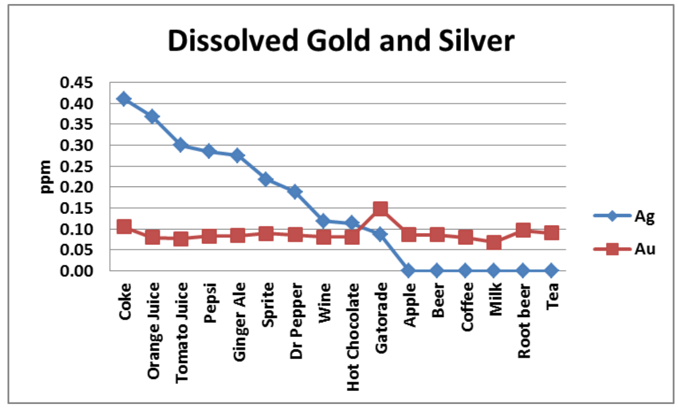

Figure 11 Dissolved Gold and Silver

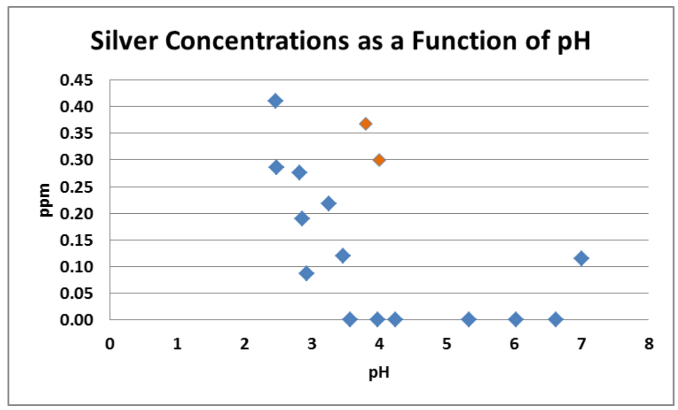

Figure 12 Silver Dissolution as a Function of pH

Table 6 contains all the average data determined by ICP for all the metals dissolved in each of the sixteen beverages. No factors were applied to the dissolution data to attempt to account for the metal strips of different sizes in different solutions volumes. The data are plotted in **Figure 13**. The metals appear to fall into three main groups: iron and zinc, silver and gold and then all the rest; with the latter group being of intermediate solubilities in the beverages used in the study.

A rather simplistic attempt has been made to correlate total metal dissolution in all the beverages to various fundamental properties of the elements. Obviously, the arbitrary nature of lumping everything together masks the nature of the complex differences between the range of beverages. All attempts have been unsuccessful. Two of the other main reasons are that none of the attempted correlations took into account kinetic factors or physical factors like differing oxide tenacity to stay attached to the parent metal. Nevertheless, in **Figure 14** the sum total of the metals dissolved in all the beverages is plotted against the EMF values in volts for the metals examined in this study. All the metals either fall on a fairly smooth curve or close to it, except aluminum. Tin, nickel and SAC305 are three others that are close to the curve but not on it. Perhaps the deviations from the curve for tin, nickel and aluminum can be explained by the tenaciousness of the oxides in question, which all form good native oxide passivations.

Table 6 Summary Table of Dissolution of All Metals in All Beverages in 3 Days

	Zn	Fe	Pb	Ni	Cu	Al	Sn from SAC 305	Sn	Ag	Au
Orange Juice	3930	867	557	31	44	23	98.0	18.6	0.37	0.080
Apple	1813	1590	498	47	5	82	82.9	99.2	0.00	0.086
Gatorade	1470	1500	97	197	338	158	28.9	18.3	0.09	0.149
Ginger Ale	829	1070	475	197	158	140	62.7	71.6	0.28	0.084
Tomato Juice	1920	483	197	22	44	13	9.3	8.7	0.30	0.077
Sprite	777	895	171	202	129	102	158.9	116.1	0.22	0.089
Wine	813	645	60	177	71	16	19.6	5.8	0.12	0.081
Milk	988	411	14	70	25	6	0.0	1.9	0.00	0.068
Root beer	276	423	248	13	110	61	19.7	30.0	0.00	0.097
Tea	243	450	272	6	134	15	0.0	2.2	0.00	0.090
Coffee	286	474	250	5	11	2	0.0	0.4	0.00	0.080
Hot Chocolate	419	438	12	8	17	35	2.1	1.6	0.11	0.081
Coke	152	331	9	206	150	43	18.4	16.9	0.41	0.105
Pepsi	179	57	99	212	143	50	23.7	17.6	0.29	0.083
Dr Pepper	137	225	15	150	129	16	19.9	5.2	0.19	0.086
Beer	212	129	50	223	26	18	10.2	4.5	0.00	0.086

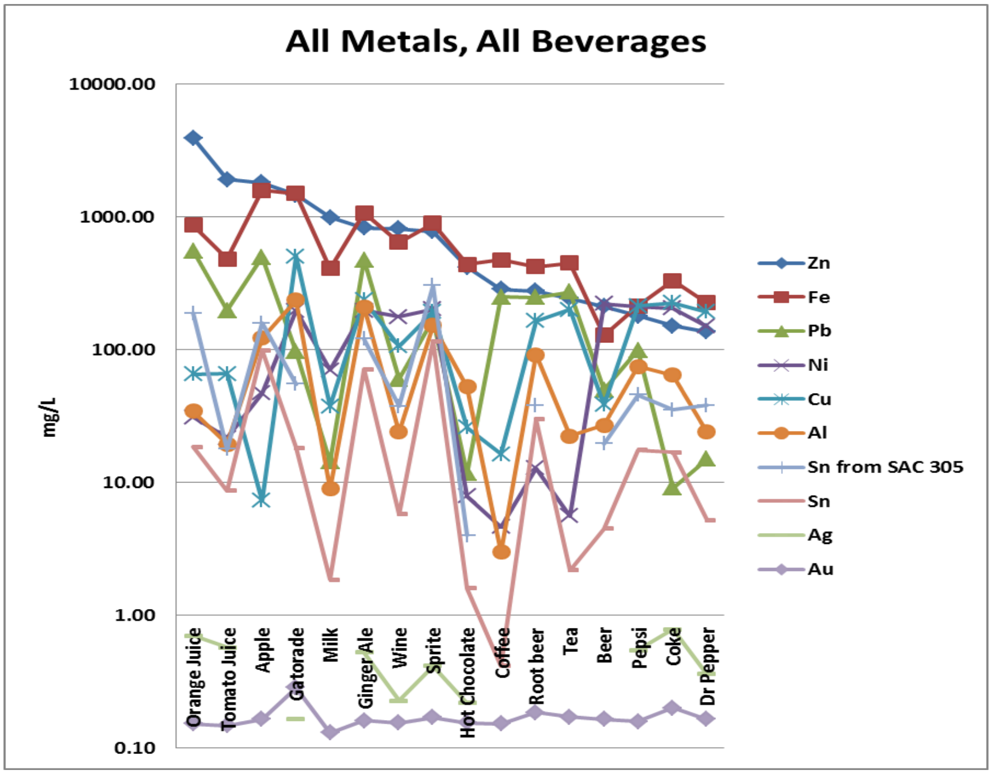

Figure 13 The Data for All Metals Dissolved in All Beverages

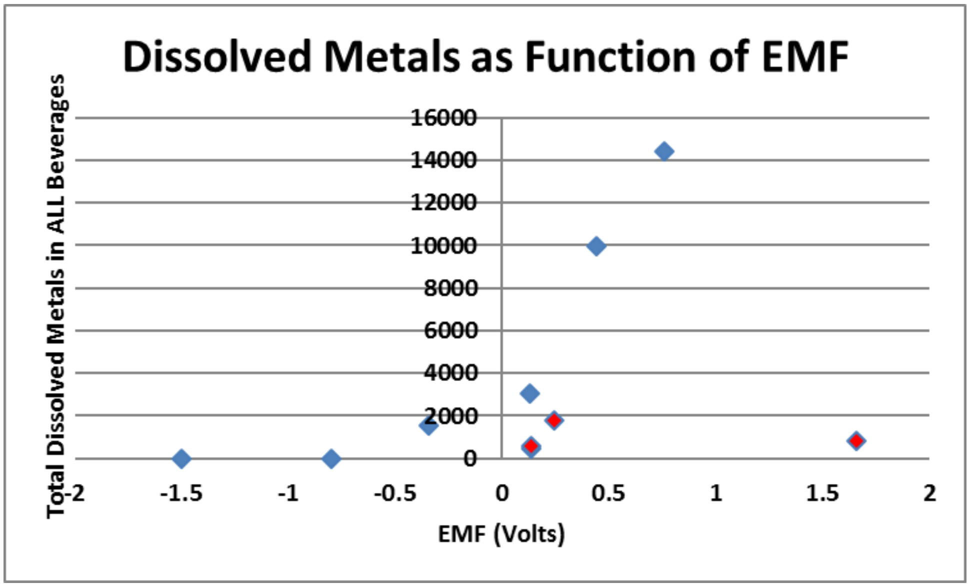

Figure 14 Milligrams of Dissolved Metals as a Function of the Primary EMF of Each Metal

Conclusions

Although the attempt to develop one master predictive equation for dissolution of the metals studied has failed, some general trends can be made from the observed results.

1) Two keys factors are pH and the concentration of carboxylic acids. Both must be taken into account to understand why juices were usually more corrosive than carbonated beverages.
2) Tomato juice is more like orange juice in terms of the order of solubilities of metals in each beverage. Among the carbonated beverages the following similar pairs were found: beer and Pepsi, ginger ale and Sprite and last, Coke and Dr. Pepper. Not surprisingly tea and coffee, both fairly high pH and low concentrations of anything were fairly similar in their dissolving power.
3) The only metal ions that would normally be expected to be in beverages are zinc and iron. Iron is found in Pepsi and tomato juice at 156 and 10 mg/mL, respectively. Zinc is in some carbonated beverages at about 20 mg/mL, while only at 4 to 10 mg/mL for other beverages. [12] As a result, only the initial iron concentration in Pepsi has been subtracted from the value measured for the dissolved zinc. This revised value is what is shown in **Table 6**. None of the other amounts are enough to influence the trends discussed and thus have been ignored with respect to the values in Table 6.
4) In decreasing likelihood of dissolution, the following overall trend was observed: Zn>Fe>Pb>Ni>Cu>Al>Sn>Ag>Au. Except for aluminum, the trend more or less follows the oxidation EMF values for metal to lowest oxidation state.

Acknowledgements

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