Food Quality and Preference 22 (2011) 628–637

Contents lists available at ScienceDirect

Food Quality and Preference

journal homepage: www.elsevier.com/locate/foodqual

The use of standard electrode potentials to predict the taste of solid metals

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ARTICLE INFO

Article history:
Received 25 July 2010
Received in revised form 18 March 2011
Accepted 30 March 2011
Available online 7 April 2011

Keywords:
Copper
Gold
Metallic
Standard electrode potential
Taste
Zinc

ABSTRACT

Not all metals taste equally metallic when placed in the mouth. While much work has been done to examine the metallic taste sensations arising from metal ions in solutions, there is comparatively less known about the taste of solid metals. In this study seven metals in the form of spoons were used to compare the perception of taste arising from solid utensils placed inside the mouth. Thirty-two participants tasted seven spoons of identical dimensions plated with each of the following metals: gold, silver, zinc, copper, tin, chrome and stainless steel. More negative standard electrode potentials were found to be good predictors of solid metals that had tastes scoring highest for the taste descriptors strong, bitter and metallic. Thus, it was found that both gold and chrome (having the most positive standard electrode potentials) were considered the least metallic, least bitter and least strong tasting of the spoons. Zinc and copper (having the most negative standard electrode potentials) were the strongest, most metallic, most bitter, and least sweet tasting of the spoons. We conclude that gold and chrome have tastes that are less strong than metals with lower standard electrode potentials.

1. Introduction

'Metallic' has not been widely accepted as a taste quality descriptor in the psychophysical literature (Bartoshuk, 1978), even to describe sensations induced by electrogustometry (Ajdukovic, 1990). Recently, however, there has been growing evidence that iron ions, particularly in the form of aqueous ferrous sulphate ($FeSO_4$), may act as metallic chemosensory stimuli. Using a multidimensional scaling approach, Stevens, Smith, and Lawless (2006) showed that ferrous sulphate produces a distinctly different sensation from the traditional basic taste descriptors of sweet, sour, bitter, salty, and umami, which have been shown to have unique receptors (Chandrashekar, Hoon, Ryba, & Zuker, 2006).

Yang and Lawless (2005) evaluated the sensory characteristics of 10 divalent metallic salt solutions and showed that among the compounds tested, iron compounds were highest in metallic taste; zinc compounds had higher astringency and a glutamate-like sensation, with magnesium and calcium salts producing a bitter sensation. More recent work has shown that metallic sensations are evoked both by rinses with metal salts and from electrical tongue stimulation (Epke, McClure, & Lawless, 2009; Lawless, Stevens, Chapman, & Kurtz, 2005, 2006). Metallic taste sensations have been shown to be multimodal; iron and copper salts in particular have complex olfactory and gustatory properties including a metallic flavour component that is decreased by nasal occlusion.

Such studies use metallic salt solutions in varying concentrations to test the taste of a particular metal ion. Oral contact was shown to be important for enhancing the impact of the metallic perception in the case of iron and copper (Epke et al., 2009). This result provides evidence that metal salts such as ferrous sulphate generate volatile lipid oxidation products in the mouth that are perceived retronasally as metallic flavours. To a lesser extent, copper salts also evoke metallic taste responses, although they are more complex in their sensory properties, which include bitter, metallic, sour and salty sensations (Cuppett, Duncan, & Dietrich, 2006; Lawless et al., 2005).

The focus in the literature on the taste sensations of iron and copper salts seems partly due to their position as convenient, soluble non-toxic mediators of 'metallic' tastes, and also because they are important for human health and occur naturally in the water supplies (Hoehl, Schoenberger, & Busch-Stockfisch, 2010) and in food (Hurrell, 1999). Both tap water and spring water contain varying concentrations of metal ions, which affect the taste of the water (Bruvold & Pangborn, 1966) and affect its acceptance as 'drinking water' (Whelton, Dietrich, Burlingame, Schechs, & Duncan, 2007). Copper in drinking water can be an important source of dietary copper for humans (Zacarias et al., 2001). Several iron salts have been introduced as food additives for the prevention of iron deficiency, although their use is not straight-forward because they are strong tasting and can also lead to premature spoilage (Hurrell, 2002).

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doi:10.1016/j.foodqual.2011.03.012

Metallic tastes arising from metals that are less soluble than iron and copper have been less studied, especially metals that might come into contact with the mouth not via food or drink, but through the utensils during eating and drinking (Himsworth, 1953). These metals tend to have very low solubilities and are hard to obtain in solution form.

One chemical property of solid metals that may influence their perceived metallic taste is their standard electrode potentials, which is a standard chemical measure of the tendency of a chemical species to acquire electrons and change its ionic state (Atkins and Jones, 2005). As such it indicates broad trends of chemical activity such as inertness and solubility when a metal is placed in an aqueous solution. Since metal atoms must become ions in solution before they can interact with putative taste receptors, it was our hypothesis that standard electrode potentials might be a predictor of the concentration of metal ions, and thus correlated

Fig. 1. The spoons used in the study are pictured. They are stainless steel spoons electroplated with the following metals (from left to right): zinc, copper, gold, silver, tin, stainless steel and chrome.

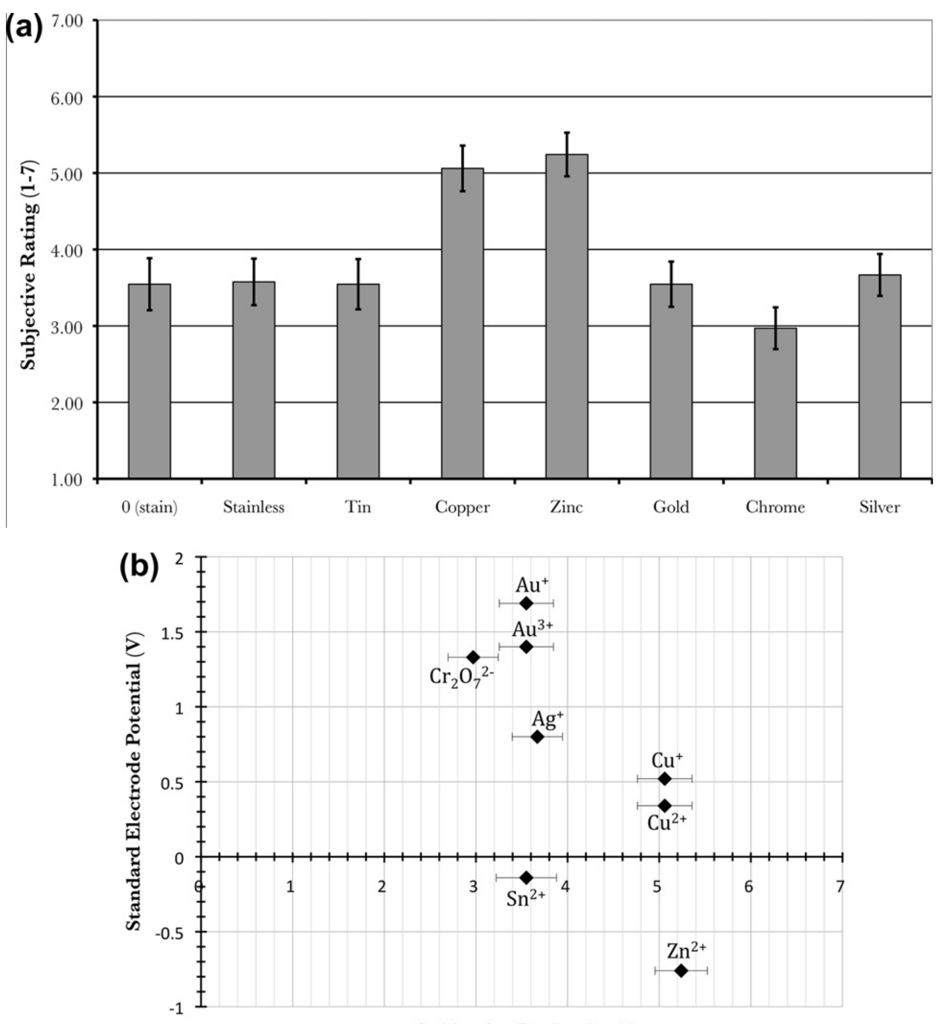

Fig. 2. (a) The subjective ratings of each of the eight spoons in response to the adjective "metallic"; (b) Perception of metallic plotted as a function of standard electrode potential.

with the metallic taste perceived when a solid metal is placed in the mouth. The value of standard electrode potentials are inversely proportional to the tendency of a metal to form metal ions in a standard solution, thus in general we expected an inverse correlation between taste and standard electrode potential on the basis of this hypothesis.

This study considers the effects of metallic tastes arising from solid utensils (spoons), because there is obvious practical significance (i.e., for cutlery) that cannot be extrapolated directly from data associated with metallic solutions, which are more likely to contribute to the generation of volatiles that evoke metallic retronasal perception by catalyzing lipid oxidation (Epke et al., 2009). This study, which involved 32 participants, investigated the differing tastes of seven spoons of identical dimensions plated with each of the following metals: gold, silver, zinc, copper, tin, chrome and stainless steel.

The form of the spoon was chosen because it is readily associated with eating and tasting, thus providing a material form that people would be conceptually and physically comfortable with having in their mouths. Teaspoons were identified as the ideal type of spoon for this study as the bowl of the spoon would be small enough to fit into any adult mouth with ease, and to rest on the tongue without risk of choking. It was expected that the use of solid metals would provide us with novel results that could not be gleaned using metal solutions, such as determining the subjective response to the taste of gold, which is highly insoluble.

2. Materials and methods

2.1. Subjects

Thirty-two participants of mixed ages and both genders (13 males, 19 females) were recruited for the study. To participate in the study, recruits were required to be between 18 and 65 years of age, and in good general health. Specifically they were informed that if they were pregnant, suffering from a cold or flu, or afflicted by any general medical condition known to compromise the senses of taste and smell such as taste-based synaesthesia, any disorders of olfaction (anosmia, hyperosmia, hyposmia, dysosmia) and any disorders of taste (ageusia, dysgeusia), then they could not participate in the study. The upper age limit of 65 was set in an attempt to negate the effect of the loss of taste sensitivity during the normal ageing process (Schiffman, 2009). No bias was given for or against anyone as a result of their gender, ethnicity or nationality. Upon agreeing to take part in the study, all participants signed a consent form but were free to withdraw at any point. Ethical consent for the study was provided by the King's College local ethical review board.

2.2. Spoons

In making the spoons, a number of practical factors had to be taken into consideration. The mechanoreceptors in the mouth are

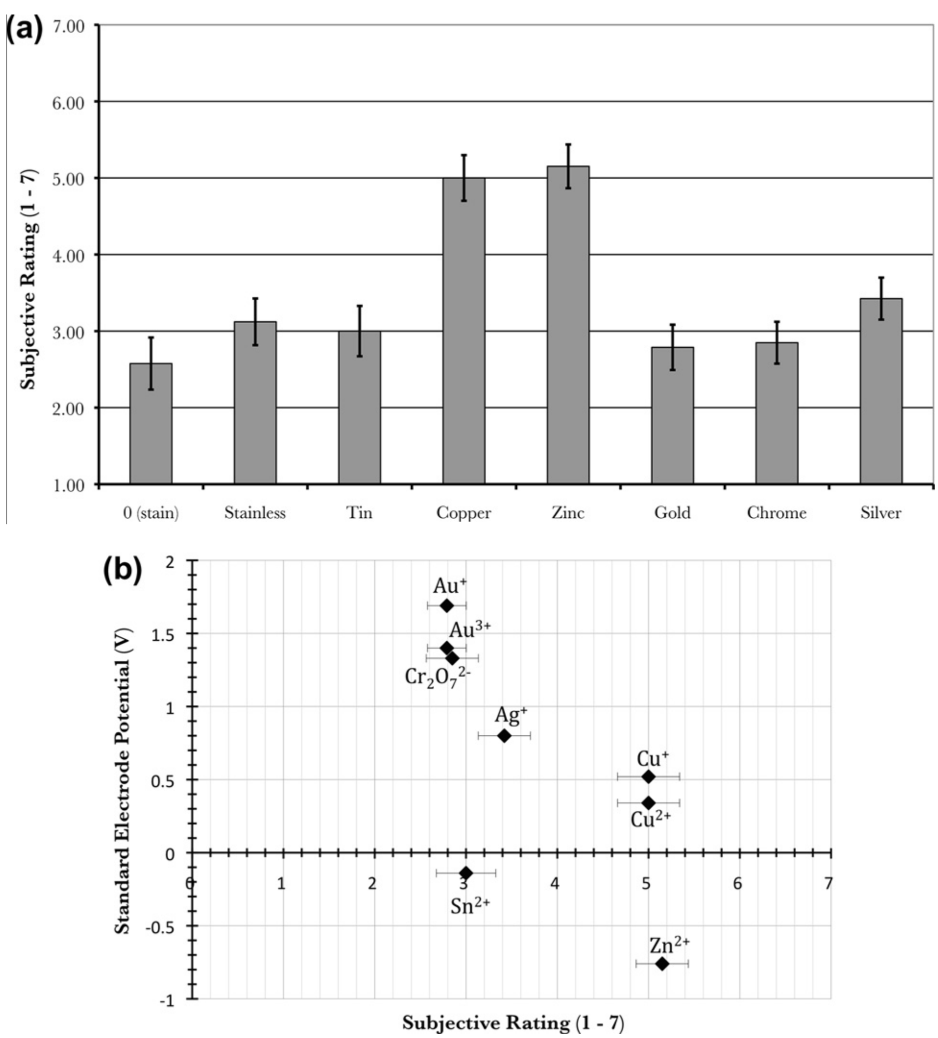

Fig. 3. (a) The subjective ratings of each of the eight spoons in response to the adjective "strong"; (b) Perception of strong plotted as a function of standard electrode potential.

sensitive to differences in size and texture of the spoons. It was, therefore, important to make the spoons of different materials using a technique that would produce spoons of exactly the same size, shape and texture. In order to resolve the issue of producing isomorphic spoons from a range of materials that could be washed between use and whose weight would be similar, it was decided to electroplate commercially available stainless steel teaspoons with a number of different metals. Thus, eight "Sunnex 18/0" stainless steel teaspoons were plated with the following pure elements: zinc, copper, gold, silver, tin and chrome to a thickness of 10 microns (0.01 mm). Although thin, 10 microns provides a homogeneous layer with no possibility of exposure to the stainless steel below it. Two of the spoons were not plated and remained as stainless steel "control spoons". One of each of the spoons of differing materials is shown in Fig. 1. Each metal was selected on the basis of its non-toxic status, suitability for contact with human skin and mucus membranes, its ability to be electroplated, and the ease with which it could be sterilized.

2.3. Testing procedure

Eight teaspoons (2× stainless steel, 1× zinc, 1× copper, 1× gold, 1× silver, 1× tin, 1× chrome) were laid out between two clean white kitchen towels. The temperature of each spoon was taken and found to be at room temperature (21 °C) at the beginning of the experiment. Participants were seated in front of the covered spoons and talked through the experimental procedure. A video camera was set to record and the participant put on a blindfold to insure the differing appearances of the spoons did not affect their responses.

The spoons were then uncovered and the handle of the first spoon placed in the hand of the participant, who then placed the bowl of the spoon into their mouth. The first spoon every participant experienced was always a stainless steel spoon (although the participants were not told that, see below for randomisation). After the spoon had been in the participant's mouth for three seconds, the participant was asked to rate the spoons on a rating scale from 1 to 7 in accordance with the following adjectives (in sequential order): cool, hard, salty, bitter, metallic, strong, sweet and unpleasant. The order of the adjectives was always the same. Our scales ranged from 1 = "not at all" to 7 = "extremely". For example: "How salty was that on a scale of 1 to 7, where 1 is not at all salty and 7 is extremely salty?" There were no verbal cues in the middle of the scale. We consistently reminded participants of the nature of the scale. The participants were required to rate the spoons in the light of all their previous experience of spoons.

Throughout the course of the study, participants were free to take the spoons in and out of their mouths at will whilst considering and rating the spoons. A glass of room temperature distilled water and a receptacle for the disposal of waste liquid was available for each participant, so that they could drink after the tasting of each spoon in order to cleanse and neutralize their palate.

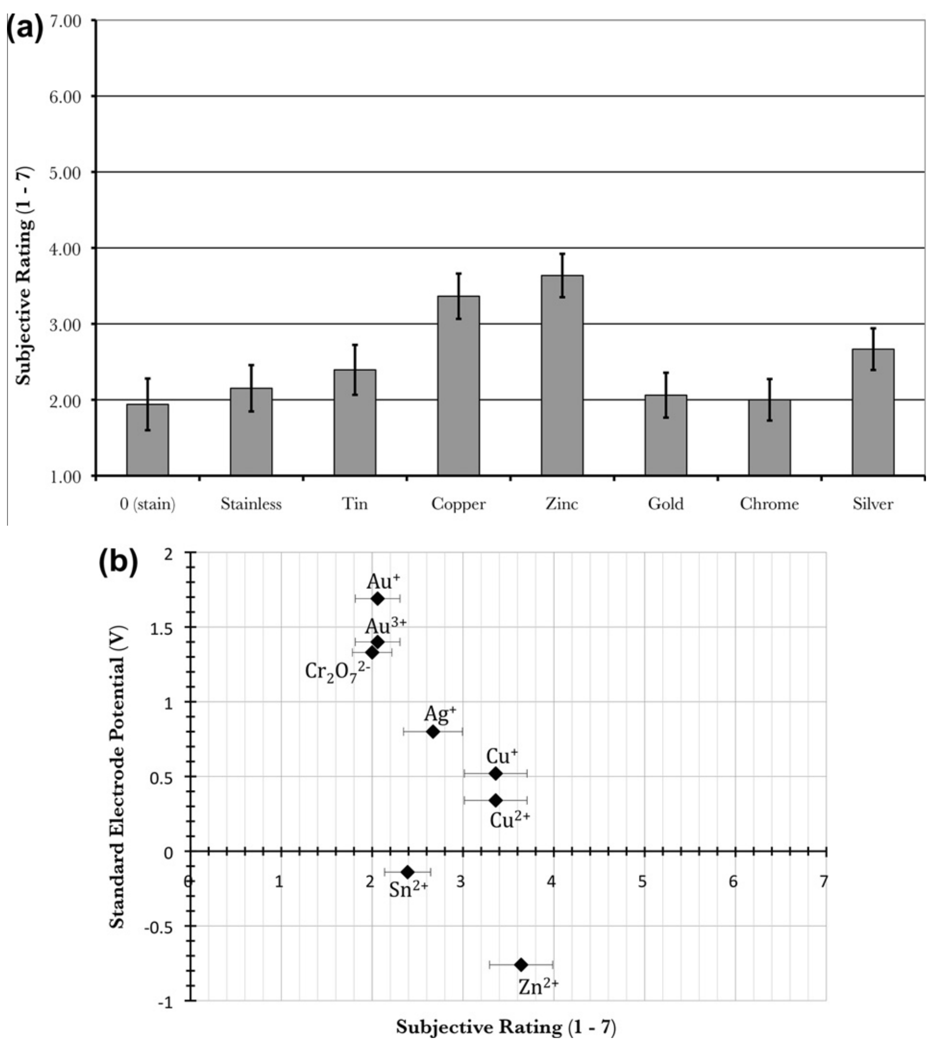

Fig. 4. (a) The subjective ratings of each of the eight spoons in response to the adjective "bitter"; (b) Perception of bitter plotted as a function of standard electrode potential.

Each participant tasted the spoons in differing, randomly generated orders, except for the first spoon, which was always one of the stainless steel spoons; the first spoon was not included in any statistical analyses (except for testing for order effects), and its inclusion in the experimental protocol was meant to eliminate putative order effects due to primacy and unfamiliarity with the experimental procedure (i.e. as a practise run). The randomisation of the order of the remainder of the spoons tasted ensured that results would take into account any cumulative effect of such tasting as well as any order effects associated with the subjective ratings. The ratings of the two duplicate stainless steel spoons were compared to test for a "first-spoon" effect on the participants' blind subjective reports.

Once the participants were finished tasting the spoons, all spoons were washed in hot soapy water and then steam sterilized for ten minutes. Once sterilized, the spoons were removed, dried and left to cool to room temperature before being wrapped in fresh kitchen towel ready for the next participant. These procedures were explained to participants before the experiment began.

2.4. Data analysis

The subjective experiential data was analysed using standard statistical techniques. Repeated measures one-way analysis of variance (ANOVA) with Tukey's Multiple Comparison Test was performed using Prism 3.0 (Graphpad Software Inc., La Jolla, California). For testing for an order effect, the randomised spoons (i.e. not the practise run) were tested via ANOVAs for each adjective. As an additional test for a "first-spoon" effect, the Tukey comparisons from the ANOVA compared the first spoon (which was always stainless steel) to the other stainless steel spoon (which was randomised in the order). For example, Fig. 8 (a) shows that the stainless steel reference spoon ('0 (stain)' in the plots) was rated as significantly warmer than all other spoons, including the other stainless steel spoon ($P < 0.0001$, Tukey's Multiple Comparison Test). All the spoons had been measured and found to be at room temperature (21 °C) at the beginning of the experiment, suggesting that coolness was significantly sensitive to an order effect (although this may be due to either a learning effect or to the participants adapting to temperature of metal spoons by rinsing with water at the same temperature between samples). Coolness was the only sensory descriptor in this study that suffered from a "first spoon effect ($P > 0.05$, Tukey's Multiple Comparison Test); the planned analysis for order effects amongst only the randomised spoons did not detect an order effect for coolness.

Analysing correlations between subjective responses and physical variables (e.g. standard electrode potentials or material hardness) was done with Intercooled Stata 7 (Stata Corp.) using the nominally nonparametric Spearman's rank order test; when analyzing standard electrode potential correlations, we did not use

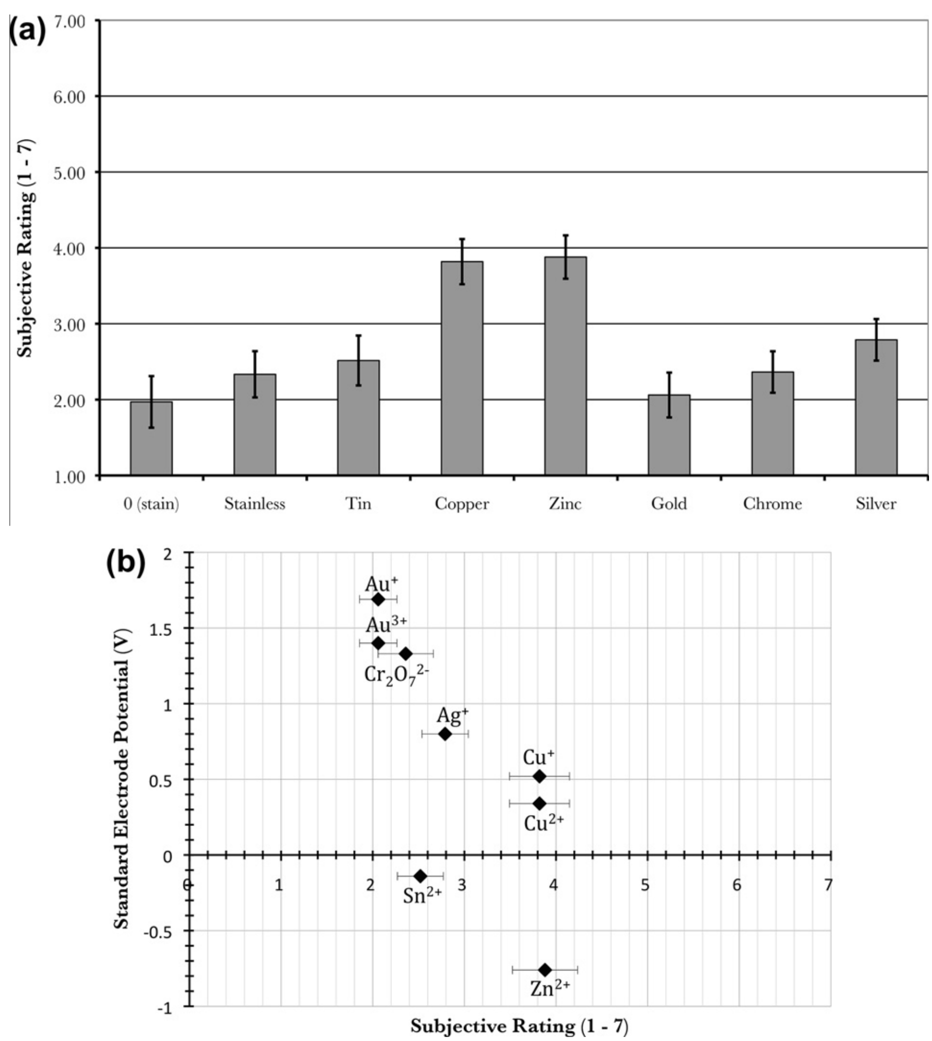

Fig. 5. (a) The subjective ratings of each of the eight spoons in response to the adjective "unpleasant"; (b) Ratings of unpleasant plotted as a function of standard electrode potential.

stainless steel since it was our control. Where Stata returned P values of 0.0000, we report $P < 0.0001$.

Part of the planned analysis was to test for correlations among the adjectives the participants rated by using Spearman's nonparametric correlation test; given that the relationships between the ratings of the spoons for metallic, strong, bitter and unpleasant were so similar, this seemed justified.

Plots investigating the correlation between the perceptions and the relevant physical or chemical property of the pure metals were obtained from standard physical (CES, 2010) and chemical data sources (Atkins and Jones, 2005; Latimer, 1952). For copper and gold, the electrode potential of two oxidation states were plotted since both could be formed in the mouth.

3. Results

The rating of the adjective "Metallic" (Fig. 2) was higher for copper and zinc than for other metals. "Metallic" ratings varied significantly by metal (Repeated Measures ANOVA, $F(6192) = 15.5$, $R^2 = 0.33$, $P < 0.0001$). In using the Tukey multiple comparison test to determine which spoons were statistically different, zinc and copper were significantly different from all other spoons ($P < 0.001$) but not from each other ($P > 0.05$); none of the other spoons (gold, silver, chrome, tin, stainless steel) were significantly different from each other ($P > 0.05$). As such, zinc and copper will be referred to as having a metallic taste, within the context of this experiment. There is an inverse correlation between the electrode potentials of metal ions and perceived metallic taste of the metals (Fig. 2b); in a Spearman rank order analysis, Spearman's $\rho = -0.31$ and $P < 0.0001$.

The adjective "Strong" (Fig. 3) was highest for zinc and copper; it varied significantly by metal (Repeated Measures ANOVA, $F(6192) = 21.7$, $R^2 = 0.40$, $P < 0.0001$). In using the Tukey multiple comparison test to determine which spoons were statistically different, zinc and copper were perceived to taste stronger than all other spoons ($P < 0.001$) but not from each other ($P > 0.05$); none of the other spoons (gold, silver, chrome, tin, stainless steel) were significantly different from each other ($P > 0.05$). In a Spearman rank order analysis between each metal and the standard electrode potential, Spearman's $\rho = -0.34$ and $P < 0.0001$.

The adjective "Bitter" (Fig. 4) was rated most highly for copper and zinc, and it varied significantly by metal (Repeated Measures ANOVA, $F(6192) = 7.9$, $R^2 = 0.20$, $P < 0.0001$). In using the Tukey multiple comparison test to determine which spoons were statistically different, zinc and copper (the strongest tasting spoons) were perceived to taste more bitter than chrome, gold and stainless steel ($P < 0.01$ for all) but not from each other ($P > 0.05$); none of the other spoons (gold, silver, chrome, tin, stainless steel) were significantly more bitter than each other ($P > 0.05$). Fig. 4(b) shows a clear inverse linear correlation between the electrode potentials of metal ions and perceived bitterness of the metals. In a Spearman

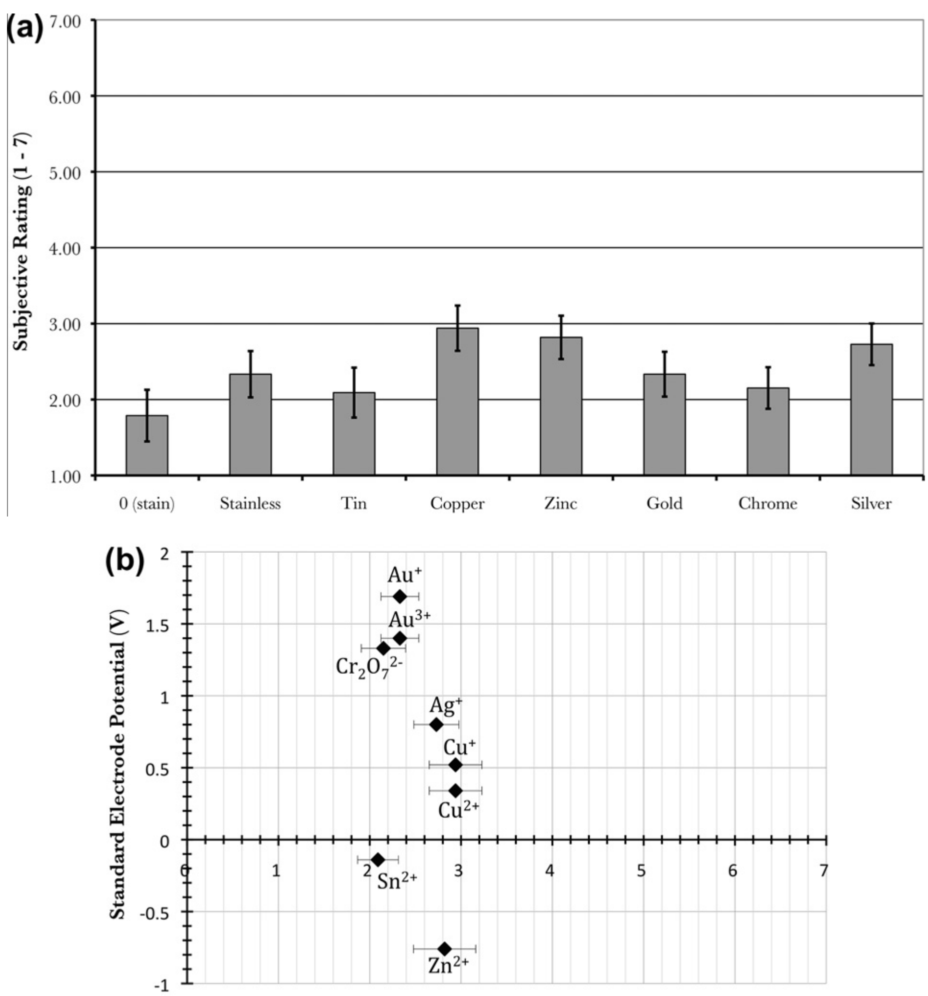

Fig. 6. (a) The subjective ratings of each of the eight spoons in response to the adjective "salty"; (b) Perception of salty plotted as a function of standard electrode potential.

rank order analysis between each metal and the standard electrode potential, Spearman's $\rho = -0.26$ and $P = 0.0002$.

The adjective "Unpleasant" (Fig. 5) varied significantly by metal (Repeated Measures ANOVA, $F(6192) = 10.8$, $R^2 = 0.25$, $P < 0.0001$). In using the Tukey multiple comparison test to determine which spoons were statistically different, zinc and copper were perceived to taste more unpleasant than all other spoons ($P < 0.001$ for all) except silver (significant at $P < 0.05$ for both copper and zinc) but not from each other ($P > 0.05$); none of the other spoons (gold, silver, chrome, tin, stainless steel) were significantly more unpleasant than each other ($P > 0.05$). In a Spearman rank order analysis between each metal and the standard electrode potential, Spearman's $\rho = -0.28$ and $P = 0.0001$.

The adjective "Salty" (Fig. 6) varied significantly by metal, with copper and zinc having the highest means, but the *R*-squared value of the Repeated Measures ANOVA was comparatively weak ($F(6192) = 2.7$, $R^2 = 0.08$, $P < 0.05$). In using the Tukey multiple comparison test to determine which spoons were statistically different, no comparison reached significance ($P > 0.05$ for all). There was no significant relationship between each metal and the standard electrode potential (Spearman's $\rho = -0.03$ and $P = 0.64$).

There were no statistically significant differences in sweetness (Fig. 7, $P = 0.05$), coolness (Fig. 8, $P = 0.50$) and mechanical hardness (Fig. 9, $P = 0.61$). In Spearman rank order analyses between each metal and the standard electrode potential, for sweetness Spearman's $\rho = 0.13$ and $P = 0.068$, for coolness Spearman's $\rho = 0.03$ and $P = 0.68$, and for hardness Spearman's $\rho = -0.05$ and $P = 0.53$.

In a Spearman rank order analysis between the subjective hardness of each metal-plated spoon and the physical hardness of the metal plating the spoon, Spearman's $\rho = -0.013$ and $P = 0.83$. This lack of correlation is not surprising, as the subjective hardness ratings may relate more to the stainless steel spoon underneath than to the 10 micron coating.

In testing the correlations between the different adjectives, the descriptor "strong" was always significantly correlated with bitter, unpleasant, metallic, and salty ($P < 0.01$ for all), but never with sweet, hard or cool. "Metallic" was invariably associated with strong and unpleasant ($P < 0.01$ for all). Bitter was inconsistent in that it varied according which spoon was considered. Although the graph suggested a trend, the descriptor sweet was never significantly inversely correlated with bitter or unpleasant, and it was never inversely correlated with metallic except for the zinc spoon.

4. Discussion

More negative standard electrode potentials appear to be good predictors of the perceived tastes of solid metals described as metallic, bitter, and strong, showing an inverse correlation. The zinc and copper spoons stand out in the plots in Figs. 2–7 as the

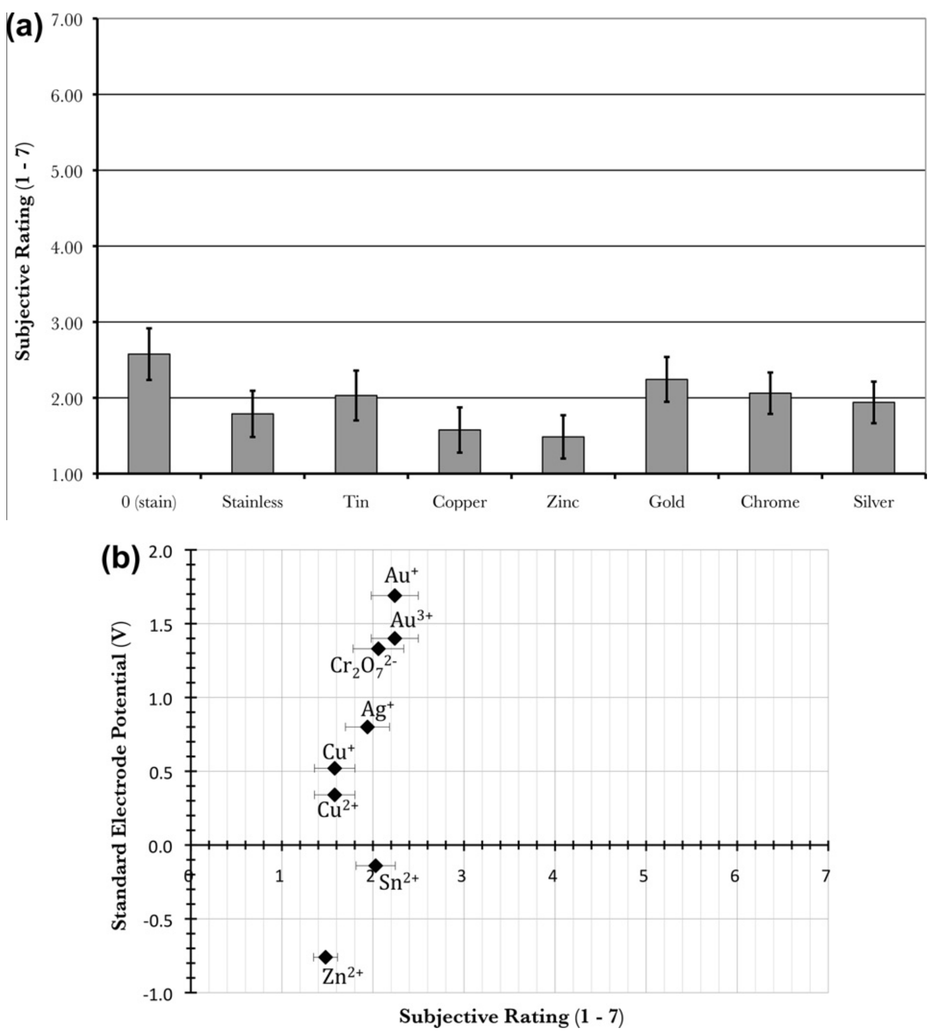

Fig. 7. (a) The subjective ratings of each of the eight spoons in response to the adjective "sweet"; (b) Perception of sweet plotted as a function of standard electrode potential.

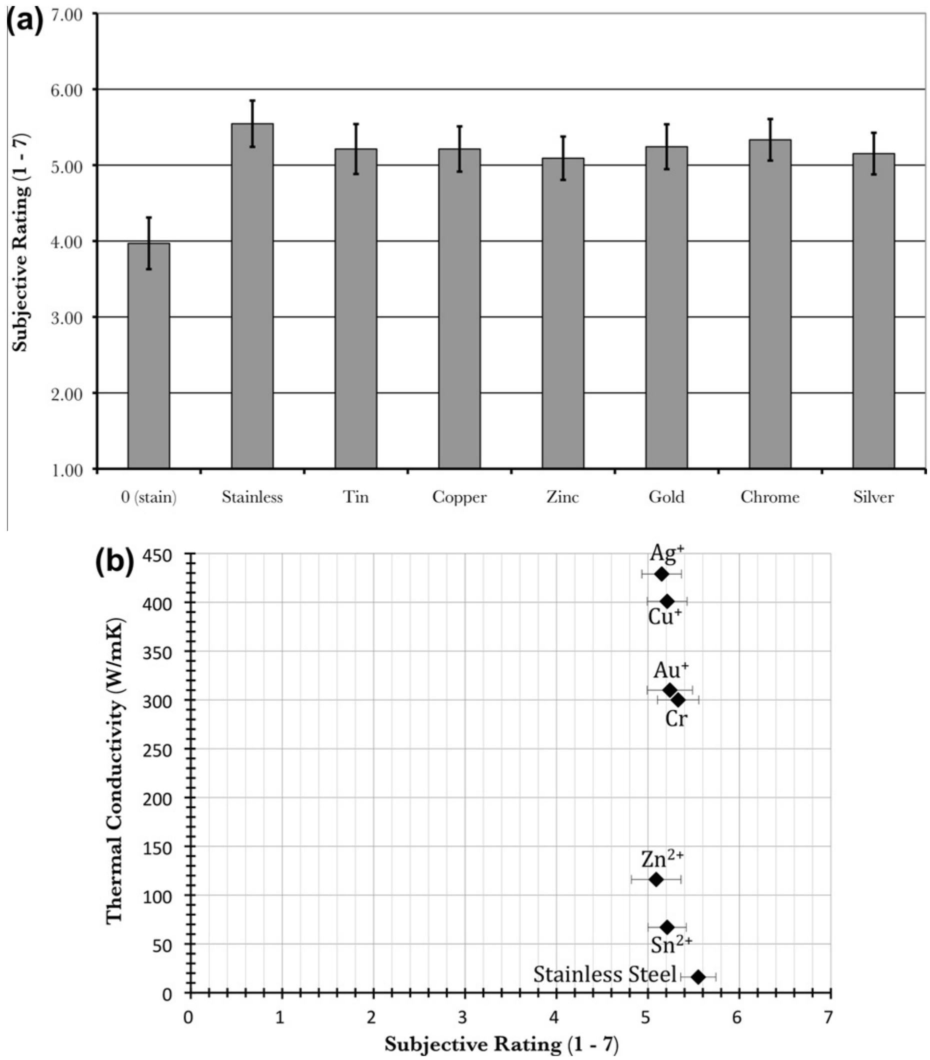

Fig. 8. (a) The subjective ratings of each of the eight spoons in response to the adjective "cool"; (b) the perception of cool plotted as a function of measured Thermal Conductivity.

most significantly strong tasting spoons, they were rated highest for the adjectives bitter, metallic, and strong. The gold and chrome spoons were frequently commented on by many participants as the most pleasant tasting of the spoons. On placing them in their mouths, participants would often say how they liked these spoons or were at least struck by the absence of taste, but our testing methodology was not adequate to confirm this. Gold was determined as being the least strong tasting of the spoons, closely followed by chrome. The chrome spoon was rated as even less metallic in taste than the gold spoon, making it the least metallic tasting of all the spoons. The taste descriptors sweet (Fig. 6b) and salty (Fig. 7b) do not seem to be strongly correlated with electrode potential. Despite this, the gold spoon emerged with the highest sweet rating of all the spoons.

The results in the present study complement, from a materials science perspective, the work by Lawless et al. (2005) by comparing other solid metals (silver, tin, chromium and gold, some of which are used in modern cutlery) to those known to produce metallic tastes directly in the oral cavity (e.g. zinc and copper). In past experiments, some of which date back all the way to 1752 (for review, see Bartoshuk (1978)), the metallic taste was thought to be similar to those produced by tasting individual solid metals and to electrogustation (including putative electrical stimuli elicited by tasting combinations of solid metals (Lawless et al., 2005)). More generally, in experiments on metallic taste, the standard stimuli are a ferrous sulphate solution and a clean copper penny (solid metal) (Civille & Lyon, 1996). Lawless et al. (2005) have shown that, although these two different stimuli seem to share a metallic taste, the taste is elicited by different sensory mechanisms: the metal solutions have a significant olfactory component requiring retronasal sensation, whereas solid metals do not contribute to generation of volatile oxidation products.

We have tested other solid metals in the oral cavity and shown that their taste is not as metallic as either copper or zinc – as would be predicted from their more positive standard electrode potentials. One of the other clear results from Lawless et al. 2005 is that when tasting two adjacent metals with different standard electrode potentials (e.g. zinc and copper), this produces an intensified metallic taste that is more akin to electrogustation than to the taste of a single metal. For electrogustation (as well as for solid metals), the taste sensation appears to be a genuine taste and not affected by retronasal stimulation of olfaction, implying that the receptors for solid metals are oral (and mostly concentrated on the tongue). This result is consistent with our hypothesis, because a combination of copper and zinc in the mouth, produces an effective battery which drives a current of metal ions into the mouth, increasing their concentration. Together, the present study and Lawless et al. (2005) suggest that the receptors of the taste of solid metal

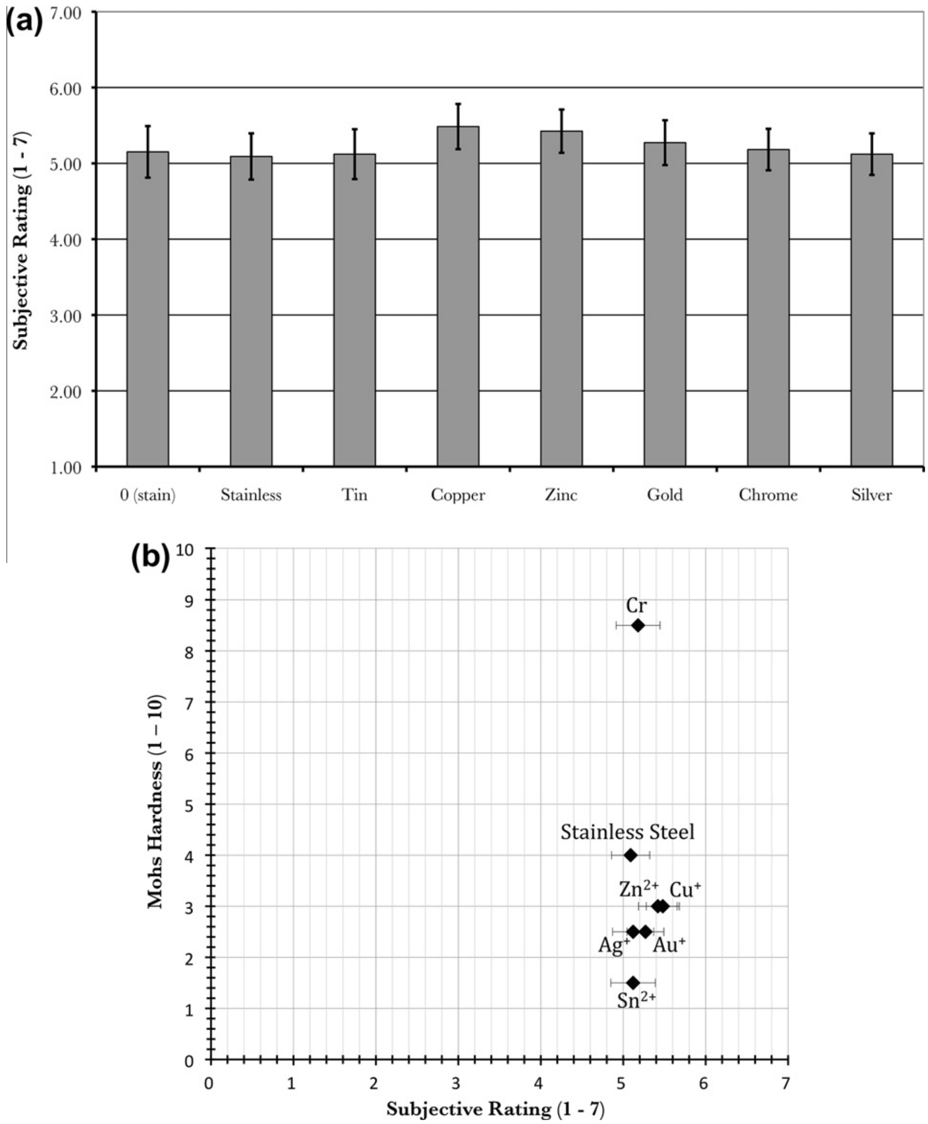

Fig. 9. (a) The subjective ratings of each of the eight spoons in response to the adjective "hard"; (b) Perception of hard plotted as a function of measured Hardness.

(possibly associated with fungiform taste buds) will be most stimulated by metals with a low or negative standard electrode potential.

The standard electrode potential of tin ($Sn^{2+} + 2e^{-} \geqslant Sn(s)$) is -0.13 V, which according to the correlations in Figs. 2–5, would tend to suggest that tin should be rated higher than copper for the taste sensations strong, metallic, and bitter. The fact that it consistently scored lower than copper may indicate that dissolution of tin in the aqueous pH neutral environment of the mouth is hampered in some way, perhaps by a stable oxide layer as in the case of chromium. The stable oxide layer of tin has been proposed to account for the success of tin plating of steel cans (Hassan & Fahmy, 2008) for the preservation of the flavour and appearance of food (Blunden & Wallace, 2003), which has been common practice in the food packaging industry for more than one hundred years. Stainless steel and chromium have similar transparent oxide layers, which are mechanically stable, chemically inert, and are responsible for their lack of taste. Hong, Duncan, and Dietrich (2010) have shown that astringency due to the presence of copper ions changed as a function of pH, as a result of lower solubility. In aqueous solution at pH > 2, Sn^{2+} will form $Sn(OH)_2$, which has very low solubility (Duffield, Morris, Morris, Vesey, & Williams 1990) and creates a passivation layer. Nevertheless, tin has been shown to diffuse into canned food at appreciable levels depending on storage conditions without unduly affecting the taste of the food (Blunden & Wallace, 2003). Thus, we suggest that the relatively reduced taste of tin is most likely due to the formation of a passivation layer preventing the formation of tin ions in the mouth, but we cannot rule out that any putative taste receptors in the mouth have a lower sensitivity to tin ions.

The silver spoon rated above all but the zinc and copper spoons for saltiness, bitterness and strength of flavour. It was some way behind the zinc and copper spoons, however, suggesting that, despite being more pronounced than for some spoons, the taste of the silver spoon was still subtle. Silver nitrate solutions are extremely bitter tasting, suggesting perhaps that the reason for the muted flavour of solid silver is its low solubility in the pH neutral environment of the mouth. Alternatively the bitter taste of sliver nitrate may be due to nitrate anion: nitro- and nitroso-compounds in plants, alkaloids, produce a very bitter taste (Luch, 2009).

In popular lore there is a general presumption that metallic tastes are unpleasant, and this can be seen in some papers in the literature as well (e.g. Lawless et al., 2004). In the present study, the descriptor metallic was statistically correlated with both the adjectives "unpleasant" and "strong", which may suggest that when considering metal spoons, that a metallic taste is considered both strong and unpleasant (Spearman's $\rho > 0.35$, $P < 0.05$ for all seven metals). This raises the possibility that our measurements of metallic tastes (where gold and chrome were the least metallic) may correlate with 'preference' for different metals. Further study with larger population will provide better understanding on hedonic responses to different metals.

We conclude that the taste of solid metals are dependent on their standard electrode potentials. Gold and chrome have tastes that are less metallic, less bitter and less strong than metals with lower standard electrode potentials, especially zinc and copper.

Acknowledgements

We gratefully acknowledge Henry Roeland Byrd for technical insights. This research was supported by a grant F/07 040/AF from the Leverhulme Trust; they did not contribute to the design or analysis within this study.

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