



Kinetics and equilibria of tea infusion. Part 11—The kinetics of the formation of tea scum

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(Received 28 April 1993; accepted 1 June 1993)

A detailed study has been made of the rate of scum formation in hard London mains water at 80°C. From a given black tea infusion the scum grew continuously and, in the first hour, proportionately with time. The amount formed depended on the exposed surface area and not on the volume of the tea brew. Longer infusion of a given mass of tea leaf first increased and eventually decreased the mass of scum. Surprisingly, the more tea leaf that was used the less scum was produced, a phenomenon largely caused by the concomitant decrease in pH.

Tea scum only formed in water which contained both calcium (or magnesium) ions and bicarbonate ions. No scum developed if the former were removed by complexing agents or the latter by lowering the pH. Boiling the water reduced but did not eliminate scum. More scum formed if the period of scum development took place at a higher temperature and the activation energy of scum formation was found to be 34 kJ mol⁻¹. This relatively high value, as well as the absence of any effect when the infusion was stirred, showed that the overall process is chemically- and not diffusion-controlled.

Decaffeinated tea gave a similar amount of scum as normal black tea while green Chun Mee leaf produced about 80% as much. Moreover, scum formation was inhibited by passing nitrogen over the surface but enhanced by passing oxygen. Taken together, these experiments suggest that scum is produced by oxidation of tea solubles mediated by calcium carbonate formation.

INTRODUCTION

A film of tea scum forms on tea infused in hard water. In the preceding paper (Spiro & Jaganyi, 1994) the scum was shown to consist of high molecular weight organic material together with islands of calcium carbonate. The present paper reports on the rate of formation of such films and on the factors which influence the rate.

MATERIALS AND METHODS

The method of scooping the scum from the surface of tea infusions was described in the previous paper (Spiro & Jaganyi, 1994). The 'standard procedure' was to infuse one teabag of a popular black tea blend (Typhoo) for 5 min in 800 ml stirred London mains tap water at 80°C, and to allow the resulting infusion to stand for 60 min at 80°C before collecting and drying the scum. For the kinetic studies several of the above parameters

were varied, in particular the amount of tea leaf, the composition of the water, the 'infusion time' during which the leaf was immersed in the hot water, the subsequent 'standing time' allowed for the scum to form after removal of the tea leaf, and the temperature and gaseous atmosphere during this period. Every experiment was repeated at least once and the results below refer to the mean values. On average, the standard deviations of the means were 0.3–0.4 mg.

RESULTS AND DISCUSSION

Variation of standing time

For given infusion conditions, the mass of scum always increased linearly with time during the first hour (Fig. 1) with a somewhat slower increase for several hours thereafter (Fig. 2). Once this had been established, it became unnecessary to measure the scum weight at several different times for each change of conditions and a standard standing time of 60 min was adopted. During this period the pH increased by *c.* 0.1.

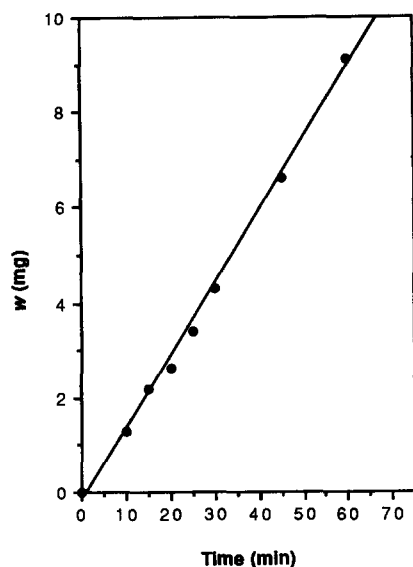


Fig. 1. Variation of the mass of tea scum with time over the first hour at 80°C, after a standard infusion of one teabag in 800 ml mains water for 5 min.

Variation of infusion time

Figure 3 shows the results of experiments carried out at 80°C with one teabag, 60 min standing time, and infusion times varying from 15 s to 15 min. The amount of scum is seen to increase up to a fairly flat maximum value of 9.1 mg after 5 min infusion and to decrease thereafter. For this reason an infusion time of 5 min was chosen for the standard procedure. However, the spectra of samples of the tea infusion taken at the same times showed a continuous increase in absorbance at all wavelengths over the whole period.

The corresponding pH values exhibited a trend opposite to that of the scum mass, with a fall from 7.66 in the hard water to 6.62 after 5 min infusion. This

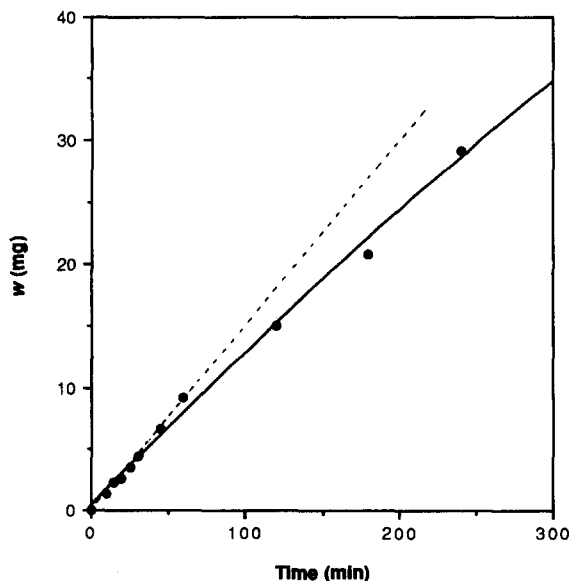


Fig. 2. Variation of the mass of tea scum with time during 4 h at 80°C, after a standard infusion. The dotted line represents a continuation of the linear plot in Fig. 1.

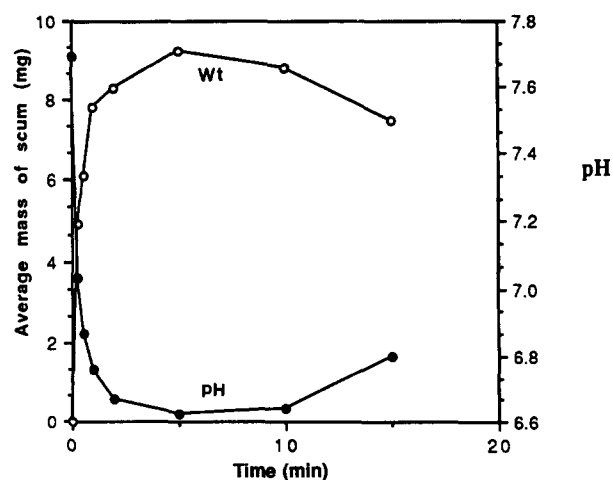


Fig. 3. Effect on scum mass after 60 min standing time (left-hand scale) and on the pH of the infusion (right-hand scale) of variations in the time over which one teabag was infused, all at 80°C.

decrease will have been caused by the acid nature of the tea polyphenolics.

Variation of surface area and volume

To test whether scum formation was a purely surface phenomenon, the normal vessel of diameter 10.28 cm (83.0 cm² area) was replaced by one of 9.0 cm diameter (63.6 cm² area). The rate of scum formation then decreased by 19%, close to the 23% decrease in surface area. In another experiment with the normal vessel, one-quarter of the tea brew was removed at the end of the 5 min infusion time. After standing for 60 min the mass of scum was the same as usual, showing that the volume of the infusion was immaterial. Thus, the scum not only accumulates at the surface but its formation depends upon the exposed surface area and is independent of the volume of the tea solution.

Variation of the mass of tea

Surprisingly, the amount of scum decreased sharply as more teabags were used. With five teabags no scum at all was observed even after a standing time of 4 h (Table 1). As shown below, these results can be largely explained by the accompanying drop in pH.

Table 1. Effect of varying the number of teabags on the pH and on the mass of scum (w) formed after 5 min infusion and 60 min standing at 80°C

No. of teabags	pH (at 80°C)	w (mg)	
		Experimental	Eqn (1)
1	6.62	9.4	9.3
2	6.26	5.6	5.1
5	5.67	0.0	1.9

Variation in the type of tea

A set of the Typhoo tea bags kept for 4 years yielded the same amount of scum as bags from the same batch tested shortly after delivery from the factory. Ageing is therefore not a significant factor in scum formation.

In a standard experiment carried out with teabags of Typhoo decaffeinated tea, the mass of scum did not differ significantly from the usual value. Caffeine is therefore not implicated in the formation of tea scum. Since caffeine is a vital component of tea cream (Rutter & Stainsby, 1975), we may conclude that the formation of tea scum and tea cream are quite separate phenomena.

To find out to what extent green tea also formed scum, Chun Mee leaf was sieved to provide the 250–850 μm fraction which corresponds in size to 98% of the Typhoo blend. The same mass of this leaf as in the teabags, 3.17 g, was then placed in a 75 μm mesh container (see below) and infused for 5 min. In contrast to black tea, little foam was produced and the brew colour was pale. After 60 min standing time the mass of scum was 7.6 mg, *c.* 20% less than for black tea.

Effect of the container material

Two sets of experiments were carried out to see if the teabag material affected scum formation. First, nine empty teabags were put into the 80°C hard water as well as one filled teabag. The pH after 5 min infusion was 6.65 and the mass of scum after 60 min standing time was 8.4 mg. The same result was obtained if nine empty teabags were stirred in the 80°C water for 5 min and then removed before adding a filled teabag and proceeding in the usual manner. The paper from a single teabag therefore slightly reduced the amount of scum, by *c.* 0.1 mg.

In a second series of experiments, 3.17 g of bulk Typhoo tea was placed in fine mesh bags made from 45 μm and 75 μm stainless-steel mesh (Endecotts Ltd). The pH values were very similar, 6.65 and 6.67 respectively, and the resulting weights of scum agreed, within experimental error, with that normally obtained with a teabag. These findings reinforce the conclusion reached above.

Effect of water composition

According to an analysis provided by Thames Water plc, the London mains tap water employed had a mean total hardness of 272 ppm as CaCO_3 and a mean alkalinity as CaCO_3 of 205 ppm. These figures correspond to a mean calcium ion concentration of 2.72 mM ($M = \text{mol dm}^{-3}$) and to a mean bicarbonate concentration of 4.1 mM (Dearborn Chemicals, Widnes, Cheshire, UK). We found its average pH at 80°C to be 7.66. A series of standard experiments was now carried out with one Typhoo teabag infused at 80°C in distilled water (pH 5.71) containing either no added electrolyte or

various combinations of CaCl_2 , MgCl_2 and NaHCO_3 (all BDH AnalaR). The results are summarised in Table 2.

An infusion in distilled water formed a brightly coloured solution with no foam and no scum even after 2 h standing at 80°C. Contrary to popular belief, the presence of calcium (or magnesium) ions on their own produced no scum either. Nor did bicarbonate ions on their own, although the tea liquor was darker than before and the colour seemed to increase more rapidly, in agreement with a study on the rate and appearance of theaflavin extraction in more alkaline media (Spiro *et al.*, 1987). There was much foam at the beginning of the infusion period but it disappeared after 3 min during stirring. Here, and in the hard water experiments, the foam will have been the result of CO_2 evolution from the reaction between bicarbonate ions and the acidic polyphenolic tea components, with the gas bubbles stabilised at the surface by various surface active tea compounds. In the experimental procedure used (Spiro & Jaganyi, 1994), the surface foam was always removed before the start of the standing period.

Scum was produced, however, whenever both calcium and bicarbonate ions were present. Foam then appeared during the infusion period and a white precipitate of CaCO_3 floated to the surface. These phenomena had also been observed in the London tap water experiments. The pH of the infusion and the mass of scum formed in the latter agreed well with the corresponding values in synthetic hard water which contained 2.72 mM calcium and 5.44 mM bicarbonate (pH 7.34). Doubling the concentration of both ions increased the mass of scum by 50%. However, only half the normal amount of scum was produced when calcium ions were replaced by the same concentration of magnesium ions. No similar experiment was possible with strontium ions because a white precipitate formed as soon as the strontium and bicarbonate solutions were mixed.

The necessity for the presence of calcium (or magnesium) ions had now been established, but it was not yet clear whether bicarbonate ions were required in their own right or just because they raised the pH to an appropriate level. Trials were therefore carried out with

Table 2. Effect the addition of various electrolytes to distilled water on the mass of scum (w) formed after 5 min infusion of one teabag and 60 min standing, all at 80°C

[Ca^{2+}] (mM)	[HCO_3^-] (mM)	w (mg)	Infusion pH	Infusion colour	Foam produced
0	0	0	5.01	Bright	None
2.6	0	0	4.67	Bright	None
0	4.1	0	6.66	Dark	Much
2.72	5.44	8.9	6.60	Dark	Much
5.44	5.44	10.9	6.45	Dark	Much
5.44	10.9	13.4	6.59	Deep	Much
2.6 ^a	0	0	4.66	Bright	None
2.7 ^a	5.44	4.5	6.67	Dark	Much

^a [Mg^{2+}] instead of [Ca^{2+}].

distilled water containing 2.7 mM CaCl₂ and selected buffer substances. Precipitation of calcium phosphate ruled out the use of phosphate salts but addition of 0.1 M boric acid and 0.004 M NaOH gave a suitable medium of pH 7.28 at 80°C. In this solution less than half the calcium ions will have been complexed by borate (Smith & Martell, 1989). The resulting tea infusion had a pH of 6.76; no appreciable foam or white precipitate were observed during infusion and only 1.3 mg scum formed after 4 h (0.3 mg h⁻¹). This experiment showed that bicarbonate ions are also essential for the formation of tea scum since almost no scum was produced even though the infusion possessed the appropriate pH and calcium ion concentration.

Variation of pH

In order to ascertain the effect of pH on scum formation, a series of standard 5 min infusions was carried out in London mains water at 80°C. After the removal of the teabag the pH was 6.66 and the foam and any scum on the surface were removed. A known small amount of 11.8 M HCl (Fisons AR) was added, the solution stirred thoroughly, the surface again cleared, the timer started and the pH of the solution measured. The resulting masses of scum formed after 1 h are listed in Table 3.

The mass of scum is seen to fall dramatically as the acidity of the solution increases. The results are represented quite well by the following empirical relation:

$$\log_{10}(w/\text{mg h}^{-1}) = 0.714\text{pH} - 3.758 \quad (1)$$

as demonstrated in the last column of Table 3. This equation between scum mass and pH also explains why much less scum was formed when two teabags were used, as shown in Table 1. The fact that no scum at all appeared with five teabags may be due to a second factor, additional complexation between calcium ions and certain tea polyphenols which will have been present in high concentration.

Effect of complexing agents

It seemed likely that scum formation could be reduced or avoided altogether by adding substances to complex calcium ions. Addition of EDTA (ethylene diamine tetra-acetic acid disodium salt, BDH AnalaR) to form a 0.001 M solution after the normal 5 min infusion period changed the pH to 6.03 and no scum formed after 60 min. In a related experiment, the EDTA was added to the London mains water to begin with, giving a pH of 6.45 at 80°C. The resulting infusion was bright with a pH of 6.26. Although foam was produced as usual, no white precipitate was observed. Only 0.7 mg of scum formed after 1 h.

Similar experiments were carried out with the addition of citric acid (BDH AnalaR) to form a 0.001 M solution. When the acid was added after the infusion period the pH dropped to 5.43 and again no scum was

Table 3. Effect on pH and scum mass (*w*) of adding various electrolytes to tea infusions in London mains water at 80°C

Addition of	pH	<i>w</i> (mg/h)	
		Experimental	Eq (1)
—	6.66	9.5	9.9
0.05 ml HCl	6.44	7.1	6.9
0.10 ml HCl	6.14	4.1	4.2
0.15 ml HCl	5.78	1.7	2.3
0.20 ml HCl	5.25	1.1	1.0
0.25 ml HCl	4.65	0.0	0.4
0.001 M EDTA	6.03	0.0	3.5
0.001 M citric acid	5.43	0.0	1.3

produced after 1 h. When citric acid was added to the mains water before infusion to give pH 6.06, the resulting tea liquor was bright with a pH of 5.82. No white precipitate was seen and only a negligible amount of foam. After 4 h just 1.2 mg scum had formed, or 0.3 mg h⁻¹. It is interesting that Pendlington (1977) has patented the impregnation of teabag paper with citric acid or other soluble acids at a level of 0.03–5 mg per cm² (2–25 mg per teabag) in order to improve the colour of tea brewed in hard water. This invention will also have had the beneficial effect of reducing the amount of tea scum but nothing about this was mentioned in the patent.

The effects on scum mass of the pH changes brought about by the additions of EDTA and citric acid can be calculated by eqn (1). These figures are listed in the last column of Table 3 and can be compared with the experimental results in the third column. It is apparent that EDTA and citric acid reduce the amount of scum much more than would be expected from the pH changes alone. This can clearly be ascribed to complexation of calcium ions by the substances added (Smith & Martell, 1989).

Finally, an experiment was carried out to see what effect the addition of a complexing agent would have on scum which was already growing. After a standard infusion, scum was allowed to form for 1 h and then sufficient EDTA solution was carefully added and briefly stirred magnetically to make the solution 0.001 M. The pH then fell to 6.19. After 1 h the scum was scooped off and found to weigh 9.2 mg. The expected weight of scum was 9.1 mg after the first hour plus 4.6 mg during the second hour at pH 6.19 according to eqn (1), a total of 13.7 mg. Thus the addition of EDTA had completely stopped the further growth of the tea scum. It is relevant to add that particles of CaCO₃ were found to dissolve rapidly in a 0.001 M solution of EDTA in water at 80°C. Calcium carbonate is therefore needed not only as an initiator of scum formation but is also an essential accompaniment to its growth. However, sprinkling CaCO₃ powder (BDH AnalaR) on to the surface of distilled water had no effect: the powder sank to the bottom and, after a teabag had been infused in the water, no scum formed even after 4 h.

Variation in standing temperature

For these experiments the teabag was infused at 80°C for 5 min as usual, and then taken out. To study scum formation at 70°C, the vessel was removed and cooled by inserting a copper coil through which cold tap water flowed. It took 2 min for the temperature to drop to 70°C. The vessel was then transferred to a water bath set at 70°C, the foam and scum on the surface were scooped off, and the timer started. The pH at 70°C was 6.65 and 6.35 mg scum formed after 60 min.

To measure scum formation at 90°C, the vessel was removed from the 80°C thermostat bath after the infusion period and the solution heated to 90°C on a hotplate. It was then transferred to a 90°C thermostat bath. Foam and scum on the surface were removed before starting the timer. The pH was again 6.65 and 12.2 mg scum were produced after 60 min.

The activation energy, E , of scum formation can be calculated from the Arrhenius equation:

$$w = A \exp(-E/RT) \quad (2)$$

where A is the pre-exponential constant, R the gas constant and T the absolute temperature. A plot of $\ln w$ against $1/T$ gave a straight line with the following equation:

$$\ln(w/\text{mg h}^{-1}) = 13.60 - 4030/T \quad (3)$$

The activation energy is therefore 34 kJ mol⁻¹. This is smaller than the activation energy of 46 kJ mol⁻¹ for the crystallisation of calcite from calcium bicarbonate solutions (Cassford *et al.*, 1983).

This value can be compared with the activation energy expected if the rate-determining step for scum formation were diffusion of some relevant species through the solution towards the surface. For such a situation the rate would be proportional to the diffusion coefficient D of the species, and we shall assume that its value would be given by the Stokes–Einstein equation:

$$D = kT/6\pi\eta r \quad (4)$$

where k is the Boltzmann constant, η the viscosity of the water and r the effective radius of the diffusing species. If r is independent of temperature the rate constant will then be proportional to T/η . Insertion of the numerical values (Robinson & Stokes, 1959) leads to an activation energy of 15.8 kJ mol⁻¹. This is less than half the experimental value and shows that the reaction is not simply diffusion-controlled.

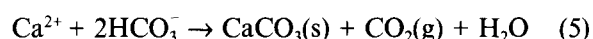
Effect of stirring

Whether diffusion is the rate-determining step was tested more directly by stirring the tea solution just below the surface. For this purpose a vertical glass stirrer ending in two twisted glass blades with an overall diameter of 5 cm was employed. At the beginning of the standing time this stirrer was lowered into the tea solution such that the blades were 2 cm below

the surface and it was rotated at known speeds with a Citenco FHP motor. After 1 h the mass of scum was 9.4 mg with a rotation speed of 42 rev min⁻¹ and 8.5 mg with 120 rev min⁻¹. These values agree, within the normal experimental uncertainties, with the value of 9.1 mg obtained without stirring and also show no trend with increasing stirring speed. This confirms that the rate-determining step of scum formation is not diffusion of material from the solution towards the scum layer.

Effect of boiling the water

In normal tea brewing, the water is briefly boiled before adding the tea leaves. Boiling is also a process for softening temporary hard water since it drives off carbon dioxide and removes calcium ions in the form of CaCO₃:



In these experiments the London mains water was boiled for a predetermined time on a hotplate and then rapidly cooled to 80°C as described above. The teabag was inserted and standard infusion and standing times were employed. When the water was boiled for 1 s the pH fell to 6.67 and 7.9 mg scum formed in 1 h, 13% less than usual. Boiling for 10 min reduced the pH to 6.53 and the mass of scum to 6.2 mg. Thus boiling the water beforehand significantly decreases the amount of scum formed but does not eliminate it altogether as claimed by Kooijmans (1940) and Schurer (1960). Since application of eqn (1) shows that only part of these decreases can be explained by the slight fall in pH, most of the effect must be attributed to the lowering of calcium and bicarbonate concentrations via the reaction in eqn (5) (*cf.* Table 2).

The amount of scum was decreased further by removing the CaCO₃ precipitated by boiling. For this experiment a litre of tap water was boiled for 10 min and then filtered hot through a sinter-glass frit. The appropriate volume of this water was equilibrated at 80°C and a standard scum experiment performed. Only 4.5 mg of scum was produced after 60 min, significantly less than with boiled water which had not been filtered.

The influence of dissolved and atmospheric gases

Several experiments were carried out to discover the effects, if any, of oxygen and carbon dioxide on the development of scum. The results are summarised in Table 4.

The first of these experiments was designed to remove O₂ but not CO₂. Distilled water was de-aerated by bubbling nitrogen for 8 h, and calcium chloride and sodium bicarbonate were then added to make the solution 2.72 mM in Ca²⁺ and 5.44 mM in HCO₃⁻. This allowed carbon dioxide to form by the reaction in eqn (5) on heating the liquid to 80°C. Nitrogen was passed over the solution during the standard infusion and

Table 4. Effect of dissolved and atmospheric gases on the formation of scum in London mains water at 80°C

Water degassed with	Gas passed over surface	w (mg)	Infusion pH	Infusion colour	Foam produced
N ₂ ^a	N ₂	4.0	6.88	Normal	Normal
N ₂	N ₂	2.0	6.51 ^b	Dull	Minimal
O ₂	O ₂	11.9	6.46 ^b	Very dark	Minimal
—	N ₂	5.2	6.67	Normal	Normal
—	O ₂	22.3	6.66	Normal	Normal
—	Air	17.2	6.70	Normal	Normal

^a Using synthetic hard water made from de-aerated distilled water to which CaCl₂ (2.72 mM) and NaHCO₃ (5.44 mM) had been added.

^b The pH of the tap water after passage of gas was 7.94 compared with 7.69 normally.

standing times. The resulting scum mass was 4.0 mg, less than half the normal amount.

Even less scum, only 2.0 mg, was found in the next experiment with London mains water which had been saturated with nitrogen both at room temperature and while it was being heated to 80°C, with nitrogen then being passed over the surface during the infusion and standing periods. Both oxygen and CO₂ had thereby been excluded. When this experiment was repeated with O₂ in place of N₂, a larger amount of scum than normal was produced, 11.9 instead of 9.1 mg.

Finally, three further sets of runs were performed in which the London mains water was not degassed but either N₂, O₂ or air was continuously passed over the surface during the 1 h standing time. As can be seen from Table 4, the mass of scum was much less than normal under N₂ (5.2 mg) but had greatly increased under an atmosphere of O₂ (22.3 mg) and even under a flow of air (17.2 mg). The formation of tea scum therefore involves oxidation by oxygen from the gas phase. However, the high activation energy of 34 kJ mol⁻¹ rules out diffusion of oxygen towards the surface as the rate-determining step. The above experiments, as well as those in the previous section, also indicate a role for CO₂ in the aqueous phase, perhaps indirectly in connection with the equilibrium in eqn (5).

Parallelism with other tea processes

The oxidation of tea solubles by atmospheric oxygen to form tea scum resembles the oxidation by oxygen of the tea flavanols in green tea to form the black tea theaflavin and thearubigins (Roberts, 1962; Graham, 1983). The former process is mediated by calcium carbonate while the latter is catalysed by tea leaf polyphenolase. Both processes, moreover, occur most readily in an alkaline environment where the various tea polyphenolics are partially ionised. In hard water they can then form ion pairs or complexes with calcium ions which may well be the first step in the production of scum.

Another parallelism emerges from a recent study by Robertson and Hall (1989). These workers reported

that theaflavin undergoes oxidation to thearubigins not only during fermentation and storage periods but also during high-temperature infusion where the breakdown of theaflavin passes through a maximum after about 10 min (at 100°C?). This behaviour is similar to scum formation which, as seen in Fig. 3, passes through a maximum after *c.* 5 min infusion at 80°C.

Arguing by analogy, and in view of the fact that green tea also produces scum, it is tempting to suggest that tea scum is made up of theaflavin and thearubigin components. However, green tea scum was not analysed. The chemical microanalysis of black tea scum (Spiro & Jaganyi, 1994) led to an empirical formula in which the O:C ratio was much higher than in theaflavin and thearubigin compounds of similar molecular weight which points to a still greater degree of oxidation, particularly in the early stages. To discover the true organic precursors of scum one would need to study the scum produced from separate hot hard-water infusions of specific flavanols, theaflavin, theaflavin mono- and di-gallates and, were it possible, thearubigins.

SUMMARY

Tea scum is a complex substance consisting of high molecular weight organic matter together with islands of calcium carbonate (Spiro & Jaganyi, 1994). The present study has shown that the latter is crucial to the development of scum, for none forms in the absence of free calcium (or magnesium) and bicarbonate ions. The rate of the reaction depends on a fractional power of the calcium ion concentration and inversely on the 0.7th power of the hydrogen ion concentration. Scum formation increases with the concentration of oxygen above the surface and thus involves aerial oxidation of tea solubles which are probably the polyphenolic components such as catechins, theaflavin and thearubigins. The overall kinetics are therefore complex and involve one or more chemical steps. Mass transport in the solution or from the gas phase is not a rate-determining step.

The fairly large activation energy of 34 kJ mol⁻¹ explains why only small amounts of scum are normally observed on a cup of tea which is cooling rapidly, and why more scum is often found inside a teapot whose temperature stays relatively high.

ACKNOWLEDGEMENTS

The authors thank Dexter Nonwovens for a Research Bursary awarded to D.J. and Premier Brands Ltd for supplying the Typhoo tea.

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