

Copyright is owned by the Author of the thesis. Permission is given for a copy to be downloaded by an individual for the purpose of research and private study only. The thesis may not be reproduced elsewhere without the permission of the Author.

Rendering complex colour inside 3D printed foods

**A thesis presented in partial fulfilment of the requirements for the degree of
Doctor of Philosophy in Food Technology**

at Massey University, Palmerston North, New Zealand

Sandra Kim

2015

Abstract

Three-dimensional (3D) printing refers to a group of digitally controlled, additive manufacturing technologies increasingly used to fabricate customised objects from a range of possible materials, including food ingredients, using a digital image file representing the object. A novel variation on 3D *food* printing is being developed to customise the appearance of foods with an embedded 3D colour image by the selective blending of primary colorants. This capability is beyond what is needed usually for the coloration of bulk, single food matrices.

In this thesis, non-food techniques of colorimetric matching (used in computer match prediction) and colour gamut mapping (from cross-media colour reproduction), were investigated as potential methods for dye recipe computation by the new 3D colour food printer. The aim was to develop a model for transforming image RGB data to dye recipe data, taking into account the variable effects of food properties. The two techniques were applied to the problem of matching a set of standard tile colours using a set of primary colorants in model food substrates. Kubelka-Munk (K-M) blending equations underlying both techniques were developed for blends of Brilliant Blue, Ponceau 4R (red) and Tartrazine (yellow) food dyes when added to a microwave-baked cake and to four variants of a wheat starch gel. Validation of the model for the cake blends was shown by $\Delta E^*_{ab,10}$ differences between computed and measured $L^*_{10}a^*_{10}b^*_{10}$ colours falling within range of a visually acceptable match (three $\Delta E^*_{ab,10}$ units). For some of the gel blends, the $\Delta E^*_{ab,10}$ differences reached five units.

Dye recipes computed by a modified colorimetric matching algorithm to match target tile colours with cake colours at times called for negative quantities, or totals that exceeded the legal limits for foods containing dyes, indicating that the target colour was outside the range (gamut) of the cake-dye system. In these recipes, individual negative dye quantities were increased to zero, and totals scaled back to within the legal limit, retaining relative dye proportions. This resulted in close differences between tile and cake before scaling (with computed $\Delta E^*_{ab,10}$ values of less than three units for as many as eight of the twelve target colours) becoming much larger

after scaling (up to 39 $\Delta E^*_{ab,10}$ units), though visual inspection of the colour pairs suggested that the matches might be closer.

The gamut of perceived colours from a coloured food is not only constrained by legal restrictions on dye addition, but dependent on the properties of the food itself, such as its background colour (seldom white) and the light-scattering effects of surface texture. Compared with colour images, foods are likely to have a more limited colour gamut, the size of which is expected to vary with changes in formulation and processing. Gamut mapping techniques were used to investigate the extent to which the target tile colours themselves needed to be scaled back before matching solutions and corresponding dye recipes could be computed. Using four samples of the gel that differed only in their level of (artificial) browning, including white, the impact of browning on the colour gamut was determined. Using the cake, solutions from gamut mapping were compared with those from colorimetric matching.

A gamut of discrete colours is treated as a continuous volume in colour space. In the absence of a published gamut calculation for coloured foods, a technique was developed to compute a mesh of points on the colour gamut boundary. Boundary colours were computed using dye blends not exceeding the legal limit, and spaced such that $\Delta E^*_{ab,10}$ did not exceed three units. This method was applied to the white (non-textured) gel containing dye blends, to generate a 'base' gamut. The absorption behaviour of each dye was found to be largely consistent among the white and browned gels which enabled quick computation of colour gamuts for the brown gels by substituting the absorption spectrum of a brown gel for that of the white in the K-M equation. The colour gamut was found to decrease in size and to shift position with increased gel browning. The dye blends that were used to compute the colour gamut boundary for the whitened gel were combined with the absorption spectrum for the cake to compute the gamut boundary for the cake colours. All colour gamuts were specific to the standard D65 illuminant and 10 degree standard observer.

In the investigation of the effects of browning, colour gamut mapping began with the initial replacement of each tile colour with a colour from the white gel gamut. All colours were replaced gradually by a darker, and often less chromatic, colour, as the level of browning in the gel was increased. As a result of the reduction in gamut size with increased gel browning, the difference between tile colours and their replacement targets in each of the reduced gamuts was smaller for tile colours having ‘brown’ characteristics (such as Orange, Red and Yellow) than they were for blue-, pink- and green- coloured tiles. Larger increases in total dye quantity with increased gel browning were needed for the latter group of colours than for the former. For most colours an increase in the relative proportion of the darkest dye in the recipe was also needed. The actual dye quantities computed for each replacement colour depended on the availability of mesh points in the region of colour space in which the tile was located.

Colour gamut mapping required a heavier computational load than the colorimetric matching technique to provide solutions for tile colours in the cake-dye gamut. Although not always giving solutions in the same angular region of colour space as the tile colours, colorimetric matching was able to produce similar $\Delta E^*_{ab,10}$ differences between tile colour and best cake match as did colour gamut mapping, for not necessarily more or less total dye.

Two forms of a generalised algorithm are proposed for the computation of dye recipes by the 3D colour food printer. One algorithm is modelled on a workflow for cross-media colour reproduction. A series of transformations that account progressively for the effect of individual characteristics of the food printing substrate on the achievable gamut from dye blends is incorporated into the main series of transformations that transcribes RGB image data to dye recipe data. In the other algorithm, modelled on colorimetric matching, it is the progressive effect of each individual characteristic on the light-absorption characteristics of the un-dyed food printing substrate that is accounted for, and incorporated into the main matching workflow.

Preface

This thesis is written in the style of (but not formally complying with) a submission based on publications, as described in the Handbook for Doctoral Study, Doctoral Research Committee, Massey University, Version 7, January 2011; each chapter following the Literature Review and preceding the final Overall Discussion is formatted as an extended research paper. Within research Chapters Four, Five and Six, the methods and results are described and discussed for each stage of the work, rather than in separate Methods and Results sections for the entire body of work. All research chapters (Chapters Four to Seven) have their own Introduction and General Discussion sections. Appendices follow each of these chapters. Due to the format of the thesis some of the detail that is covered in the Literature Review is repeated in the remaining chapters, which are presented in chronological order.

A shorter form of Chapter Four was published as an original research paper in the Journal of Food Science (Kim *et al.*, 2012). A copy is not included here. It is intended to use other chapters as the basis for future manuscripts.

Unless stated otherwise, the figures in this thesis are the work of the author.

Acknowledgments

I have many to thank for their contributions, large and small, to making this thesis a reality. Firstly, I thank my supervisors, Professor Richard Archer and Professor Matt Golding, for their guidance, vision and enthusiasm, and in particular for their patience and understanding during the drawn-out latter stages.

The 3D food printing research which is the subject of this thesis is part of the wider Technofoods project of the Riddet Institute, and was supported by a Riddet PhD Scholarship. On a day-to-day basis my 'home' has been at the Institute of Food, Nutrition and Human Health (IFNHH) at Massey University. There, I have been able to draw upon the generous support of numerous members of staff - academic, technical and administrative. IFNHH staff members that I must single out for mention are Warwick Johnson and Sue Nicholson, Managers of the Food Product Development and Postharvest Laboratories respectively, and Dr Richard Love – Warwick for being so willing and able to help, with a laugh thrown in for good measure, Sue for access to her lab's spectrophotometer, and for training in its use, and Richard for helping a first-time user of MATLAB[®].

I am grateful for the company and friendship of my office-mates, past and present especially Elham and Hayley – the challenges that come with being a PhD student have been much easier to bear because of you. And this from Teresa Wegrzyn, my fellow 'foundation' PhD student in the 3D food printing project: "at least we know what the other person's on about – even if no-one else does". Thank you Teresa.

Outside of the office, two individuals have given to this thesis in their own special way: Don Ravine with his photography (Figure 4.8 in Chapter Four) and little Poppy with her quiet presence beside me during many late nights writing.

To Duncan, I am so very lucky to have you by my side in life and love. I could not have done this without you.

Table of Contents

Abstract	i
Preface.....	iv
Acknowledgments	v
Chapter One: Introduction to 3D colour food printing	1
Chapter Two: Literature Review, Part 1 – Principles of colour and coloration	4
2.1. Scope of this Review	4
2.2. Colour and Appearance	5
2.2.1. The stimulus: light and object	5
2.2.2. Colour perception: the observer	14
2.3. Describing colour and the basis of colour measurement.....	16
2.3.1. Colour coordinates and colour coordinate systems (colour spaces)	16
2.3.2. Visual colour description – The Munsell and NCS Colour Systems	20
2.3.3. Instrumental colour measurement - the CIE systems	22
2.3.4. Colour differences: formulae.....	29
2.4. Producing colour: colour blending and colour reproduction	35
2.4.1. Additive mixing systems: colours based on the RGB primaries	35
2.4.2. Subtractive mixing systems: colours based on dyes and pigments.....	35
2.4.3. Mathematical models for colour blending	37
2.4.4. Instrumental colour matching based on the K-M blending equations.....	41
2.5. Colour management.....	43
2.5.1. Overview and definitions.....	43
2.5.2. Device characterisation (profiling)	45
2.5.3. Colour gamut mapping	46
2.6. Colour in food	52
2.6.1. The role of colour in food: Colour is a part of total appearance	52
2.6.2. Describing and measuring food colour.....	54
2.6.3. The addition of colour to food: reasons colorants are added.....	63
2.6.4. Synthetic food colorants.....	63
2.6.5. Formulation of dye blends for food applications	72
2.6.6. Factors affecting coloration in food materials: examples	75
2.7. The effects of changes in substrate on the modelling and prediction of colour and appearance	79
2.7.1. Alternative model for print, based on Principal Components Analysis (PCA).....	82

Chapter Three: Literature Review, Part 2 – Customising foods using 3D colour printing	83
3.1. The concept of customisation.....	83
3.2. 3D printing technologies.....	84
3.2.1. Fused deposition modelling, or extrusion deposition	84
3.2.2. Granular materials binding	84
3.2.3. Laminated object manufacturing.....	85
3.2.4. Advantages and applications	85
3.3. Customisation of food.....	86
3.4. 3D food printing	87
3.4.1. 3D food printing: inputs and outputs	88
3.5. Baked goods as model substrates for the development of printable food pastes.....	95
3.5.1. Description of cake characteristics and the role of ingredients and processing	96
3.5.2. The effect of formulation and processing changes.....	101
3.5.3. Alternative (rapid) baking technologies: Jet impingement oven technology and microwave cooking	110
3.5.4. A ‘case study’: low-fat cake doughnuts	112
3.5.5. Predictive studies: modeling the development of structure and colour in food systems of relevance to baking, and to 3D colour food printing.....	113
3.6. Controlled 3D coloration in food and non-food matrices	119
3.6.1. Summary and conclusions	123
3.7. Conclusions from this Review	128
3.7.1. Required experimental approach	129
3.7.2. Consumer aspects	132
3.8. Research aims and objectives of thesis	132
Chapter Four: Food coloration using computer colour matching	134
4.1. Introduction	134
4.2. Materials and General Methods	137
4.2.1. Dyes.....	137
4.2.2. Microwave cake substrate	137
4.2.3. Colour targets	139
4.2.4. Colour measurement	139
4.2.5. Calculation of colour coordinates from measured reflectance	140
4.2.6. Calculation of colour differences	141
4.3. Development and validation of the dye blending model	143

4.3.1. Background.....	143
4.3.2. Derivation of unit absorption coefficients for dyes in the cake substrate.....	145
4.3.3. Validation of unit absorption coefficients by investigation of dye blends.....	150
4.4. Matching cake colours to tile colours using the colorimetric method.....	155
4.4.1. Background.....	155
4.4.2. Methods.....	156
4.4.3. Results and Discussion.....	160
4.5. General Discussion: Visual implications.....	169
4.5.1. Interpretation of total colour differences between tile and cake colours.....	169
4.5.2. Differences in lightness, hue and chroma.....	171
4.5.3. The appropriateness of using tile colours as matching targets.....	172
4.6. Conclusion.....	172
4.7. Appendix.....	173
Chapter Five: Colour gamut boundary computation allowing for the effects of browning.....	181
5.1. Introduction.....	181
5.2. Materials and General Methods.....	184
5.2.1. Model system.....	184
5.2.2. Colour measurement.....	188
5.3. Development and validation of dye blending models.....	189
5.3.1. Derivation of dye unit absorption coefficients and substrate absorption coefficients.....	189
5.3.2. Validation of dye unit absorption coefficients (1).....	193
5.3.3. Validation of dye unit absorption coefficients (2): Comparison of dye unit absorption spectra derived from the different substrates.....	197
5.4. Colour gamut boundary computation.....	199
5.4.1. Colour gamut boundary for the 'White' gel containing primary dye blends.....	199
5.4.2. Effect of substrate browning.....	204
5.4.3. Visualisation of the gamut boundaries in colour.....	206
5.5. General Discussion.....	208
5.5.1. Strength of colour predictions and implications for gamut shape.....	208
5.5.2. Gamut boundary sampling technique: density of sampling points.....	212
5.6. Conclusions.....	213
5.7. Appendix.....	214
5.7.1. Comparison of gel and microwave-baked cake substrate colours without added primary dyes.....	214

5.7.2. Unit absorption spectra at λ_{\max} for each primary dye in the Brown1 and Brown3 gel substrates.....	214
5.7.3. MATLAB [®] code for the gamut boundary colour figures (Figure 5.8).	215
Chapter Six: Food coloration using colour gamut mapping.....	217
6.1. Introduction	217
6.2. Materials and General Methods	219
6.2.1. Selection of appropriate methods	219
6.2.2. General procedure	220
6.3. Gamut boundaries at planes of constant hue angle.....	221
6.3.1. Computation	221
6.3.2. Results	223
6.4. General colour gamut mapping procedures.....	226
6.4.1. Lightness compression.....	226
6.4.2. Chroma compression	226
6.4.3. Clipping	227
6.5. Colour gamut mapping to a ‘standard’ colour gamut	227
6.5.1. Lightness compression.....	227
6.5.2. Chroma compression	228
6.5.3. Clipping	229
6.6. Colour gamut mapping: effects of browning.....	229
6.6.1. Lightness compression.....	229
6.6.2. Chroma compression	229
6.6.3. Clipping	230
6.7. Results from colour gamut mapping.....	231
6.7.1. Description of the changes in C* and L* across the mapped colours	233
6.8. Computing dye recipes for mapped colours.....	240
6.8.1. Recipe correction method	240
6.8.2. Effect of browning on computed dye quantities.....	243
6.9. Dye recipes for mapped colours: Results and discussion	244
6.9.1. Extent of recipe correction	244
6.9.2. Relationships between gel browning and computed dye quantities	246
6.9.3. Models fitted to the relationships between gel browning and computed dye quantities for mapped colours.....	251
6.10. General Discussion.....	253

6.10.1. Observed trends in the lightness, chroma and dye recipes of the mapped colours	254
6.10.2. Effects of gamut boundary detail and choice of mapping algorithm	255
6.10.3. Implications for 3D colour food printing	257
6.11. Appendix.....	258
Chapter Seven: Comparing colorimetric matching and colour gamut mapping.....	276
7.1. Introduction.....	276
7.2. Materials and Methods	278
7.2.1. Boundary for the entire cake gamut	278
7.2.2. Gamut boundaries at planes of constant hue angle	279
7.2.3. Gamut mapping	279
7.2.4. Dye quantities for mapped colours	279
7.2.5. Comparison of outputs from colour gamut mapping and from colorimetric matching	279
7.3. Results and Discussion.....	280
7.3.1. Gamut boundaries.....	280
7.3.2. Gamut mapping.....	287
7.3.3. Dye quantities for mapped colours	290
7.3.4. Differences between outputs from colorimetric matching and colour gamut mapping, in providing the best equivalents of the original tile colours.....	291
7.4. Conclusions.....	310
7.5. Appendix.....	311
Chapter Eight: Overall Discussion.....	322
8.1. Recap: The background to the thesis	322
8.2. Research objectives and summary of main findings	323
8.3. Translation to 3D colour food printing	328
8.3.1. Transformations models for use with colour gamut mapping.....	329
8.3.2. Transformations models for use with colorimetric matching	338
8.3.3. Summary: transformations models.....	340
8.3.4. Alternative printing technologies	341
8.3.5. Single-coloured model substrates vs. multi-coloured printed foods	342
8.3.6. Quality of reproduction	343
References.....	345

