

**DRYING CHARACTERISTICS AND QUALITY
ATTRIBUTES OF BLACK TEA UNDER
SUPERHEATED STEAM DRYING**

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SUPERHEATED STEAM DRYING**

by

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LIST OF SYMBOLS

h	Heat transfer coefficient
D_{eff}	Effective moisture diffusivity (m^2/s)
E_{ni}	Energy input (kJ)
E_{no}	Energy output (kJ)
W	Work (kJ)
n_{en}	Energy efficiency
N	Drying rate ($\text{g H}_2\text{O}/\text{m}^2\cdot\text{min}$)
W_{w}	Weight of the water (g)
W_{d}	Weight of the dry matter (g)
M_{s}	Mass of dried tea (g)
A	Area of drying (m^2)
X	Moisture content (g)
t	Drying time (min)
M_{o}	Initial moisture content (g)
M_{e}	Equilibrium moisture content (g)
a	Dimensionless empirical constant
n	Dimensionless empirical constant
k	Drying rate coefficient
l	Thickness of dried tea leave (m)
n	Number of terms
D_{o}	Diffusivity at an infinite temperature (m^2/s)
E_{a}	Activation energy (kJ/mol)
T	Temperature ($^{\circ}\text{C}/\text{K}$)
R	Gas constant ($\text{J}/\text{mol}\cdot\text{K}$)

C	Measured value of colour variables
C_o	Initial value of colour
R^2	Coefficient of determination
R^2_{adj}	Adjusted coefficient of determination
R^2_{pre}	Predicted coefficient of determination
X^2	Reduced chi-square
$MR_{exp,i}$	Experimental moisture ratio
$MR_{pre,i}$	Predicted moisture ratio
N	Total number of observations
Abs	Absorbance reading
DM	Percentage of dry matter (%)
x_i	Coded value of independent variable
z_i	Actual value of independent variable
D	Desirability function
m_{ev}	Experimental value
m_{pv}	Predicted value
γ	Measured response
β_o	Constant coefficient of the model
β_1 and β_2	Linear model
β_{11} and β_{12}	Quadratic model
β_{12}	Interaction model
\bar{C}_p	Specific heat capacity (J/kg°C)
P	Power of the dryer (W)
m_{ew}	Mass of the evaporated water (kg)
λ_{wp}	Latent heat of the dried tea leaves (J/kg)
λ_w	Latent heat of free water (J/kg)

LIST OF ABBREVIATIONS

ANOVA	Analysis of variance
C	Catechin
CCD	Central composite design
CTC	Crush, Tear and Curl
CV	Coefficient of variance
DPPH	2,2- diphenyl-picrylhydrazyl
EC	Epicatechin
ECG	Epicatechin gallate
ECP	Endless chain dryer
EGC	Epigallocatechin
EGCG	Epigallocatechin gallate
FRAP	Ferric reducing antioxidant power
GA-ANN	Genetic algometric neural network
GC	Gallo catechin
GRG	Generalised Reduced Gradient
HAD	Hot air drying
HPLC	High performance liquid chromatography
IBMK	Isomethyl butyl ketone
MC	Moisture content
MR	Moisture ratio
PDA	Photodiode array detector
PE	Percentage error
PRESS	Predicted error sum of squares
QDA	Qualitative descriptive analysis

RMSE	Root mean square error
RSM	Response surface methodology
RSS	Residual sum of squares
SD	Standard deviation
SSD	Superheated steam dryer
SSIB	Superheated steam impingement blanching
TF	Theaflavin
TF2	Theaflavin-3-gallate
TF2B	Theaflavin-3'-gallate
TF3	Theaflavin-3,3,-digallate
TPC	Total phenolic content
TPTZ	2,4,6-Tri (2-pyridyl)-s-triazine
TR	Thearubigin
USM	Universiti Sains Malaysia

CIRI-CIRI PENGERINGAN DAN KUALITI TEH HITAM MENGGUNAKAN PENGERINGAN STIM PANAS LAMPAU

ABSTRAK

Keseimbangan rasa di dalam teh hitam dapat dicapai melalui proses pengeringan. Walau bagaimanapun, pengering konvensional yang digunakan dalam industri teh kini menggunakan tenaga haba yang tinggi daripada kayu api sebagai sumber bahan api utama. Hal ini menimbulkan kebimbangan terhadap kesannya kepada alam sekitar. Pengering stim panas lampau (SSD) merupakan teknologi pengeringan terkini yang menggunakan stim sebagai medium pemanasan. Kesan penggunaan SSD dalam proses pengeringan teh hitam masih terbatas dari segi pemahaman ciri dan mekanisme pengeringan, sifat kualiti, dan penggunaan tenaga. Oleh itu, kajian ini dijalankan untuk mengkaji keupayaan SSD sebagai pilihan praktikal untuk mengeringkan teh. Pertama, kesan julat suhu (120–200°C) SSD terhadap ciri pengeringan teh hitam telah dikaji. Berdasarkan pemerhatian, peningkatan suhu pengeringan dapat meningkatkan kadar pengeringan dan hal ini membantu mengurangkan tempoh masa pengeringan. Model baharu yang menggabungkan Model Page serta Model Henderson and Pabis juga berjaya menepati data eksperimen pengeringan. SSD juga dapat mengurangkan tenaga pengaktifan (16.17 kJ/mol) 25 kali lebih rendah berbanding kajian yang dilaporkan menggunakan ketuhar konvensional (406.02 kJ/mol). Hasil kajian ini membuktikan bahawa SSD boleh memulakan pemindahan jisim kelembapan dalam pengeringan teh lebih cepat daripada ketuhar konvensional. Kedua, kaedah gerak balas permukaan (RSM) dengan reka bentuk komposit pusat (CCD Pemusat-Muka) telah digunakan untuk mengenalpasti keadaan pengeringan optimum (140–180°C dan 20–25 minit)

berdasarkan jumlah kandungan fenolik, aktiviti antioksidan, flavonoid individu dan asid amino dan sifat sensori teh hitam (warna teh, bau, rasa, dan kepahitan teh). Analisis statistik menunjukkan bahawa kebanyakan gerak balas memberikan kesan positif terhadap suhu pengeringan, dan negatif terhadap masa pengeringan. Keadaan pengeringan optimum boleh laksana SSD telah diperolehi pada suhu 161°C selama 22 minit dengan menggunakan model ramalan. Pada bahagian terakhir kajian ini, perbandingan antara keadaan optimum pengeringan SSD dengan pengering plat pinggir (ECP) (kini digunakan di ladang BOH) telah dibandingkan. Kajian mendapati bahawa teh kering SSD mengandungi lebih tinggi kandungan fenolik asid, aktiviti antioksidan, theaflavin, thearubigins, dan asid galik berbanding teh kering ECP sebanyak 92%, 96%, 50%, 86% dan 96%. Para panelis sensori juga lebih menggemari SSD teh kering berbanding teh ECP dalam semua sifat sensori. Dari aspek penggunaan tenaga spesifik, SSD menggunakan 55% tenaga lebih rendah (2.6 kWh/kg) daripada pengering ECP (5.82 kWh/kg). Secara keseluruhannya, kajian ini berjaya membuktikan SSD sebagai pilihan yang lebih sesuai dalam pengeringan teh kerana dapat menghasilkan kualiti teh hitam yang lebih baik dengan penggunaan tenaga yang lebih rendah.

DRYING CHARACTERISTICS AND QUALITY ATTRIBUTES OF BLACK TEA UNDER SUPERHEATED STEAM DRYING

ABSTRACT

The balanced flavour of black tea can be achieved from its tea leaves drying process. However, conventional dryers used in the current tea industries consume a large amount of thermal energy that comes from firewoods as their primary source of heat. This eventually raises concerns on the environmental impacts of firewood usage. Superheated steam dryer (SSD) is an advanced drying technology that utilises steam as the heating medium. The impact of SSD on the black tea drying process is still limited in terms of understanding the drying characteristics and mechanisms, quality attributes, and energy consumption. Therefore, this study aims to explore the capability of SSD as a practical option for tea drying. Firstly, the effect of SSD temperature range (120–200°C) on black tea drying characteristics was investigated. It was observed that an increase in drying temperature caused a higher drying rate that helps to shorten the drying period. The drying experimental data were also successfully fitted to a new model (Page and Henderson and Pabis model) which helps to predict the behaviour of black tea drying. SSD was able to lessen the activation energy (16.17 kJ/mol) by 25 times lower than the previous study which used the conventional oven (406.02 kJ/mol). This proves that SSD can initiate the mass transfer of moisture in tea drying faster than the conventional oven. Secondly, response surface methodology (RSM) with central composite design (CCD-Face centered) was employed to optimize the selected drying condition (140–180°C and 20–25 min) based on the phenolic content, antioxidant activities, individual flavonoids and amino acid, and sensory attributes (infusion colour, aroma, flavour and mouthfeel). Statistical analysis showed that most responses

were positively affected by SSD drying temperature and negatively affected by SSD drying time ($P<0.05$). The feasible optimal condition of SSD was achieved using predictive model at 161°C and 22 min. The final part of this study compared the optimum drying conditions of SSD with endless chain plate (ECP) dryer (currently used in BOH factory). Result revealed that SSD dried tea had significantly higher phenolic acid, antioxidant activities, theaflavins, thearubigins, and gallic acid than ECP dried tea by 92%, 96%, 50%, 86% and 96%, respectively. The sensory panelists also preferred SSD dried tea over ECP dried tea in all sensory attributes. In term of specific energy consumption, SSD consumed 55% energy lower (2.6 kWh/kg) than the ECP dryer (5.82 kWh/kg). Overall, this study had successfully proved SSD as a suitable option for tea drying because it could produce a better quality of black tea with a lower energy consumption.

CHAPTER 1

INTRODUCTION

1.1 Background

Tea is one of the beverage that all ages can drink worldwide. Today, tea is claimed to be the second most-consumed beverage after water and can be found in almost 80% of all households across the globe (Goggi, 2018). The numerous health benefits of tea have gained recognition and interest from both consumers and scientists alike (Yang *et al.*, 2015). Therefore, particular attention is given towards tea processing as it directly influences the quality of tea (Botheju *et al.*, 2011). The balanced flavour can be achieved during drying because moisture is removed, thus halting fermentation and increasing tea shelf life (Roshanak *et al.*, 2016).

Drying has been categorised as an energy demanding process that consumes an average of 20–25% national energy in Germany and Denmark and 10–15% in most developing countries (Sehrawat *et al.*, 2016). According to Kumar and Pou (2016), most hot air tea dryers adopted conventional endless chain pressure (ECP) or fluid bed (FB) dryer that consumed a large amount of thermal energy that varies from 4 – 10.4 kWh/kg of made tea. The main challenges faced here are: (1) the low drying rate at the initial period of drying leads to an intensive energy consumption; (2) unwanted particle in a moving part of dryer can come into contact with tea leaves; and (3) exposing the tea leaves to oxidation during drying will cause the final black tea product to lose its quality (Temple & Van Boxtel, 2000). Particularly in main tea producer countries such as Kenya, Sri Lanka, India, and Malaysia, conventional tea dryer utilises firewood, coal, natural gas, and furnace oil as their primary source of fuel (Saikia *et al.*, 2013).

Consequently, this raises concerns on the environmental impacts in terms of deforestation and air pollution (the release of carbon dioxide after burning) (Okoth, 1991).

This results in a search for an alternative and innovative dryer which its inherent advantages are better than a conventional dryer. Superheated steam drying (SSD) is one of the advanced drying technology. SSD offers many advantages such as a high energy efficiency due to higher drying rate than hot air, a low net energy consumption, the elimination or reduction of odour emission, a lower oxygen environment (no fire and explosion hazards), and a final product with lower nutrient loss and lesser impact on quality (colour, texture, shrinkage, and rehydration) when compared to a conventional dried product (Mujumdar, 2014). Numerous studies have been reported on superheated steam drying of foodstuffs which include tortilla chips (Li *et al.*, 1999), potatoes (Tang & Cenkowski, 2000), shrimp (Prachayawarakorn *et al.*, 2002), noodles (Markowski *et al.*, 2003), and cocoa beans (Zzaman *et al.*, 2014). However, information on the effects of superheated steam drying of black tea is scarce in published literature.

Besides, numerous studies on different tea drying techniques such as microwave (Chan *et al.*, 2009), vacuum (Kishore *et al.*, 2014), and freeze-dryer (Roshanak *et al.*, 2016) had been conducted to investigate their effect on the quality of black tea. Their results highlighted the influence of drying time and temperature towards qualitative characteristics of polyphenols content, its antioxidant ability, and sensory attributes of black tea (Teshome *et al.*, 2013). Despite their promising findings, each of these drying technologies face several drawbacks which hinder their ability to improve the black tea quality further. For instance, hot air dryer has the advantages of being simple and having a low operational cost but can damage the nutritional value of tea leaves (Roshanak *et al.*, 2016; Qu *et al.*, 2019), while vacuum and freeze-drying were able to produce high

quality of black tea with more phenolic compounds and high aroma tea powder (Xiangyang *et al.*, 2010; Ye *et al.*, 2014).

Nonetheless, vacuum and freeze-drying are not suitable as their duration is considerably longer than other drying methods (Mujumdar, 2014). A freeze dryer also requires a high operational and maintenance cost that is 4 to 8 times higher than hot air drying (Cieurzyńska & Lenart, 2011). Whilst microwave drying may provide many benefits such as shortening the time of drying, but the food material itself may undergo interior burning due to excessive heating (Jeni *et al.*, 2010).

1.2 Problem statements

The conventional tea drying process is known to be an energy-intensive operation that consumes up to 71.7% energy within the whole tea production (Munasinghe *et al.*, 2017). This energy accounts for both thermal and electrical energies consumption to reduce the moisture content of tea leaves from 70% to 3% (Khanali *et al.*, 2017). Hence, energy crisis and environmental impacts of the conventional dryer has been addressed in many research works (Ni & Hang, 1992; Gesimba *et al.*, 2005; Azapagic *et al.*, 2016; Bandara *et al.*, 2016). It is necessary for new technology such as superheated steam to be adopted in black tea drying for efficient consumption of energy and environmental friendly operation.

SSD is an advanced drying technology; yet, not many study has investigated its drying capabilities and efficiency, especially on tea leaves. Tea leaves contain many heat sensitive compounds such as flavonoids and amino acids which need to be preserved for the best quality of black tea. Unlike conventional tea drying, SSD does not involve direct heating and absence in oxygen in the dryer that could be a suitable option to improve tea quality. Hence, this work aims to investigate the feasibility of

superheated steam as a dryer for black tea drying based on the drying characteristics and mechanisms, total polyphenol content, antioxidant abilities, individual flavonoids and amino acid, sensory attributes and its energy efficiency. This work might help in improving the product quality and energy consumption in the development and production of black tea.

1.3 Objectives

This study aims to explore the ability of SSD to minimise the quality degradation of black tea leaves and improve the energy consumption in the tea industry. In order to achieve the aim, experimental works were carried out based on the following objectives:

- 1) To conduct drying kinetic analysis based on moisture content and colour changes during superheated steam drying of tea leaves at 120–200°C drying temperature for 40 minutes.
- 2) To determine the optimum drying conditions (temperature and time) for SSD based on black tea total phenolic content, antioxidant activity, individuals flavonoids and amino acid, and qualitative data of sensory attributes by using response surface methodology.
- 3) To compare all chemical components and sensory acceptability of tea prepared by SSD operated at optimum condition and ECP drying condition.
- 4) To compare the energy consumption and efficiency between SSD operated at optimum condition and ECP drying condition.

1.4 Scope of work and thesis outlines

The study of this research focused mainly on the application of superheated steam dryer in drying black tea leaves and its impact on drying characteristic and tea quality attributes. The whole dissertation is arranged as follows:

Chapter 1 begins by addressing the overall context of the study that includes research background, problem statements, and proposed solution. It also states each objective that this study has been focusing on.

Chapter 2 describes information on tea, its economic importance, and the whole tea production. The elaborate literature review was also included in tea quality assessment based on their bioactive constituents. The different effect on the quality parameter of tea by different dryers is extensively compared and discussed. Previous works on SSD are also presented which includes its principles, characteristics, and effect on product food quality.

Chapter 3 is a published work on the kinetic study of the moisture and colour changes of SSD dried black tea leaves. It details on the drying characteristic of SSD at certain range of temperature and time. The mathematical models that correlated well with both experimental data of moisture and colour changes are also reported. Additionally, effective diffusivity, activation energy and reaction rate constant were also reported.

Chapter 4 is a published paper of work on the optimization of drying conditions for black tea leaves. It reports on the experimental design of SSD drying conditions and optimization by using response surface methodology (RSM). The experimental design constructed consider all the desirable target for both chemical components and sensory

descriptive scores which later decides the optimum condition. The validity of the optimum condition is also discussed.

Chapter 5 is a comparison study between the optimum drying conditions of SSD and an endless chain plate tea dryer. The two dryers had contrasted in terms of the quality of polyphenol content, antioxidant activity, individuals flavonoids, and amino acid, sensory acceptability, and energy consumption.

Finally, Chapter 6 concludes all the key findings of the whole research work. Recommendations were also listed for further research.

The whole experimental design for this study is shown in Figure 1.1.

Phase 1: Kinetic study and quality attributes

Drying tea leaves
 Time: 5-40 mins (5 mins interval)
 Temperature: 120°C - 200°C (20°C interval)

1. Moisture
 - a) Drying Curve
 - b) Drying Kinetic (Thin Layer Drying model)
 - c) Diffusion Coefficient and Activation Energy
2. Colour changes
 - a) Degradation Kinetic

Phase 2: Optimization of tea drying condition using SSD

Response Surface Methodology (RSM)
 Variables (140-160°C and 20-25 min)

Extraction
 Distilled water extraction

Responses		
1. Phenolic Content Analysis	2. Individual Flavonoids and Amino Acid	3. Sensory Evaluation (QDA)
a) Total Phenol Content	a) Total Catechin	a) Liquor colour
b) DPPH Radical Scavenging Activity	b) Total Theaflavin	b) Aroma
c) FRAP	c) Total Thearubigin	c) Mouthfeel
	d) Gallic Acid	d) Flavour
	e) Caffeine	
	f) L-theanine	

Phase 3: Comparison study between SSD and ECP dryer

Optimum condition of SSD (obtained from RSM)
 vs.
 ECP dryer drying condition

Quality Attributes			
1. Phenolic Content Analysis	2. Individual Flavonoids and Amino Acid	3. Sensory Evaluation (Hedonic)	4. Energy consumption
a) Total Phenol Content	a) Total Catechin	a) Liquor colour	a) Energy efficiency
b) DPPH Radical Scavenging Activity	b) Total Theaflavin	b) Aroma	
c) FRAP	c) Total Thearubigin	c) Mouthfeel	
	d) Gallic Acid	d) Flavour	
	e) Caffeine	e) Overall acceptability	
	f) L-theanine		

Figure 1.1 Overall experimental design of the study

CHAPTER 2

LITERATURE REVIEW

2.1 Origin and botanical varieties of tea

The ancient legend of discovering tea happened as far back as 3000 BC in China (Mair & Hoh, 2009; Sigley, 2015). The fascinated incident happened when Emperor Shen Nong accidentally tested the infusion of leaves from a tea plant that fell into his cup of boiling water. He was intrigued with its flavour and aroma, and thus declared the practice of tea drinking (Weisburger, 1997; Sigley, 2015). This custom soon spread to India and Japan, then to European countries which later resulted in four million acres of land devoted to tea cultivation in over 30 countries (Ferrara *et al.*, 2001).

Cultivation of tea originates from plants belonging to the family of *Theaceae* that comes in two varieties: *Camellia sinensis*: var. *sinensis* (China tea) and var. *assamica* (Assam tea) (Hasimoto & Simura, 1978; Yang *et al.*, 2016). China tea cultivar can be characterised by its smaller leaves, 5 – 12 cm long (Figure 2.1-a), which grow in big shrubs that are up to 6 m in height. This particular plant, which can either be a shrub or an evergreen tree, can be grown in the tropical and subtropical climates with adequate annual rainfall up to 1,200 mm (Mukhopadhyay & Mondal, 2017). It also requires shade and mildly acidic soil for optimum cultivation (Silva, 2007; Hajiboland, 2017). Tounekti *et al.* (2013) also stated the Assam tea has very large leaves, 15 – 20 cm long (Figure 2.1-b), which come from a small tree that grows up to 15 m with a straight trunk and robust branches (Takeo, 1992; Tounekti *et al.*, 2013). As in Malaysia, major tea manufacturers cultivated Assam tea variety (Figure 2.1-b) which most suited with Malaysia's climate (Krishnan, 2000)

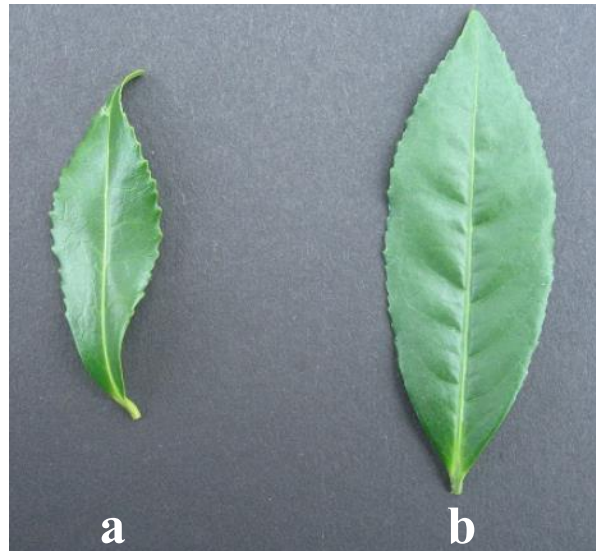


Figure 2.1 Different sizes of tea leaves based on two different varieties; a) *Camellia sinensis*: var. *sinensis* (China tea) and b) var. *assamica* (Assam tea). Source from Nursery (2017)

2.2 Economic importance of the tea industry

Since drinking tea has been around for thousands of years, the tea industry has become the second largest global beverage packaged with 286 billion litres consumed in the year 2018 (Figure 2.2). According to the report by Bolton (2019b), the global tea market was estimated to be worth about USD 12.62 billion in 2019 to 2023 with an annual growth rate of 5% between 2019 to 2023. This significant growth is mainly driven by a tremendous leap in world consumption and demand. Bolton (2019b) also stated that tea consumption is expected to increase by nearly 5% which is 30 – 36 billion litres in volume. These opportunity has brought up to 13 million employment around the world (Chang, 2015).

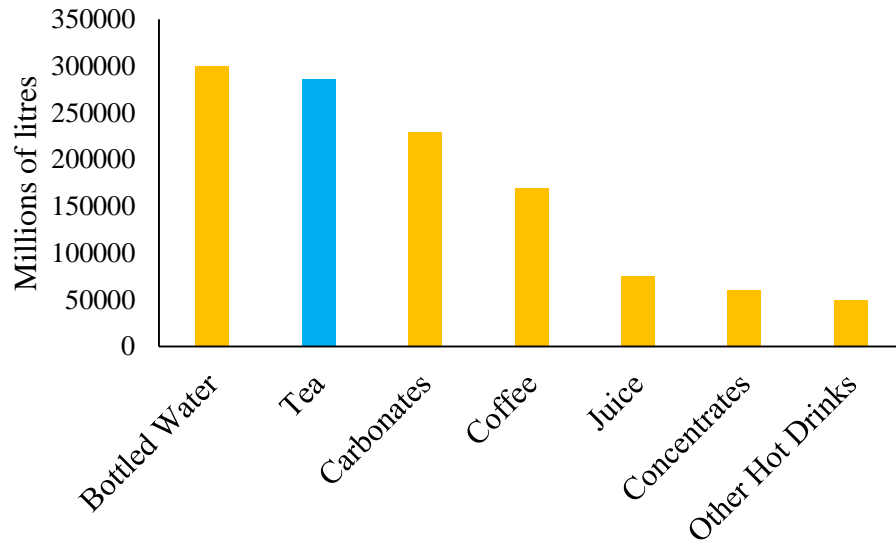


Figure 2.2 Bar chart presents the global beverage market in total volume in 2018 (Bolton, 2019b)

The main producers of tea are China, India, Kenya, and Sri Lanka, accounting for nearly 76% of the total world production (Soni *et al.*, 2015). China, as the largest producer, can produce an output of 2.8 million tonnes that contribute to more than 35% of the world's tea production (Bolton, 2019a). Compared to these giant tea producers, Malaysia is not well known as the main tea grower in the global market. However, Malaysia is doing its best to contribute to the global tea production stage.

By 2011, Malaysia produced up to 0.45% of the world's total tea production that leads to an approximate \$100 million in revenue per year (Tang, 2015). Overcoming to get recognition, Malaysia tea's manufacturer aims to venture a mass market brand to the foreign market such as the United States. Tang (2015) also reported from his interview with Caroline Russell, CEO of BOH Plantations, that they believed the unique and very distinct flavours tea from Malaysia can make the connoisseur market to be interested. Hence, she also hopes to obtain an increase of sale up to 30% from the overseas market in the future.

The biggest tea company in Malaysia belongs to BOH that also owns the largest tea plantations with 1,200 hectares out of a total of 2,533 hectares of land property

dedicated to tea cultivation, which mark its dominant household name in Malaysia (Figure 2.3) (Verwoerd & Facenda, 2013).



Figure 2.3 Overview of BOH Plantation, Cameron Highlands.

These properties are all located in Cameron Highlands, Pahang. Its ideal condition has helped in Malaysia's tea production in having robust, flavourful, and aromatic tea (Verwoerd & Facenda, 2013). The ideal location above 1,500 metres of sea level, constant temperature of 15–22°C, an abundant rainfall, fertile and acidic soil prompted J. A. Ruselle to create Malaysia's first highland tea plantation back in 1929. Since then, 70% of the country's total tea production has been from BOH manufacture alone, which is being reserved for the local consumers as well as being exported internationally.

2.3 Black tea and it's chemical composition

Freshly plucked leaves of tea plant can be processed to give five major types of tea; black tea are from fully fermented leaves, oolong tea are semi-fermented, pu-erh tea are post fermented tea, and green and white tea are from unfermented tea leaves (Figure 2.4) (Carlioni *et al.*, 2013).



Figure 2.4 Selection of tea types; white, green, oolong, black and pu-erh tea retrieved from Valley (2018)

Even with the various types of tea, consumers of black tea usually far outweigh other types of tea consumers. Statically, 80% of people mostly consume black tea which is the most popular drink around Europe, North America, and North Africa, whereby Asians mostly prefer green and white tea (Beresniak *et al.*, 2012; Waugh *et al.*, 2017). Particularly in Malaysia, different ethnic prefers different types of tea. As mentioned by Kong (2013), chairman of the Tea Trade Association, most Malays will go for strong teas (black tea), while Chinese consumers tend to favour light tea such as green, white or oolong tea. Nonetheless, a mixture of strong black tea with condensed milk known as ‘Teh Tarik’ (pulled tea) has become the most popular request among all ethnics in Malaysia (Foon, 2015).

Black tea refers to a fully fermented or oxidised form of tea. The oxidation manages the chemical compositions to undergo changes and result in great taste, aroma, colour, nutritional, and desired biological properties in black tea (Gramza-Michałowska, 2016; Duan *et al.*, 2018). Those changes have promoted black tea consumption with significant health benefits such as cancer prevention (Yang *et al.*, 2008), anti-inflammation (Nurlaily *et al.*, 2012), and reduced occurrence of cardiovascular diseases and cerebral ischemic damages (Pinto, 2013). These health-promoting aspects are mainly attributed to the bioactive compounds in black tea.

2.3.1 Bioactive constituents in black tea

Fresh tea leaves differ greatly in composition after being oxidised into black tea. Specifically, the main constituents of fresh tea leaves that belong to the polyphenol group (25% to 35% on a dry weight basis) will undergo a process of oxidation and polymerisation (Li *et al.*, 2013). Major polyphenols in tea consist of six main groups; flavanols, hydroxyl-4-flavanols, anthocyanins, flavones, flavonols, and phenolic acids (Mukhtar & Ahmad, 2000). Among these flavanols, (+)-catechins (C) is predominant followed by (-)-epicatechin (EC), (-)-epicatechin gallate (ECG), (-)-epigallocatechin (EGC), (-)-epigallocatechin gallate (EGCG), and (+)-gallocatechin (GC) as depicted in Figure 2.5 (Liang *et al.*, 2003; Łuczaj & Skrzydlewska, 2005).

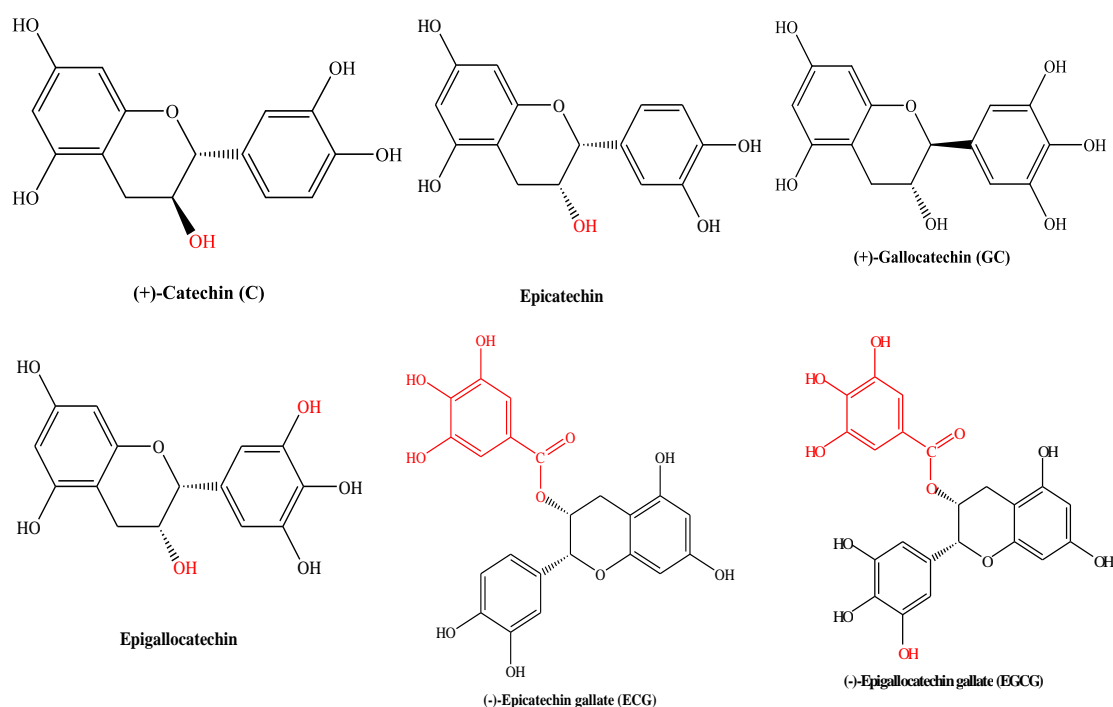


Figure 2.5 Major flavanol components in tea leaves

In black tea, these flavanols are firstly oxidised to quinones and the reaction is catalysed by polyphenol oxidase which needs to work in direct contact with atmospheric

oxygen, monophenol mono-oxidase (tyrosinase), and an *o*-diphenol: O₂ oxidoreductase that naturally exists in fresh tea leaves (Łuczaj & Skrzydlewska, 2005). Afterward, a polymerisation step occurs resulting in catechin quinones undergoing nucleophilic addition that forms gallocatechin quinones. Subsequent oxidation, carbon dioxide elimination, and rearrangement cause the complete synthesis of theaflavin (Figure 2.6) (Tanaka *et al.*, 2010).

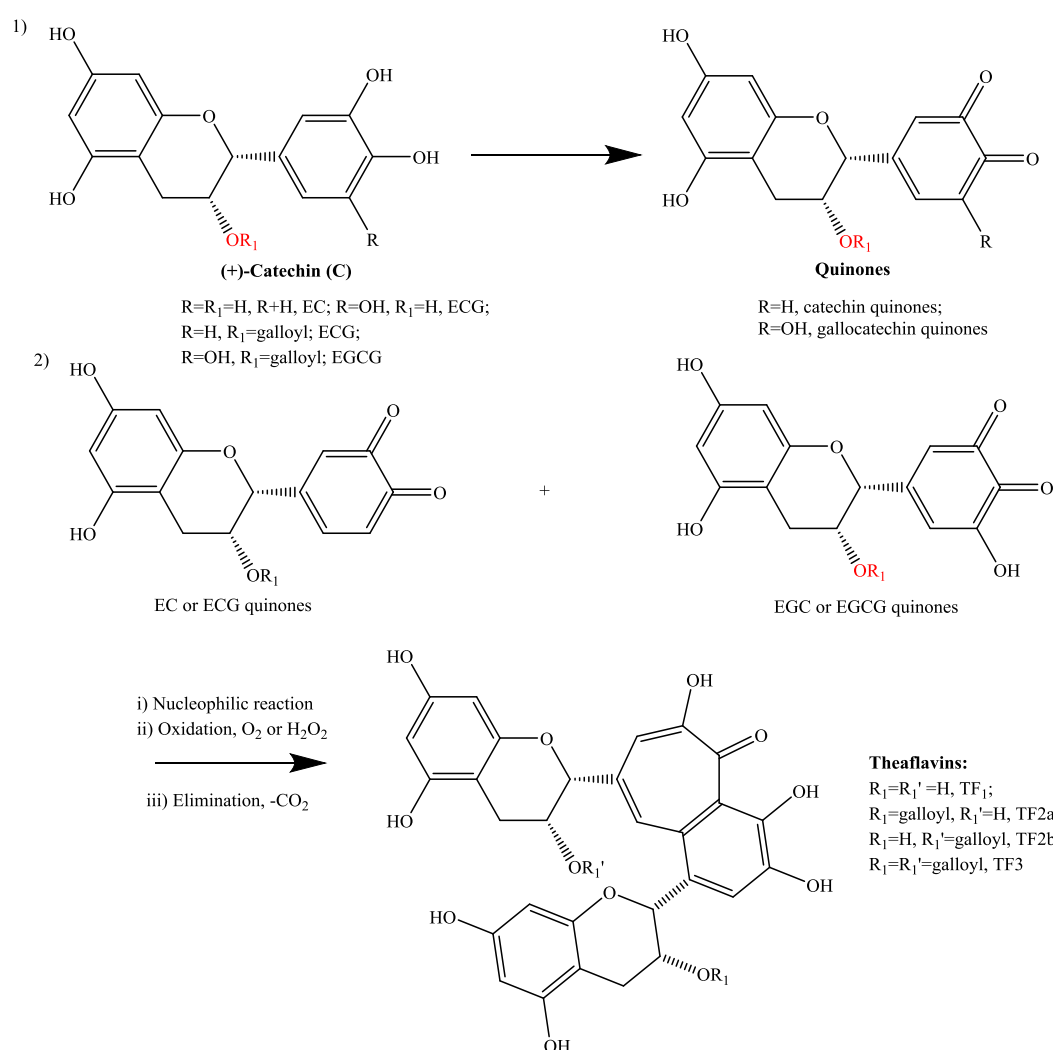


Figure 2.6 Enzymatic catalysed process of the theaflavin formation

This reaction results in the ensuing black tea to have an approximate of catechins (10–12%), theaflavins (3–6%), thearubigins (12–18%), flavonols (6–8%), phenolic acids and depsides (10–12%), amino acids (13–15%), methylxanthines (8–11%),

carbohydrates (15%), proteins (1%), mineral matter (10%), and volatiles (<0.1%) (Łuczaj & Skrzydlewska, 2005).

2.3.1(a) Flavanols group in black tea

Polyphenol groups are known to have more than one benzene ring with each containing hydroxyl group (-OH) (Riemersma *et al.*, 2001; Hodgson & Croft, 2010). The most abundant polyphenols in black tea are the flavanols, a subclass of flavonoids. These flavanols are mostly responsible for the health benefits offered from drinking tea. Hodgson and Croft (2010) further added that a single cup of tea (2 g tea in hot water) could provide up to 150 – 200 mg of flavanols. These flavanols include catechins (monomers of EC, GC, ECG, EGC, and EGCG), theaflavin, and thearubigin (Peterson *et al.*, 2005). However, the presence of catechin in black tea largely decreases after the oxidation process. This colourless soluble compounds transform into orange-brown colour compounds: theaflavin and thearubigin (Khan & Mukhtar, 2007).

Theaflavin, which consists of the bicyclic ring with tropolone structure (Figure 2.7), is a product from catechin oxidation.

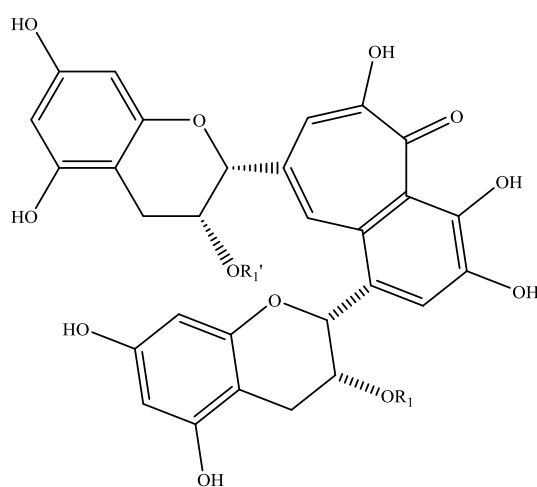


Figure 2.7 Theaflavin structure

This yellowish, reddish compound is known as the colour contributor to black tea (Rana & Singh, 2012). It can also be produced in four major isomers which are theaflavins (TF1), theaflavin-3-gallate (TF2), theaflavin-3'-gallate (TF2B), and theaflavin-3,3'-digallate (TF3) (Li *et al.*, 2013). Theaflavins can also impart astringent taste, a mouth-coating sensation known as 'briskness' of black tea infusion (Scharbert, Jezussek, *et al.*, 2004). As a dimer of catechin, theaflavins are able to possess strong antioxidant activity that is able to help in the prevention of DNA oxidative damage (Feng *et al.*, 2002), anti-cancer (Koňariková *et al.*, 2015), anti-inflammatory (Aneja *et al.*, 2004), anti-microbial (Friedman *et al.*, 2006), anti-mutagenicity (Gupta *et al.*, 2002), and anti-viral effect (He, 2017).

The major flavanols in black tea are thearubigins, with 60–70% from the total of phenolic content in dry tea (Tounekti *et al.*, 2013). Thearubigin is also known as a polymeric and oligomeric compound that is formed after theaflavins was further oxidised; this may include nucleophilic reacting with oxidised flavanols (Yassin, 2014). This oligomer is also defined by its brown colour and gives out the specific taste in black tea. It is similar to theaflavins with an added major contribution towards health as an anti-initiating agent against cancer cells (Butt *et al.*, 2014). However, the exact structure of thearubigins is still unknown despite the attempt and efforts done by many researchers (Wang *et al.*, 2018).

2.3.1(b) The amino acids, alkaloids, and phenolic acids

Amino acids comprise an important component in black tea and are present in the range of 1–4% of the leaves total dry mass (Hilal, 2017). Various types of amino acid can be found in tea such as theanine, glutamic acid, aspartic acid, serine, glutamine, alanine, and arginine. Among all, L-theanine is the majority (>70%) in black tea (Chen

et al., 2009). This glutamic acid γ -ethyl amide (L-theanine) (Figure 2.8) is very important in tea because of its flavour characteristic that can give out a sweet umami taste in tea (Kausar *et al.*, 2013). Theanine not only gives out the delicate taste but produces a calm sensation effect and is able to regulate blood pressure in human being (Thippeswamy *et al.*, 2006).

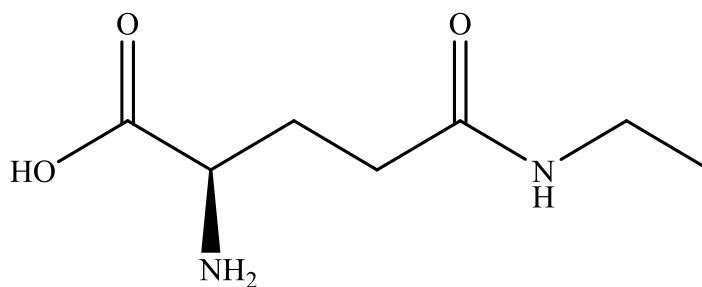


Figure 2.8 L-theanine structure

Black tea chemical composition consist of minor group of alkaloids that include caffeine, theobromine, and theophylline (Figure 2.9). Caffeine being the richest (3–4%) among alkaloids in black tea even after fermentation (Rahim *et al.*, 2014), and is known for its pharmacological properties that can help stimulate the central nervous system, heart, and respiratory system of the human body (Yang *et al.*, 2007). Nonetheless, unlike other components in black tea, high doses of caffeine have some negative effects such as being a diuretic, causes anxiety, increases blood pressure, and triggers sleeplessness (Yang *et al.*, 2007).

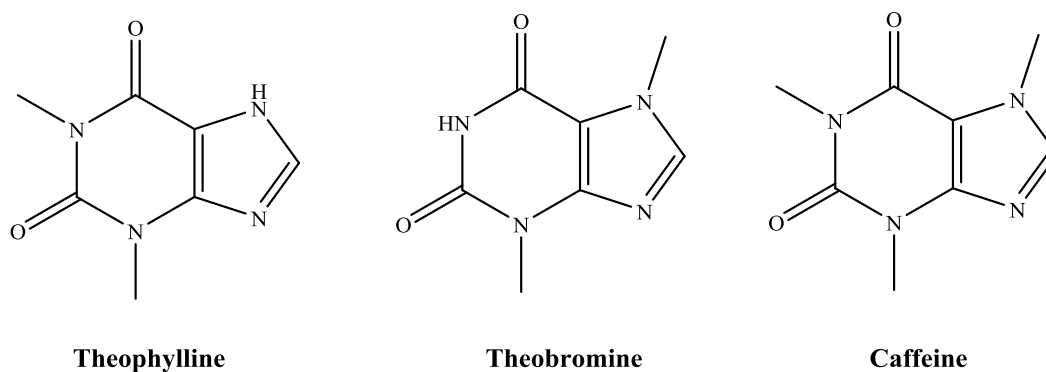


Figure 2.9 Alkaloids structure

Another key component present in black tea includes gallic acid which possesses an antioxidant property (Karori *et al.*, 2007), anti-fungal (Koech *et al.*, 2013), and anti-carcinogenic effect (Perva-Uzunalić *et al.*, 2006) (Figure 2.10). The rest of the black tea components consist of vitamins (B, C, and E), minerals and trace elements (calcium, magnesium, manganese, copper, zinc, selenium, and potassium), triterpenoids, carbohydrate, and fat (Perva-Uzunalić *et al.*, 2006).

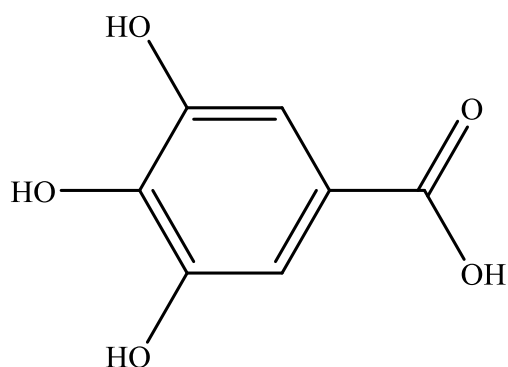


Figure 2.10 Gallic acid structure

2.3.2 Black tea processing

“All ‘true’ tea comes from the same plant, called Camellia sinensis. Whether the tea becomes white, green, oolong or black, it depends on how the leaves are processed and oxidised.” (“True tea,” 2010)

The statement above gives a clear spectrum about tea processing. Essentially, the processing is an important factor to determine the type of tea being made. There are five distinct stages to manufacture black tea, which are plucking, withering, rolling, oxidation, and drying. The later 4 stages are presented in Figure 2.11.



Figure 2.11 Main processing steps in producing black tea which involve withering, CTC or Orthodox method, oxidation, and drying

In the first step, the tea leaves are normally being harvested by hand plucking. Selection between the unopened bud to the top three leaves at the bud depends on the type of tea being created. In the second stage, plucked tea leaves are laid out to wilt and wither for several hours (as in Figure 2.11). This stage is a partial drying where the water content in tea leaves is reduced up 50 to 70% (Turkmen *et al.*, 2009). This serves as physical preparation for the next phase; as moisture content decreases, it allows the catechin to penetrate out from the cytoplasm and interact with the oxidase in the cell membrane of the leaves (Kilel *et al.*, 2018). The leaves can be withered under sunlight or even in dark or hot room. Rolling is the third step that involves the crushing of withered tea leaves into smaller sizes (Vargas & Vecchiatti, 2016). Black tea is usually processed in either orthodox or Crush, Tear, and Curl (CTC) processing (as in Figure 2.11). The softened or withered tea leaves are macerated fully to produce smaller size

tea in the CTC method, while in orthodox processing, tea leaves are pressed, rolled, and twisted by the Orthodox roller or Rotarvane to get a bigger size tea (Kilel *et al.*, 2018). These techniques help in breaking the cell walls of the tea leaf to make the enzymes and essential oil ready for the oxidation process (Kumar *et al.*, 2013).

The fourth step is the most crucial step of all the processes. In this step, the leaves are laid out to rest for several hours for the oxidation process to take place. At this moment, good contact with air is a must to ensure that the oxidation process occurs (Vargas & Vecchietti, 2016). This oxygen interacts with the exposed enzyme and turn the leaves to reddish brown colour as well as develop the flavour characteristic of black tea (Tanaka *et al.*, 2010). In the fifth and final step, the goal is to reduce the water content of the leaves down to 4 or 3% in the drying process. This process is to halt all the enzymatic activity and preserve the chemical and flavour compounds for longer tea's shelf life and minimise packaging requirement (Roshanak *et al.*, 2016). Drying is physically achieved in an oven-like room where the leaves are laid out in thin layers and dried at 100–120°C.

2.3.2(a) Black tea drying

The purposes of drying tea are mainly for stopping the fermentation process, removing the moisture, and producing an excellent quality of tea with longer shelf life (Akhtaruzzaman *et al.*, 2013). Therefore, the final moisture content of tea leaves before packing needs to reach an optimum of 3 to 2.5% of the final weight (Temple & Van Boxtel, 2000). On one hand, if the dried tea is below the moisture content, it disrupts the total quality; and on the other hand, if moisture content is above the standard, the tea easily deteriorates during storage (Hampton, 1992; Kohn, 2015).

There are numbers of different dryers that are applied to reduce the moisture content of the tea leaves. Previously, selection ranges of drying are extensively used in tea industry such as sun-drying (Gulati *et al.*, 2003), fluidised bed dryer (Temple & Van Boxtel, 1999), freeze or vacuum dryer (Mujumdar, 2014) to microwaves (Chan *et al.*, 2009) and radio-frequency dryer (Mujumdar & Law, 2010). Even though each dryer has its advantages, they also lack in certain terms as listed in Table 2.1.

Table 2.1 List of dryers used in the black tea drying process

Drying method	Example	Outcome	References
Sun drying	None	Free process but unhygienic, non-uniform dried product, longer time is required, and is highly weather dependant	Joubert and Schultz (2012)
Convention dryer	Fluid bed, rotary, spray dryer	High rates of heat and mass transfer, however low thermal efficiency, high product exposure time, airflow able to disturb or contaminate the product, and have slower startup	Temple and Van Boxtel (2000)
Conduction dryer	Vacuum dryer	Thermal efficiency is higher but suitable only for products using low temperature	Greensmith (1998)
Radiation	Microwave, Infra-red, solar and radio frequency	The capital of operating is higher, high potential for overheating, and temperature control is harder	Chan <i>et al.</i> (2009) and Mujumdar (2014)

All the above outcomes of each dryer deals with the limitation on operating circumstances that restrict their ability to perform well. Nevertheless, the drying process also highly influences the quality attributes of black tea, such as the colour, its heat sensitive components, and flavour characteristics in tea. Therefore, it is important to investigate the effect of drying on black tea quality under different dryer instruments.

2.3.2(b) Quality parameter of black tea under different drying instruments

Commercial values of black tea leaves highly depend on its quality parameters that emphasise on the need to produce good quality tea. Since drying is the last step in the tea manufacturing process, many works had investigated the effects of different drying methods on the quality parameters of black tea. All the quality parameters of black tea under different dryers are listed in Table 2.2.

Table 2.2 Studies conducted on the effect of different drying modes on black tea leaves

No	References	Dryer types	Dryer conditions	Parameters	Highlights
1	Temple <i>et al.</i> (2001)	Batch fluidised bed dryer	60–140°C 1–64 min	<ul style="list-style-type: none"> • Polyphenols • Sensory 	<ul style="list-style-type: none"> • Lowest temperature (60°C) needed a longer time to reach standard moisture content and lead to undesirable colour and flavour. • Highest temperature (140°C) can cause ‘case hardening’ to the black tea leaves • 100–120°C can help in preserving flavour as it shortened the time of drying.
2	Teshome <i>et al.</i> (2013)	Hot air drying	90–130°C 20–30 min	<ul style="list-style-type: none"> • Sensory evaluation • Theaflavins and thearubigins 	<ul style="list-style-type: none"> • Most biochemical composition decreased at elevated temperature and time. • 100°C and 25 min were suggested as the optimum drying conditions.
3	Shahabi <i>et al.</i> (2014)	Hot air-convection dryer	50, 60, 70, 80, and 90°C 0.5, 1, and 1.5 m/s	<ul style="list-style-type: none"> • Colours (L^*, a^*, and b^*) 	<ul style="list-style-type: none"> • An increase of redness was obtained at increasing temperature and time. • While lightness and yellow/blue coordinate decreased in values.
4	Kishore <i>et al.</i> (2014)	Vacuum dryer	75, 85, and 95°C	<ul style="list-style-type: none"> • Colour • Aroma index 	<ul style="list-style-type: none"> • CTC black tea achieved the highest redness levels at high temperature (95°C), while orthodox black tea at moderate temperature. • The yellowness of black tea reached the highest value at moderate and high temperature for CTC and orthodox black tea, respectively.
5	Demirhan and Özbek (2015)	Microwave dryer	180, 360, 540, 720, and 900 W	<ul style="list-style-type: none"> • Colours 	<ul style="list-style-type: none"> • Colour parameters (L^*, a^*, and b^*) of black tea shifted towards the darker region as the microwave power output increased.

					<ul style="list-style-type: none"> • Browning index increased with an increased of microwave power.
6	Karadağ <i>et al.</i> (2016)	Microwave dryer	860 W/h and 18 min	<ul style="list-style-type: none"> • Total phenolic content • Antioxidant activities • Sensory attributes 	<ul style="list-style-type: none"> • Microwaved black tea leaves improved in quality constituents (phenolic content and antioxidant activities) compared to commercial Turkish black tea. • The sensory result also presented higher acceptability among consumers.
7	Tuan <i>et al.</i> (2016)	Hot air dryer	80, 90, 100, 110, 120, 130, and 140°C	<ul style="list-style-type: none"> • Aroma constituents 	<ul style="list-style-type: none"> • Volatile compounds increased at drying temperature 80–120°C and decreased above 120°C.
8	Polat <i>et al.</i> (2018)	Hot air dryer	100, 130, and 160°C	<ul style="list-style-type: none"> • Volatile compounds 	<ul style="list-style-type: none"> • Volatile groups behaved differently at increasing temperature. • Aldehyde, ketone, terpenes, acid, and lactone significantly increased when the drying temperature increased. • Furans, hydrocarbons, and esters decreased at increasing temperature.
9	Qu <i>et al.</i> (2019)	Hot air dryer Far-infrared dryer Microwave dryer Halogen lamp dryer Halogen lamp microwave dryer	150°C and 20 min 135°C and 30 min 700 W and 6 min 700 W and 16 min 343 W and 10 min	<ul style="list-style-type: none"> • Sensory evaluation • Polyphenols contents • Amino acid • Volatile compounds 	<ul style="list-style-type: none"> • The distinct impact was observed on colour, aroma, and taste of black tea dried under the different dryer. • Halogen lamp-microwave and microwave showed better sensory qualities of black tea leaves. • The microwave also obtained the highest retention of polyphenols, catechins, and theaflavins. • Far-infrared and hot air dryer gave minimum loss of amino acid and soluble sugars in black tea. • Microwave and halogen lamp dryer were able to improve the black tea quality and perform better than conventional hot air dryer.