Scientific substantiation of the absence of combustion in the Electrically Heated Tobacco Product (EHTP) and that the aerosol emitted is not smoke

Markus Nordlund, Maurice Smith, Serge Maeder, Thomas McGrath,
Jean-Pierre Schaller, Pascal Pratte, Patrick Picavet, Manuel C. Peitsch

Philip Morris International Research & Development

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1   Executive summary

This report consolidates scientific evidence substantiating that no combustion of the tobacco material occurs in Philip Morris International's (PMI) Electrically Heated Tobacco Product (EHTP) when used in the Electrically Heated Tobacco System (EHTS) (also known as the Tobacco Heating System (THS), which is marketed in various countries under the *IQOS* brand name) and that the aerosol generated is not smoke. A review was completed of the physical and chemical processes occurring in tobacco as a function of temperature, including the fundamentals behind aerosol formation in general and smoke formation in particular, as well as a consideration of the available technical and scientific definitions of smoke and combustion (see Appendix). Based on these reviews, the scientific evidence comprehensively demonstrates that no combustion of the tobacco material occurs during use of the EHTP in the EHTS Holder and that the aerosol generated from the EHTP tobacco material is not smoke.

Key takeaways of this report include the following:

- Combustion (burning) is defined as a chemical process of oxidation that occurs at a rate fast enough to produce heat and usually light in the form of either a glow or flame.

- Combustion includes both complete and incomplete (partial) combustion processes, such as smoldering (flameless) and flaming combustion.

- Combustion is initiated by ignition and is self-sustaining as long as the exothermic (heat-generating) oxidation reaction is sufficiently strong to overcome heat losses and endothermic (heat consuming) processes, such as vaporization and endothermic thermal decomposition.

- For combustion (burning) of the EHTP tobacco material to occur, the temperature of the tobacco material would need to exceed about 400°C.

- The maximum temperature measured in the tobacco material of the EHTP during use in the EHTS Holder is 320°C. This is well below the temperatures required for combustion of the tobacco material to occur.

- There are no signs of net exothermic (heat-generating) processes in the EHTP tobacco material during use in the EHTS Holder. In fact, when power to the heater is switched off, there is an immediate decrease in the temperature of the tobacco material.

- As the EHTP is heated to temperatures below the ignition temperature of the EHTP tobacco material, the tobacco material undergoes processes such as drying and vaporization, and close to the Heater also thermal decomposition (torrefaction and low-temperature pyrolysis), but no combustion (neither incomplete nor complete).

- The comparison of the chemical composition of the EHTS aerosol generated in oxidative (air) and non-oxidative (nitrogen) environments indicated that oxygen (necessary for combustion to happen) does not play a major role in the thermal decomposition of the EHTP tobacco or the aerosol formation.

- The presence of low levels of gaseous compounds in the EHTS aerosol, such as carbon monoxide, carbon dioxide, nitrogen oxides, etc. (usually associated with combustion), is not evidence that combustion has occurred, as low amounts of these compounds are

generated during low-temperature thermal decomposition of tobacco components during heating, independent of the oxygen availability.

- Smoke is an aerosol containing liquid and solid particles (particulate matter).

- Smoke particulate matter is formed when products of combustion and high-temperature pyrolysis (e.g., hydrocarbons) reach supersaturation and either condense to form droplets or react together to form particles, or by the nucleation and growth of positively charged hydrocarbon ions to form soot particles.

- While smoke is an aerosol, not all aerosols are smoke. The EHTS aerosol liquid particulate matter (droplets) is not formed from condensation of byproducts of combustion or pyrolysis. The droplets are instead generated when glycerol (added to the tobacco material during processing as an aerosol former) is vaporized and reaches supersaturation and condenses on cooling, forming nuclei, onto which more glycerol, water, nicotine, and other constituents can condense to form liquid aerosol droplets.

- Aerosols formed from the condensation of water vapor, vaporized e-liquids, or vaporized glycerol from the EHTP tobacco material are not smoke and are very different in terms of origin and chemical and physical composition compared with smoke aerosols formed from the combustion and associated high-temperature pyrolysis products generated from the burning of tobacco.

- The aerosol generation process in the EHTS is equivalent to the aerosol generation process in most e-vapor products (EVP), for which aerosol formers (glycerol and propylene glycol) in the e-liquid are vaporized during heating and are subsequently cooled down to form liquid aerosol droplets.

- The EHTP, when used as intended in the EHTS Holder, does not generate solid particles, as demonstrated by detailed experiments involving scanning electron microscopy (SEM) analysis. Thermodenuder and thermodilution techniques are not able to determine the presence of solid particles alone without being combined with microscopy analysis, such as SEM.

- As the tobacco material in the EHTP is heated and not burned when used in the EHTS Holder, the levels of the majority of the analyzed harmful and potentially harmful constituents in the EHTS aerosol were reduced by >90% compared with the levels in the mainstream smoke of a 3R4F reference cigarette.

- More than 97.8% w/w (mass fraction) of the EHTS mainstream aerosol constituents transfer via vaporization/direct transfer processes to the aerosol from the multicomponent EHTP. Less than 2.2% w/w of the EHTS mainstream aerosol constituents are likely to be formed by torrefaction and low-temperature pyrolysis processes, as suggested by the limited influence of oxygen on the aerosol composition.

- The mere presence of low amounts of pyrolysates in an aerosol does not imply that the aerosol is smoke, as the amount of the pyrolysates must be high enough to reach supersaturation to condense (nucleate) or to react with each other to form particulates. For example, the aerosols generated by EVPs contain low amounts of pyrolysates and are not considered to be smoke.

- The absence of combustion in the EHTP, when used as intended in the EHTS Holder, and that the aerosol generated is not smoke have been substantiated by scientific evidence and have been verified by third-party scientific experts in numerous countries as well as by independent research organizations.

To summarize, scientific evidence comprehensively demonstrates that there is no combustion of the EHTP tobacco material occurring during intended use of the EHTS and that the aerosol generated is not smoke.

2   List of abbreviations

BDL	Below the limit of detection
CO	Carbon monoxide
CO_2	Carbon dioxide
CH_4	Methane
EHTP	Electrically Heated Tobacco Product
EHTS	Electrically Heated Tobacco System
EVP	E-vapor products
HCI	Health Canada Intense
HPHC	Harmful and potentially harmful constituents
LLOQ	Lower limit of quantification
LOD	Limit of detection
LOQ	Limit of quantification
NFPA	National Fire Protection Association
NFPDM	Nicotine-free dry particulate matter
NO_x	Nitrogen oxides
NQ	Not quantified (below the limit of quantification)
PAH	Polycyclic aromatic hydrocarbons
PMI	Philip Morris International
R	Radial distance
SEM	Scanning electron microscopy
TG	Thermogravimetric
TGA	Thermogravimetric analysis
TGPM	Total gaseous and particulate matter
TPM	Total particulate matter
THS	Tobacco Heating System
TUM	Technical University of Munich

3   Introduction

The purpose of this report is to review the physical and chemical processes occurring when heating tobacco as well as the fundamentals behind aerosol formation in general and smoke formation in particular. In addition, available technical and scientific definitions of key terms are reviewed together with a summary of the scientific evidence demonstrating that there is no combustion of the tobacco material occurring and that the aerosol generated during use of the Electrically Heated Tobacco System (EHTS) (also known as the Tobacco Heating System (THS)), is not smoke. EHTS uses a heat not burn technology to heat the tobacco (without burning it) and generates an inhalable aerosol.

4   Electrically Heated Tobacco System

4.1   Product description

The EHTS, developed by Philip Morris International (PMI), has three distinct components that perform different functions: (i) an Electrically Heated Tobacco Product (EHTP), a multicomponent tobacco product containing a specially formulated and processed tobacco material made from tobacco powder, water, glycerol, guar gum, and cellulose fibers; (ii) a Holder, into which the EHTP is inserted and that heats the tobacco portion of the EHTP by means of an electrically controlled Heater; and (iii) a Charger, which is used to recharge the battery in the Holder after each 360-second heating cycle or 14 puffs, whichever comes first. A schematic of the EHTS and of the multiple components of the EHTP inserted into the Holder (heating device) are shown in Figure 1(a) and Figure 1(b), respectively.

4.2   Product operation

To operate the EHTS, an EHTP is inserted into the Holder, and a 360-second heating cycle is initiated. The Holder contains a Heater that heats the tobacco material in the EHTP radially outwards from the center of the tobacco plug (see Figure 1(b)). During a puff, air enters the Holder through a gap between the outer and inner device walls and then exits through the components of the EHTP. The Heater (heating blade) consists of a platinum track encased in a ceramic substrate. Heat is supplied to the tobacco material for a fixed period of 360 seconds and allows for up to 14 puffs to be taken. The device switches off after 14 puffs or 360 seconds, whichever comes first. The temperature profile of the Heater is electrically controlled by a firmware and a micro-controller, and the electrical current supplied to the Heater is continuously monitored by the device throughout the 360-second heating cycle. The electrically controlled Heater in the Holder is programmed to reach an average temperature over the Heater surface between 320°C and 350°C according to a predefined profile, and if its operating temperature exceeds 350°C, the energy supply to the Heater is cut. During the controlled heating process, volatile compounds vaporize from the tobacco material, and low-temperature thermal decomposition reactions occur in the tobacco material in close contact with the Heater; however, combustion and high-temperature pyrolysis reactions are avoided [1]. When the vaporized compounds are cooled down in the EHTP, a nicotine-containing aerosol that is not smoke is formed, containing significantly lower levels of harmful and

potentially harmful constituents (HPHC) compared with those measured in cigarette smoke [2-4].

More information on the design and the operation of the EHTS during use can be found in [5].

Charger

EHTP

Holder

(a)

Battery   Heater

Aerosol cooling
element   Cellulose Acetate
                 Mouth Piece

Holder

Tobacco   Hollow Cellulose
mixture   Acetate tube   EHTP

(b)
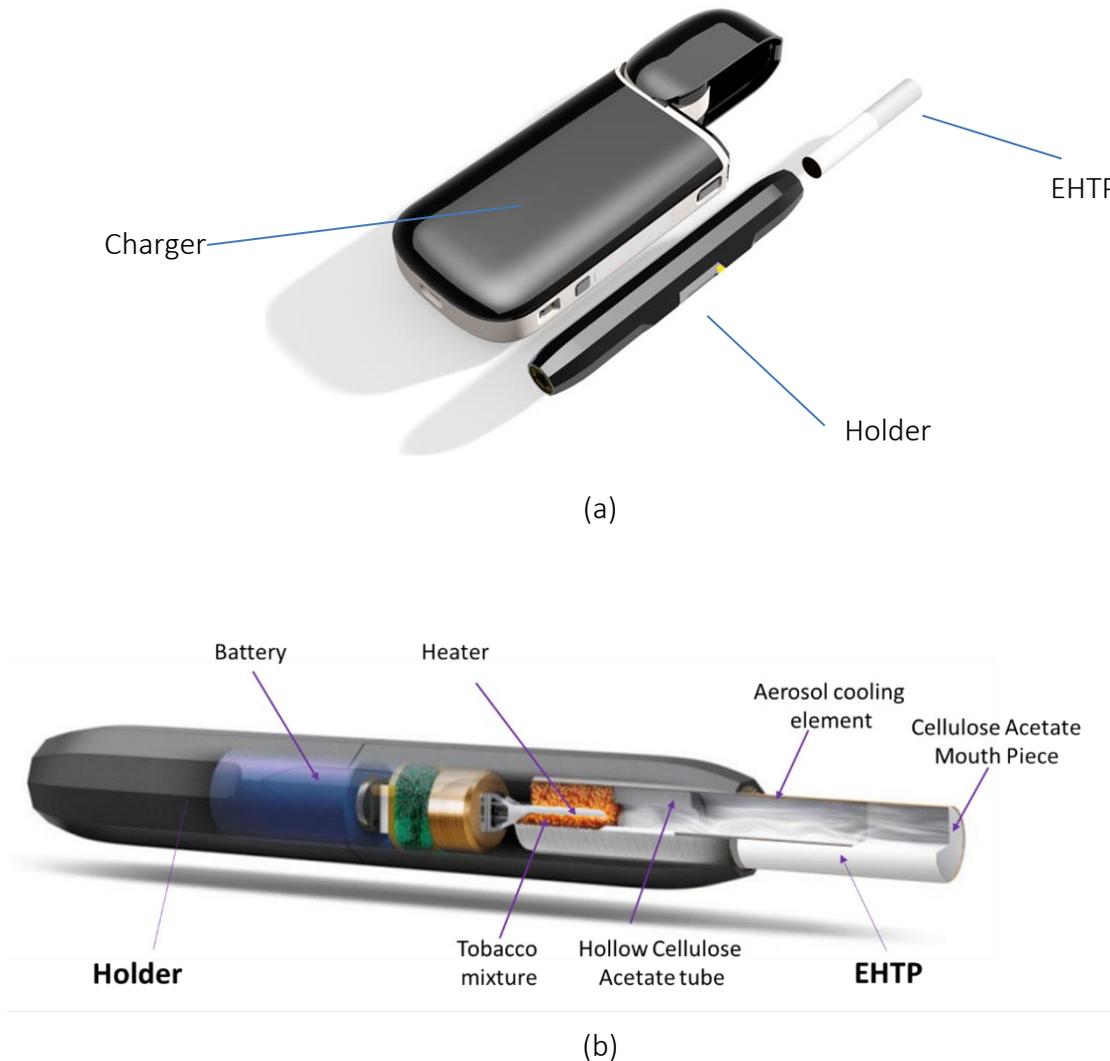
Figure 1. (a) The three components of the EHTS and (b) a schematic of the cross-sectional view of the EHTP and its components when inserted into the Holder.

5   Scientific overview of physical and chemical processes

5.1   Physical and chemical processes occurring in tobacco as a function of temperature

Tobacco is a lignocellulosic biomass built up of numerous constituents, including hemicellulose, cellulose, lignin, pectin, sugars, proteins, organic acids, amino acids, starch, polyphenols, water, waxes and nicotine [6, 7]. In the tobacco material used in the EHTP, glycerol and guar gum are also important constituents [5]. When heated, the tobacco constituents undergo various thermochemical processes depending on the heating temperature.

These thermochemical processes are briefly reviewed in this section from a fundamental perspective together with the main emissions associated with the processes. Technical and scientific definitions of terms used in this section can be found in the Appendix at the end of this report.

5.1.1   Drying

Drying is the first thermal process occurring in biomass, such as tobacco, when thermal energy is provided by an external heat source. At temperatures below the boiling point of water (100°C), liquid water absorbed by the tobacco starts to evaporate into gas phase, creating water vapor emissions [8, 9]. In addition, vaporization of water bound in the tobacco also begins from temperatures below its boiling point and is largely complete at around 150°C [10]. During drying, tobacco loses moisture and shrinks but has still the ability to retain its structure if being rewetted [10]. There is no chemical transformation of the water during the endothermic process of drying but only a phase change of water from liquid to vapor via thermodynamically controlled evaporation.

5.1.2   Volatilization

Like water, organic components in the tobacco are also vaporized when the tobacco is heated either by an external source or by exothermic (heat-generating) processes. Vaporized compounds may later condense back onto cooler parts of the tobacco. This combined vaporization and condensation process is usually referred to as distillation. Evaporation of relatively stable high-molecular weight species in the tobacco occurs mostly below 250°C [11-13]. For example, evaporation of glycerol, which is added to the tobacco as humectant and aerosol former with a boiling point of 290°C, occurs at temperatures between 120-250°C [13], while nicotine have been reported to evaporate between 110°C and 220°C [14], even though the boiling point of nicotine is 247°C. Vaporization is an endothermic phase change process controlled by thermodynamics without chemical transformation of the vaporized compounds. Under conditions where only drying and vaporization occur, temperature changes in the tobacco only relate to enthalpy changes, and mass loss from the tobacco is associated with the phase change only. These processes are therefore also referred to as inert heating [15] or non-reactive drying [10].

5.1.3   Thermal decomposition / pyrolysis

As the temperature increases and becomes sufficient to overcome the activation energy needed to break chemical bonds in the tobacco constituents by heat alone, thermal decomposition of the tobacco constituents into different, typically shorter chain compounds occurs. This process is known as thermal decomposition or pyrolysis and can be both endothermic and exothermic [10, 16-18]. Pyrolysis is sometimes referred to as destructive distillation of organic compounds in an oxygen-free environment that converts the organic matter into gases, liquids, and char [18]. During pyrolysis, molecules are subjected to elevated temperatures leading to molecular vibrations at which the molecules are stretched and shaken to such an extent that they start breaking down into smaller molecules [19]. Pyrolysis often precedes other processes, such as gasification and combustion, where partial or total oxidation

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of the treated material occurs [19]. It is important to note that thermal decomposition via pyrolysis does not require oxygen and is a completely different process than combustion, for which oxygen is indispensable.

Thermal decomposition of tobacco encompasses numerous processes depending on the degraded tobacco constituent and the temperature. At temperatures around 120°C to 150°C, the mechanical strength of the tobacco is reduced as the lignin filling the spaces in the cell wall between cellulose, hemicellulose, and pectin components starts to soften [10]. Dehydration of low-molecular weight carbohydrates (mono- and di-saccharides) starts around 150°C, leading to structural rearrangements from which the tobacco loses its ability to regain its original structure. Decomposition products include anhydro saccharides, anhydro saccharidic acids, and compounds with furan-like structures, as well as carbon dioxide (CO_2) from decarboxylation [20]. Organic acids also start to degrade around these temperatures, with malic acid starting to decompose into water, carbon monoxide (CO), CO_2, and lower carboxylic acids, such as formic acid, acetic acid, and pyruvic acid, at around 140°C [21, 22]. Amino acids have been reported to decompose between 185°C and 270°C, resulting in lactams and other heterocyclic nitrogen compounds [23]. Fats, waxes, and resins partially vaporize or decompose into smaller molecules.

Pyrolysis in the temperature range of 200-300°C is also known as torrefaction or mild pyrolysis [10, 24], which is the primary process leading to the characteristic darkening of coffee beans and the increase in their aromaticity during coffee roasting [25]. Heating tobacco to temperatures associated with torrefaction also results in similar carbonization and devolatilization, which result in the disruption of most inter- and intramolecular hydrogen bonds, C–C and C–O bonds. This results in the formation of hydrophilic extractives, carboxylic acids, alcohols, aldehydes, ether, gases including CO, CO_2, and methane (CH_4), and aromatics [10, 24, 26-28]. The chemical compositional changes from devolatilization and carbonization of the polymers makes the tobacco darken and become brittle [24] and both aliphatic and aromatic in nature [29-31].

Decomposition of compounds such as starch, hemicellulose, lignin, and pectin occurs within the temperature range associated with torrefaction. These compounds decompose mostly at temperatures higher than 220°C [10, 11, 17, 32-36]. While the onset of endothermic decomposition for cellulose occurs at slightly higher temperatures (above 300°C), hemicellulose and pectin undergo exothermic decomposition between 220°C and 250°C [17, 33, 35], and the decomposition of starch begins around 250°C to 260°C [34, 37]. The thermal decomposition of lignin has been reported to start around 150°C [17, 32] and takes place over a larger temperature range due to its diverse structure. The decomposition is most intense between 200°C to 500°C, where phenols, formaldehyde, methanol, CO, and CO_2 are typical thermal decomposition products [10, 32]. Exothermic decomposition reactions include depolymerization and the production of altered and rearranged polysaccharide structures but do not involve an external oxidizer, even if CO or CO_2 are produced [15]. At temperatures above 250°C, hemicellulose decomposes extensively into volatiles and a char-like solid product, whereas lignin and cellulose show limited devolatilization and carbonization [15].

At temperatures between 400°C and 600°C, pyrolysis of cellulose, amino acids, and esters dominate the thermal decomposition together with various combustion reactions [38]. Heating

tobacco to temperatures above those associated with torrefaction results in further increase in the aromaticity of the remaining solid with the evolution of light gases, aromatics, and polycyclic aromatic hydrocarbons (PAH), as well as oxygen and nitrogen containing aromatics and PAHs [31]. Most of the products emitted from thermal decomposition are formed from the tobacco at temperatures below 600°C [38].

At low temperatures, endothermic vaporization and decomposition reactions dominate over exothermic decomposition reactions requiring an external heat supply to sustain the thermal decomposition of the tobacco. When increasing the temperature, exothermic decomposition may overwhelm the endothermic decomposition, resulting in a net exothermic process that releases a surplus of heat [15]. If this surplus of heat or the heat provided by an external energy source surpasses the apparent activation energy of all vaporization and decomposition reactions combined and exceeds the energy required to activate oxidative chemical reactions, then oxidative decomposition occurs [15]. Contrary to combustion, the oxidation in oxidative decomposition does not occur via reactive radicals (O$^{.}$, HO$^{.}$) but rather via the significantly slower reacting oxygen molecule (O$_2$) [20]. The presence of oxidation reactions during thermal decomposition can lead to slightly enhanced thermal decomposition between 250°C and 400°C, even though the mass loss and emissions are almost the same as those observed in thermogravimetric (TG) analysis (TGA) under non-oxidative atmospheres [9, 12, 16, 39]. The net energy release, however, is not sufficient to overcome all vaporization and decomposition reactions as well as heat losses driven by heat and mass transfer processes to become a self-sustaining propagating decomposition process. Oxidative decomposition is sometimes referred to as cold oxidation or auto oxidation [20], which occurs via slow oxidation reactions. Examples of familiar cold oxidation reactions are the rusting of iron, hardening of paints, rotting of food, or the enzymatic browning of foods, such as apples [20]. Oxidative decomposition processes should not be misinterpreted as combustion reactions, which require rapid oxidation reactions.

5.1.4 Combustion

Combustion is defined as a chemical reaction between oxygen and a material (oxidation) happening at a rate fast enough to produce heat (increased temperature) and usually light. Combustion (burning) is initiated by ignition [18]. For ignition of the tobacco material (and therefore also combustion) to occur, the energy either supplied by an external source or generated from exothermic thermal decomposition reactions needs to be sufficient to overcome not only all endothermic vaporization and thermal decomposition reactions in the vicinity of where the energy is provided, but also the heat and mass transfer-induced energy losses. As long as this criteria is reached, the combustion of the tobacco material is self-sustained. In the same way that oxygen is an indispensable element for combustion, ignition energy to get the combustion reaction started is also one of the three elements required for combustion to occur, as visualized in the combustion triangle shown in Figure 2.
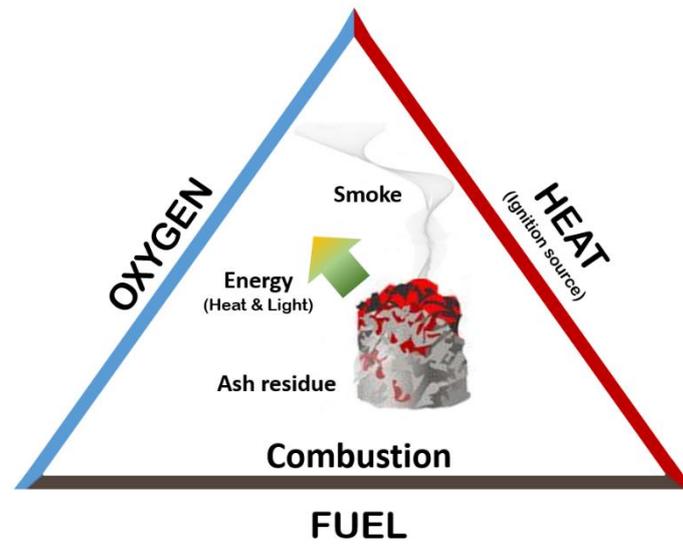
Figure 2. Combustion triangle indicating elements required for combustion of tobacco to occur, i.e., the fuel (tobacco), oxygen, and heat as ignition source.

The ignition temperature of tobacco can be determined by assessing the decomposition kinetics of the tobacco material by conducting thermal decomposition studies in non-oxidative (nitrogen or helium) and in oxidative (air) atmospheres under controlled heating conditions. TGAs of different tobacco materials were performed by various researchers in the two atmospheres at constant heating rates [9, 12, 16, 39]. As shown in Figure 4, the TG curves (mass evolution curves) from both the non-oxidative and the oxidative atmosphere experiments follow each other closely up to about 400°C for all reconstituted tobaccos tested by the various researchers. This indicates that the thermal decomposition behavior of the tobacco material is largely independent of the oxygen availability in the atmosphere below 400°C. Above about 400°C, a rapid oxidation-induced mass loss occurs in the oxygen containing (air) atmosphere, whereas this rapid mass loss is absent in the non-oxidative atmospheres. The rapid mass loss starting around 400°C in the air atmosphere experiments is directly related to combustion of the tobacco material remaining after thermal decomposition [9, 12, 16, 39]. Therefore, the ignition temperature of reconstituted tobacco under the testing conditions is around 400°C.

(a)

(b)

(c)

(d)
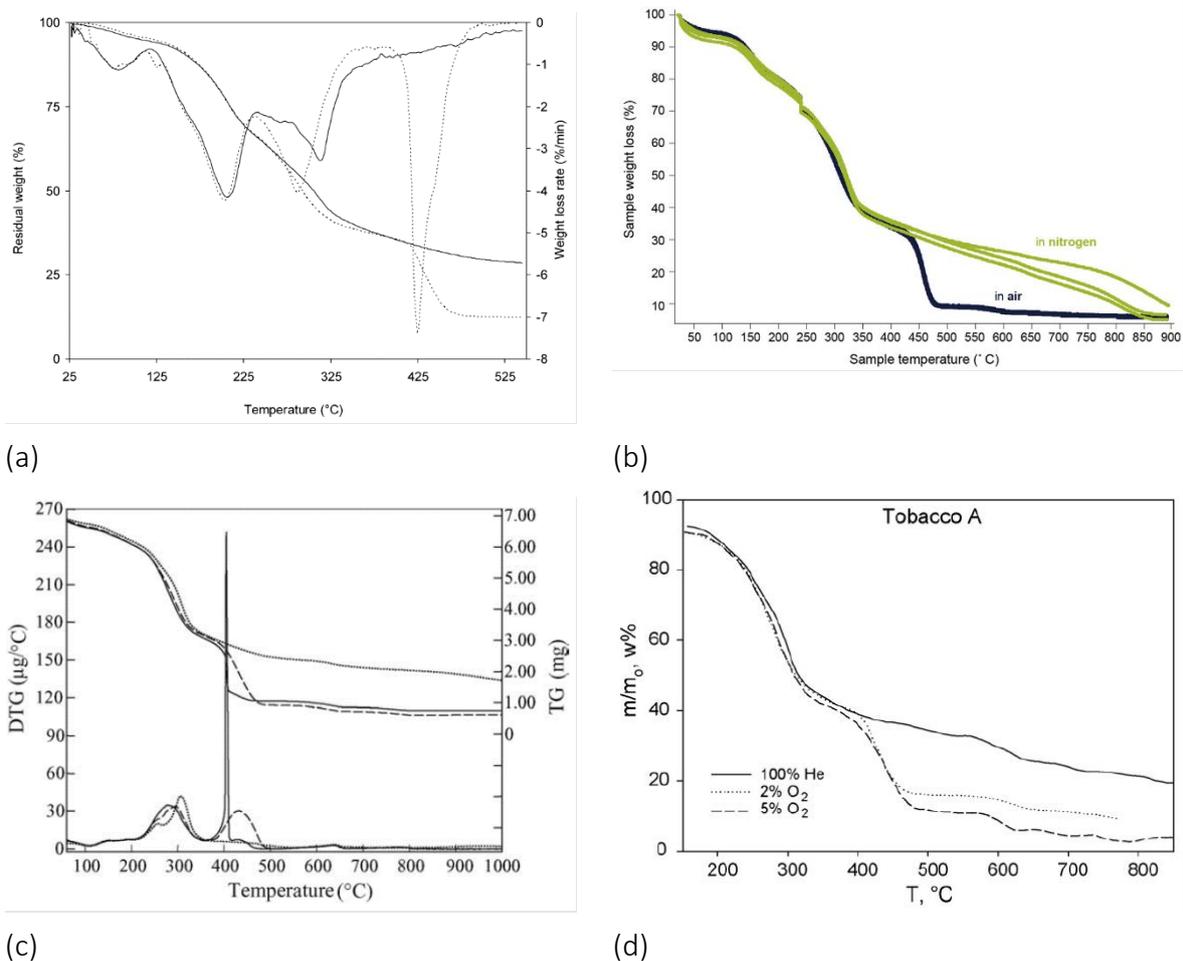
Figure 3. (a) Comparison of TG (weight loss) and differential TG (DTG) (weight loss rate) results obtained for the shredded sheet of homogenized tobacco material, which contained glycerol as an aerosol former in inert and oxidizing atmospheres using a 10°C/min constant heating rate (continuous lines: nitrogen atmosphere; dotted lines: air) [12]. (b) TGA curves of a reconstituted tobacco material heated in both air and nitrogen using a constant heating rate of 5°C/min [9]. (c) TG and DTG curves of a reconstituted tobacco sheet in different atmospheres at the heating rate of 5°C/min. Atmosphere: N_2 (·······), 5% (volume) O_2 in N_2 (— —), 20% (volume) O_2 in N_2 (—) [39]. (d) TG curves obtained at 10°C/min for a homogenized tobacco in powder form in different atmospheres [16].

As shown in Figure 3, at temperatures above 400°C and in the presence of an oxidant (such as the oxygen in air), the residual solid phase "fuel" from the thermal decomposition ignites and begins to undergo a smoldering combustion (flameless combustion) until the fuel is consumed. Combustion processes can be either complete or incomplete (partial) [40, 41]. In complete combustion, there is sufficient oxygen available so that when oxygen reacts with a fuel in a complete oxidation reaction, only CO_2 and water are produced [40]. Complete combustion rarely occurs in natural combustion situations, and almost all combustion processes found in nature are incomplete combustion processes, such as smoldering combustion [38, 42, 43] and

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most flaming combustions [40, 41]. In incomplete combustion (occurring when there is an insufficient supply of oxygen), the combustion of carbonic fuel produces incompletely oxidized carbon compounds. Combustion products produced during incomplete combustion include water, CO, CO_2, PAHs, nitrogen oxides (NO_x), polychlorinated aromatics, hexachlorobenzene, hydrogen cyanide, alcohols, and ketones [6, 38, 40-47]. Some of these products contribute to the formation of fine solid and liquid particulate matter, which together with all emitted gaseous compounds constitute smoke [6, 38, 40-47]. Incomplete or partial combustion should be distinguished from other thermal processes that are dependent on heat alone, such as drying, vaporization, and pyrolysis, where oxygen is not necessary for the processes to occur. Incomplete combustion reactions are also different from oxidative thermal decomposition processes, for which only slow oxidation reactions occur, as described previously. It is impossible in practice to have biomass completely or even partially combust in the absence of lower-temperature thermal processes, such as drying, vaporization, and pyrolysis. However, it is possible to induce such thermal processes in biomass materials without complete or partial combustion occurring by heating tobacco to temperatures below its ignition temperature.

The burning of tobacco in a cigarette is an example of a smoldering combustion process, where temperatures up to about 850°C are generated at the burning tip of the cigarette when the tobacco is lit with a match or lighter [45, 48-50]. When air is drawn through the burning tip, temperatures rise due to the increased oxygen availability, evidencing the exothermic (heat-generating) characteristics of combustion reactions.

5.2   Fundamentals of aerosol formation

The previous section describes the physical and chemical processes resulting in the emissions from tobacco when it is heated to different temperatures. In tobacco products intended for inhalation, the volatiles emitted in the zones at elevated temperatures need to be cooled down to form aerosols containing liquid and/or solid particles. In this section, the fundamentals behind aerosol formation in general and smoke formation in particular are reviewed.

5.2.1   Smoke formation

Smoke is an aerosol containing solid and liquid particles and is generally considered to be a product of combustion and sometimes also a result of high-temperature pyrolysis [18, 51-55]. The mechanisms behind smoke formation were outlined in the review by Bankston et al. (1981) [56] and Rasbash & Drysdale (1982) [57]. According to the authors, smoke particulates are formed when combustion and high-temperature pyrolysis products (e.g., hydrocarbons and generally high-molecular weight compounds) reach supersaturation and condense (nucleate) to form droplets and react with each other to form nuclei, or by the nucleation and growth of positively charged hydrocarbon ions resulting in the formation of soot particles [56]. For particulate formation to happen from pyrolysis (the temperature of a pyrolyzing material should be between 325°C and 625°C) [54]), the concentration of hydrocarbon pyrolysates in the vapor evolving from the decomposition of the tobacco components must be high enough for a hydrocarbon to reach supersaturation at the specific thermal conditions (cooling rates) occurring in the product during use. The mere presence of pyrolysates (byproducts of pyrolysis) in an aerosol does not unambiguously imply that the aerosol can be defined as smoke, as the

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amount of the pyrolysates must be high enough to reach supersaturation to condense (nucleate) or to react with each other to form particulate matter [54].

Cigarette smoke is generated when the tobacco in a cigarette is lit and a self-sustaining combustion process is established. During the smoldering combustion, the tobacco in the burning cone of the cigarette reaches temperatures above 850°C [50]. The large amount of energy released by the combustion process breaks down tobacco components, generating a complex smoke aerosol (composed of gases, liquid droplets, and solid particles (soot) suspended in air) and leaving a residue of ash (composed of inorganic compounds originally present in the tobacco material). Cigarette smoke is a complex mixture in which more than 6,000 chemicals have been identified [7]. A smaller number of these chemicals have been classified by public health authorities as likely causes of smoking-related diseases, such as lung cancer, heart disease, and emphysema. The majority of these HPHCs are generated at elevated levels by processes occurring at temperatures associated with tobacco combustion [27, 58].

5.2.2   Formation of aerosols that are not smoke

Unlike smoke aerosol formation, droplets in aerosols that are not smoke are formed from vaporized compounds that cool down to reach supersaturation and condense (nucleate) to form liquid droplets [59]. The vaporized compounds originate from vaporization from either a liquid or a solid material containing volatile compounds. During vaporization, chemical compounds change their physical state from liquid or solid phase to gas phase without undergoing a chemical reaction.

Cloud formation in the atmosphere is a typical example of how aerosols that are not smoke are formed. In this process, water is first evaporated from, for example, lakes or oceans by solar radiation. Thereafter, the water vapor rises and is progressively cooled down until it becomes supersaturated. The supersaturated water vapor condenses (nucleates) to form liquid droplets, which together constitute the cloud.

Another example is the aerosol formation in e-vapor products (EVP). In EVPs, an e-liquid, consisting of a mixture of propylene glycol, glycerol, water, flavors, and often nicotine derived from tobacco, is heated electrically by a heating element (usually in the form of a coil or a metallic or ceramic mesh) when a puff is taken. When heating the e-liquid, its constituents evaporate to form a mixture of the vaporized compounds. When this vapor mixture is mixed with air during puffing, it cools down, and compounds with low vapor pressure, such as glycerol and propylene glycol, reach supersaturation and condense (nucleate) to form liquid-based aerosol droplet nuclei, onto which other vapors (water, nicotine, flavors) can condense to form the final aerosol droplets. During heating of the e-liquid in EVPs, thermal decomposition (mild pyrolysis) of the liquid components has been reported in peer-reviewed scientific literature [60-67]. In these studies, small amounts of byproducts from pyrolysis of propylene glycol and glycerol, including acrolein, acetaldehyde, formaldehyde, glycidol, and acetol, were reported for various e-liquid formulations and EVPs [66, 68, 69]. The amount of carbonyl pyrolysates generated and found in the EVP aerosol was correlated with the energy applied to the e-liquid during vaporization. The pyrolysates found in the aerosol from EVPs were generated by thermally induced decomposition of propylene glycol and glycerol used in the e-liquids without the influence of oxidation. This implies that mild pyrolysis of the e-liquid occurs in EVPs,

especially if the temperature of the heating element becomes elevated, as in the case when the heating element is not sufficiently immersed in e-liquid (known as 'dry puffing'). However, the mere presence of low amounts of pyrolysates in EVP aerosols does not imply that the aerosol from EVPs is smoke, as the aerosol droplets in EVPs are generated by propylene glycol and glycerol vapors (evaporated from the e-liquid) nucleating when being cooled down and becoming supersaturated.

In a similar manner to the aerosol generation in EVPs, and contrary to the smoke formation in burning cigarettes, the aerosol droplets of the EHTS aerosol are generated when the vaporized glycerol (having a low vapor pressure) reaches supersaturation and condenses, forming liquid nuclei, onto which more glycerol, water, nicotine, and other constituents can condense to form droplets [70, 71]. The resulting nicotine-containing aerosol is therefore formed by glycerol (which is specifically added to the tobacco material during processing to act as an aerosol former) [70, 71]. When heated, the EHTP tobacco material undergoes a number of thermochemical processes, such as drying, vaporization, and thermal decomposition of the tobacco components close to the Heater, the latter of which is also known as torrefaction or mild pyrolysis, but high-temperature pyrolysis processes and combustion are avoided. Low-temperature thermal decomposition of the tobacco compounds results in such low amounts of pyrolysates that the vapors cannot reach supersaturation and form aerosol droplets [70, 71]. In the same way, low amounts of pyrolysates are present in EVP aerosols (from thermal decomposition of glycerol and propylene glycol) but do not contribute to the aerosol formation in EVPs, as described previously. The liquid droplet-based aerosol generated by heating the EHTP tobacco material in the EHTS Holder is not smoke and contains, significantly lower levels of HPHCs compared with the mainstream smoke of a 3R4F reference cigarette [4, 72].

6   Scientific substantiation of the absence of combustion and smoke

In this section, scientific evidence that the tobacco material in the EHTP does not combust and that the aerosol generated from the EHTP, when used as intended in the EHTS Holder, is not smoke is presented and discussed.

6.1   Ignition temperature of the EHTP tobacco material

To evaluate whether or not combustion of the tobacco material in the EHTP occurs during use, it is important to determine the ignition temperature of the material under controlled conditions. As described in Section 5.1.4, the ignition temperature of tobacco can be determined by assessing the decomposition kinetics of the tobacco material by conducting thermal decomposition studies in non-oxidative and in oxidative atmospheres under controlled heating conditions. TGA of the tobacco material used in the EHTP was performed by [12] in nitrogen and in air atmospheres at a constant heating rate of 10°C/min. Similarly to the observations made for other reconstituted tobacco materials in Section 5.1.4, the TG curves (mass evolution curves) shown in Figure 4 from both the nitrogen and the air atmosphere experiments follow each other closely up to about 400°C, indicating that the thermal decomposition behavior of the tobacco material is largely independent of the oxygen availability in the atmosphere. Above about 400°C, a rapid oxidation-induced mass loss occurs

in the air atmosphere, whereas this rapid mass loss is absent in the nitrogen atmosphere. This rapid mass loss around 400°C in the air atmosphere experiments is directly related to combustion of the remaining tobacco material [9, 12, 16, 39, 73].
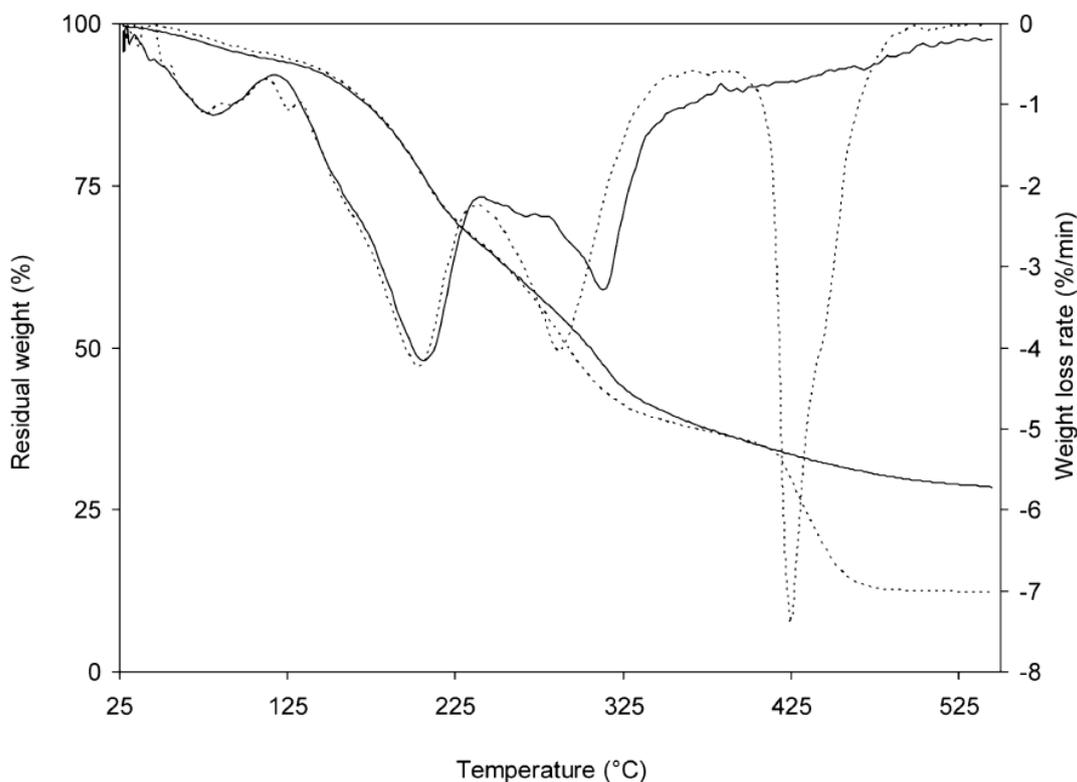

Figure 4. Comparison of TG (weight loss) and DTG (weight loss rate) results obtained for the shredded sheet of homogenized tobacco material, which contained glycerol as an aerosol former in inert and oxidizing atmospheres using a 10°C/min constant heating rate (continuous lines: nitrogen atmosphere; dotted lines: air) [12].

That the rapid mass loss around 400°C in the air atmosphere is a result of combustion, indicating that the ignition temperature of the tobacco material is about 400°C, is evidenced by visual inspection of the tobacco material after it was heated to different temperatures. Figure 5(a) shows that after being heated to 260°C in a nitrogen atmosphere, the fresh tobacco material (Figure 5 (left)) changed its visual appearance (i.e., the color changed from brown to black), which is a typical sign that thermochemical processes, including volatilization, and torrefaction (mild pyrolysis), occurred [10, 24, 31]. Even after being heated to 550°C in the absence of an external oxygen supply (nitrogen atmosphere) (Figure 5(b)), the tobacco material had a similar visual appearance as the tobacco material heated to 260°C (Figure 5(a)).

As the thermal decomposition kinetics below about 400°C were similar for the nitrogen and for the air atmosphere experiments (Figure 4), a similar visual appearance of the tobacco material as was observed in the nitrogen experiments in Figure 5(a) was also observed in the air experiments at 370°C (Figure 6(a)), just below the temperature where the rapid mass loss

occurred in the air experiments. However, in contrast to the nitrogen experiments, Figure 6(b) shows that at 550°C in an air atmosphere, in which oxygen was available, the tobacco material was combusted, leaving mainly a residue of ash composed of inorganic compounds originally present in the tobacco material.

Fresh tobacco material

Tobacco after heating up to 260°C under nitrogen

Tobacco after heating up to 550°C under nitrogen

Figure 5. Visualization of the tobacco material used in the EHTP (left) before and (a) after heating up to 260°C and (b) after heating up to 550°C in a TG oven under a constant heating rate of 5°C/min in a nitrogen atmosphere [73].

Fresh tobacco material

Tobacco after heating up to 370°C under air

Tobacco after heating up to 550°C under air

Figure 6. Visualization of tobacco material used in the EHTP (left) before and (a) after heating up to 370°C and (b) after heating up to 550°C in a TG oven under a constant heating rate of 5°C/min in an air atmosphere [73].

The numerous studies observing thermal decomposition kinetics of tobacco materials similar to those used in the EHTP, together with the presence of an ash residue in the air atmosphere experiments above 400°C, confirm that the ignition temperature of the tobacco material is about 400°C in TGA experiments, for which only limited heat losses occur due to the uniform heating environment. When the EHTP tobacco material is heated by the EHTS Holder during intended use, there are heat losses to the surrounding materials and environment due to thermal gradients. The ignition temperature of the tobacco material during actual EHTP use is

therefore above the ignition temperature observed in the TGA experiments, which serves as a lower limit for combustion to occur.

Further evidence that the ignition temperature of the tobacco material in the EHTP is above 400°C was provided by a study carried out by CRL Energy Ltd., New Zealand [74], on behalf of the Ministry of Health in New Zealand. The tobacco-containing section of the EHTP was pressed against a hot plate in an air atmosphere, and the hot plate temperature was slowly increased to determine the ignition propensity of the tobacco material. A thermocouple inserted in the center of the tobacco section measured internal temperatures of the EHTP during heating. For comparison, two selected cigarettes were also tested in the same manner. In the event that the measured sample temperature would exceed the hot plate temperature (in black), the heat generation by the sample itself would come from an exothermic activity, such as combustion. Therefore, the ignition temperature of the tobacco material would be around the temperature when the measured temperature becomes higher than the temperature of the hot plate. As shown in Figure 7, the hot plate tests indicate that the tobacco material in the EHTP is less likely to lead to combustion than the cut-filler tobaccos used in the cigarettes tested when exposed to a hot surface. Even above 450°C, the tobacco material in the EHTP did not show a net exothermic behavior.
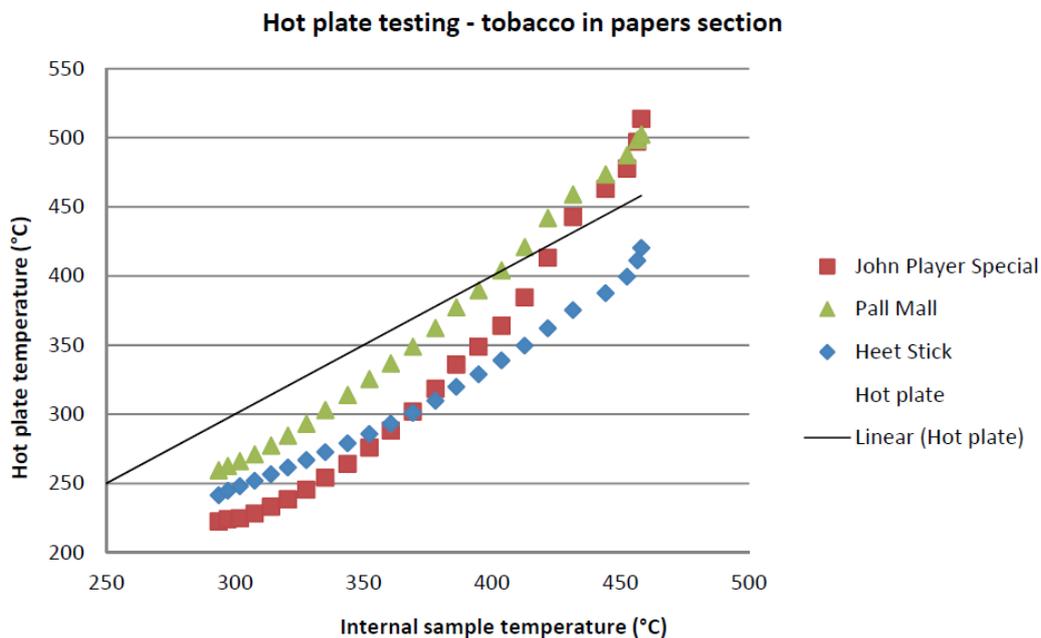
Figure 7. Hot plate auto ignition testing [74]. Heet Stick in the legend refers to the EHTP.

6.2   Temperature of the tobacco material in the EHTP during use

Temperature measurements using thermocouples inserted in the EHTP at different radial distances from the EHTS Heater surface during product use under the Health Canada Intense (HCI) machine-puffing protocol (55 mL puff volume, two-second puff duration, 30-second puff interval) show that the tobacco material touching the Heater surface reaches a maximum temperature of 320°C (Figure 8(a)). This is well below the temperatures required for

combustion of the tobacco material to occur (known to exceed 400°C [9, 12, 16, 39, 74]). Already at 0.2 mm from the Heater, the temperature of the tobacco material is reduced to below 250°C.

Contrary to the increase in the temperature of the tobacco material that occurs when air is drawn through a lit cigarette (rising to above 850°C) [50], there is a significant drop in the temperature of the tobacco material in the EHTP when a puff is taken and cool air is drawn into the EHTS (as shown in Figure 8(a)). In contrast to the exothermic self-sustaining combustion process in a cigarette, the heating of the EHTP tobacco material in the EHTS Holder demonstrates an endothermic behavior (i.e., thermal energy generated by the Heater is required to heat up the tobacco material and to maintain its temperature). When the Heater is switched off, the temperature of the tobacco material immediately drops (as shown in Figure 8(a)).
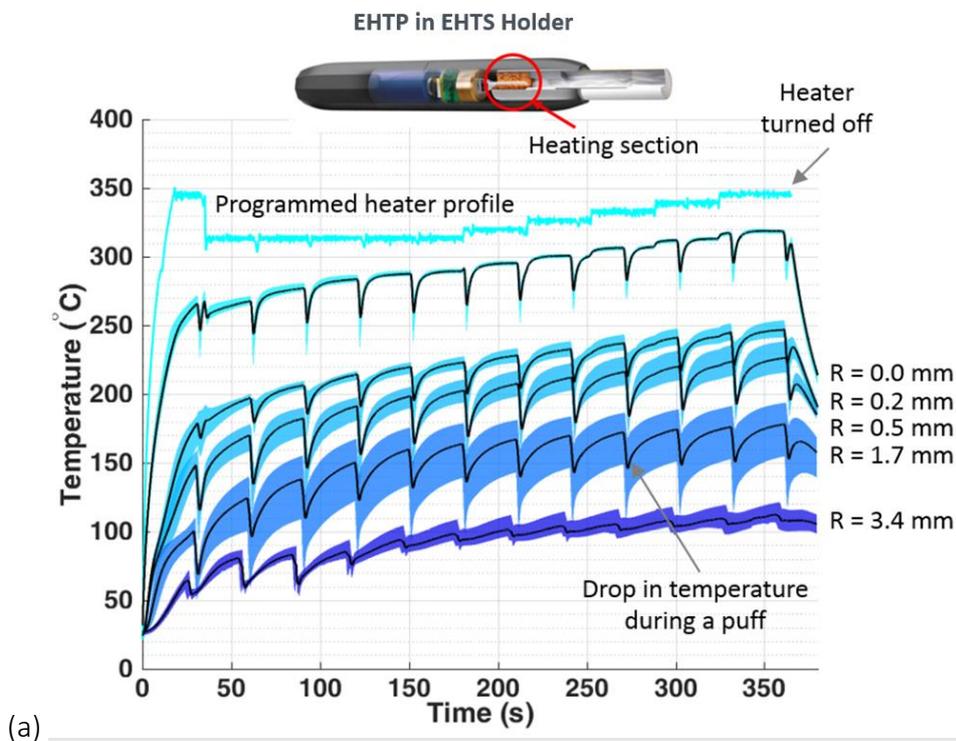
(a)
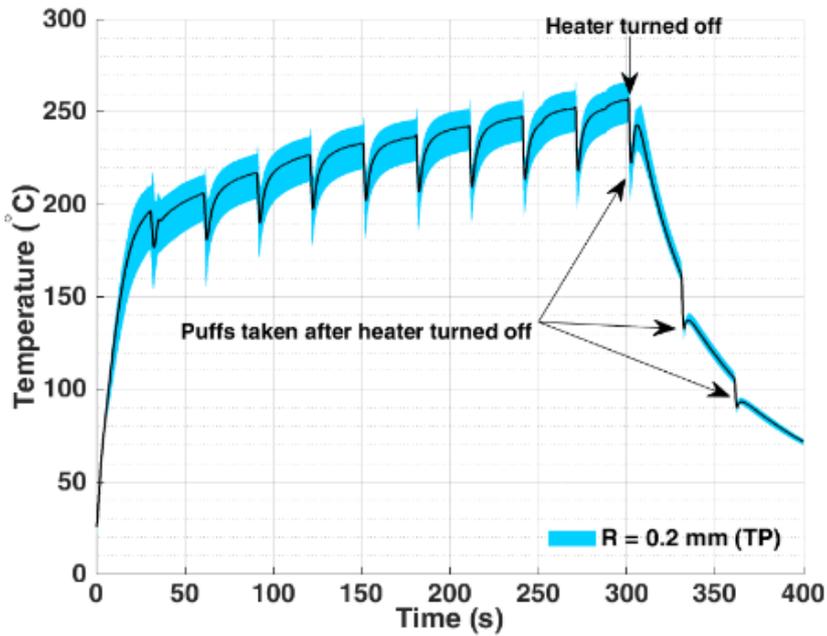
(b)
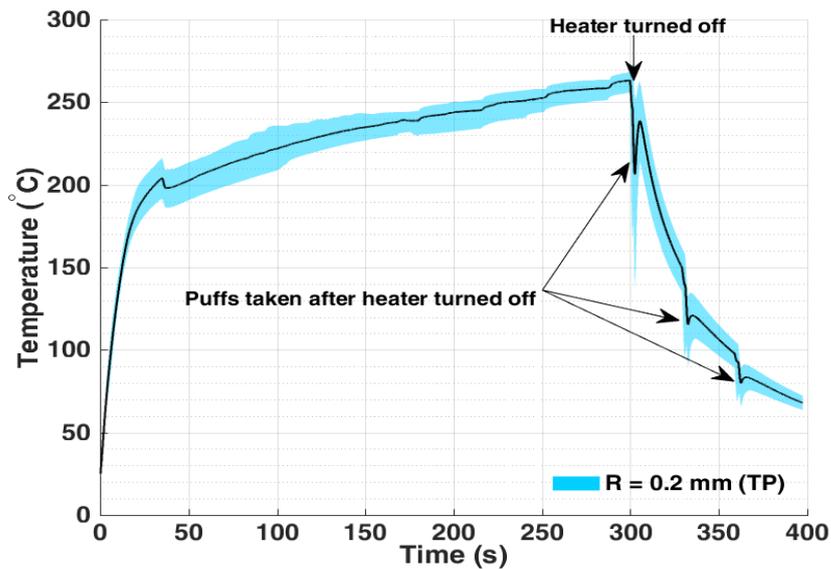
(c)

Figure 8. Temperatures measured in the EHTP during use under the HCI machine-puffing protocol (55 mL puff volume, two-second puff duration, 30-second puff interval) (a) at different radial distances (R) from the Heater surface together with the programmed Heater profile, and (b) at R=0.2 mm, when the Heater was switched off after the 10th puff. (c) No air inflow was allowed up to 300 seconds (the time when the Heater was switched off); after 300 seconds, the HCI puffing regimen was applied. The solid black lines represent the average of five replicates, and the colored region indicates the range between the minimum and maximum temperatures of the individual replicates. Data acquisition was stopped after 390 seconds.

Figure 8(b) shows the results of a similar set of experiments, in which the Heater is switched off after 300 seconds. Data acquisition was stopped after a total time of 390 seconds. The average temperature of the tobacco material recorded 0.2 mm from the surface of the Heater (based on five replicates) is shown in Figure 8(b). As the Heater was switched off, there was no recovery in the temperature of the system, and as time passed, and when additional air inflows were started ("puffs taken"), the decrease in temperature of the tobacco material accelerated.

In Figure 8(c), the results of experiments in which no air was drawn through the EHTP until 300 seconds since the start of the run (the time at which the Heater was switched off) are shown for comparison. As there is no air drawn through the EHTP, there is a continual slow rise in the temperature of the tobacco material, and no troughs in the temperature profile are present before 300 seconds. When the Heater was switched off and air was drawn through EHTP for the first time, there was a significant decrease in the temperature of the tobacco material.

The temperature in the EHTP during use has also been measured by independent external research groups [74, 75]. As shown in Figure 9, the measured temperature of the tobacco material in the EHTP (reported in Figure 8) is confirmed by independent research groups during simulated use with and without puffing.

(a)

(b)
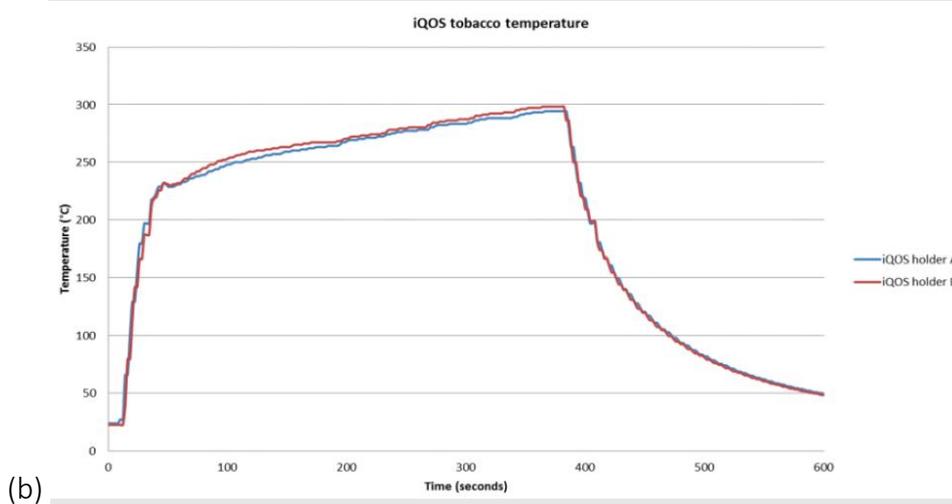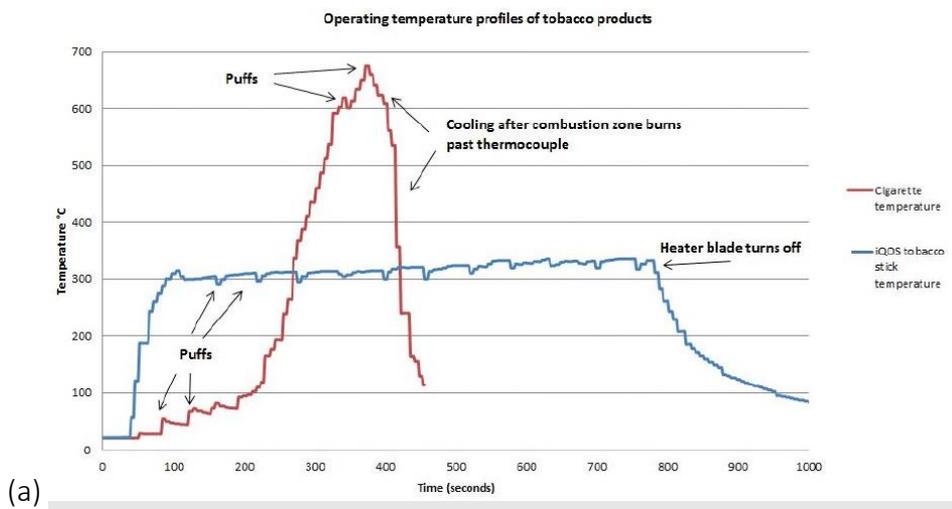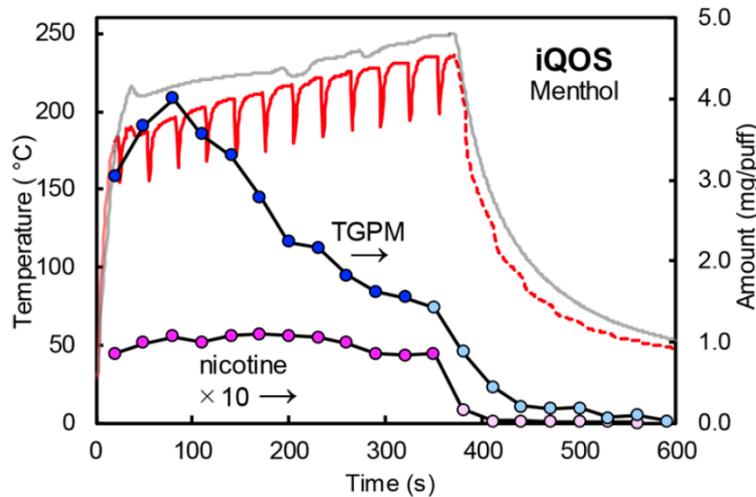
(c)

Figure 9. (a) EHTP (referred to as iQOS in the legend) and cigarette operating temperatures, with puffs [74], and (b) EHTP (referred to as iQOS in the legend) operating temperatures, without puffs, for two Holders (A and B) [74]. (c) Changes in the tobacco temperature in the EHTP (referred to as iQOS Menthol in the legend) during simulated use. Light-colored red line and broken red line indicate preheating and after puffing, respectively. Pale gray line indicates the temperature change without puffing. The generation of nicotine and total gaseous and particulate matter (TGPM) at each puff are also shown [75].

6.3   Chemical analysis of emissions

As shown in the previous section, the maximum temperature measured in the EHTP tobacco material when used as intended in the EHTS Holder is 320°C (at the Heater surface) (Figure 8(a)), and most of the tobacco material is at much lower temperatures. This temperature is lower than the ignition temperature of the tobacco material and much lower than the temperatures of the lit part of a burning cigarette, which reaches temperatures above 850°C [50]. As vaporization and thermal decomposition processes are strongly temperature-dependent, related emission of HPHCs, mostly formed by the latter process, is also related to the temperature of the tobacco material. It is widely accepted that the formation of HPHCs is reduced by decreasing the temperature, and that the main reduction is achieved when avoiding temperatures related to combustion [27, 58].

Aerosol chemistry results generated under the HCI puffing regimen for the EHTP in comparison with the mainstream smoke of a 3R4F reference cigarette were reported by [4] for 58 compounds, which included 54 HPHCs. This list includes all of the prioritized HPHCs listed by regulatory authorities, such as the abbreviated list from the U.S. Food and Drug Administration (FDA), Health Canada, and the World Health Organization. The quantification of the 58 constituents was performed in compliance with published international standards and practices, and all aerosols and smoke samples were generated using the HCI puffing regimen. The results show that for the majority of the analyzed HPHCs, their levels were reduced by >90% in the EHTS aerosol compared with the smoke from a 3R4F reference cigarette [4]. The same level of reduction was also observed when all compounds specified in the complete list of 93 HPHCs established by U.S. FDA [76] were considered (90.5% reduction, on average [77])

as well as for different tobacco blends [3] and when compared with the mainstream smoke from commercially available cigarettes [72]. The compositions of 3R4F reference cigarette smoke and EHTS aerosol are very different, as most of the EHTS aerosol mass is attributed to water, whereas for the 3R4F reference cigarette smoke, the largest fraction is comprised of compounds other than water, glycerol, and nicotine, referred to as the "Others" fraction in Figure 10. The composition of the "Others" fraction for the collected EHTS aerosol has been identified and was reported by [78, 79]. The difference between 3R4F reference cigarette smoke and EHTS aerosol becomes evident when visually inspecting Cambridge filters, onto which the smoke, or aerosol particulate phases, were collected during use (Figure 10). The collected 3R4F reference cigarette smoke strongly discolored the Cambridge filter, whereas only a slight discoloration is visible for the collected EHTS aerosol. The difference in the composition of 3R4F reference cigarette smoke and EHTS aerosol is also evident when analyzing the relative differences in the levels of HPHCs. For example, the formation of aldehydes, such as acetaldehyde, formaldehyde, and acrolein, has been assessed for the EHTS aerosol, with reductions in levels of emissions of 86.6%, 90.6%, and 93.8%, respectively, compared with the 3R4F reference cigarette smoke. As these HPHCs are typically formed during thermal decomposition, the reduced levels of these HPHCs in the EHTS aerosol compared with 3R4F reference cigarette smoke indicate that there is substantially reduced thermal decomposition (pyrolysis) activity in the EHTP tobacco material compared with the burning and high-temperature pyrolysis occurring in a lit cigarette. Similar levels of reduction of HPHCs reported by [4] in the EHTS aerosol compared with cigarette smoke have also been reported by independent studies on EHTS aerosol chemistry [2, 80-82].

As the EHTS aerosol chemistry analyzed by [2-4, 72, 80-83] focused on HPHCs, an in-depth comparative analysis of the composition of the mainstream EHTS aerosol and 3R4F reference mainstream cigarette smoke was also conducted using a non-targeted differential screening approach, combining liquid and gas chromatography coupled with high-resolution mass spectrometry, to establish a comprehensive overview of all compounds present in the EHTS aerosol during use in the EHTS Holder [78, 79, 84]. The overall results confirm that heating rather than burning tobacco results in a significant reduction in the number of compounds (750 peaks with abundance >100 ng/stick for the EHTS aerosol, representing 532 compounds [79], versus approximately 4,330 peaks for 3R4F reference cigarette smoke). The significantly reduced complexity of EHTS aerosol compared with cigarette smoke from a burning cigarette further confirms that avoiding combustion generates an aerosol that is very different from cigarette smoke.

A systematic identification of all compounds detected in the mainstream EHTS aerosol was carried out, leading to an unambiguous identification of about 99% of the aerosol mass [78, 79]. Out of the constituents detected in the EHTS aerosol, more than 97.8% w/w of the aerosol mass (based on the 50 most abundant compounds found in the EHTS aerosol [79] and the total particulate matter (TPM) reported by [85]) is comprised of compounds that are known to be present in the EHTP tobacco material from the tobacco [6], added flavors, aerosol former, etc. Therefore, less than 2.2% w/w of the EHTS aerosol mass is estimated to come from thermal decomposition of the tobacco material by torrefaction and low-temperature pyrolysis processes. This low amount of pyrolysates detected in the EHTS aerosol indicates that very limited low-temperature pyrolysis occurs in the EHTP tobacco material during EHTS operation.

In contrast, smoke resulting from burning tobacco has a very different composition, where a large portion of the smoke mass is formed by high-temperature thermal decomposition of tobacco constituents [86].
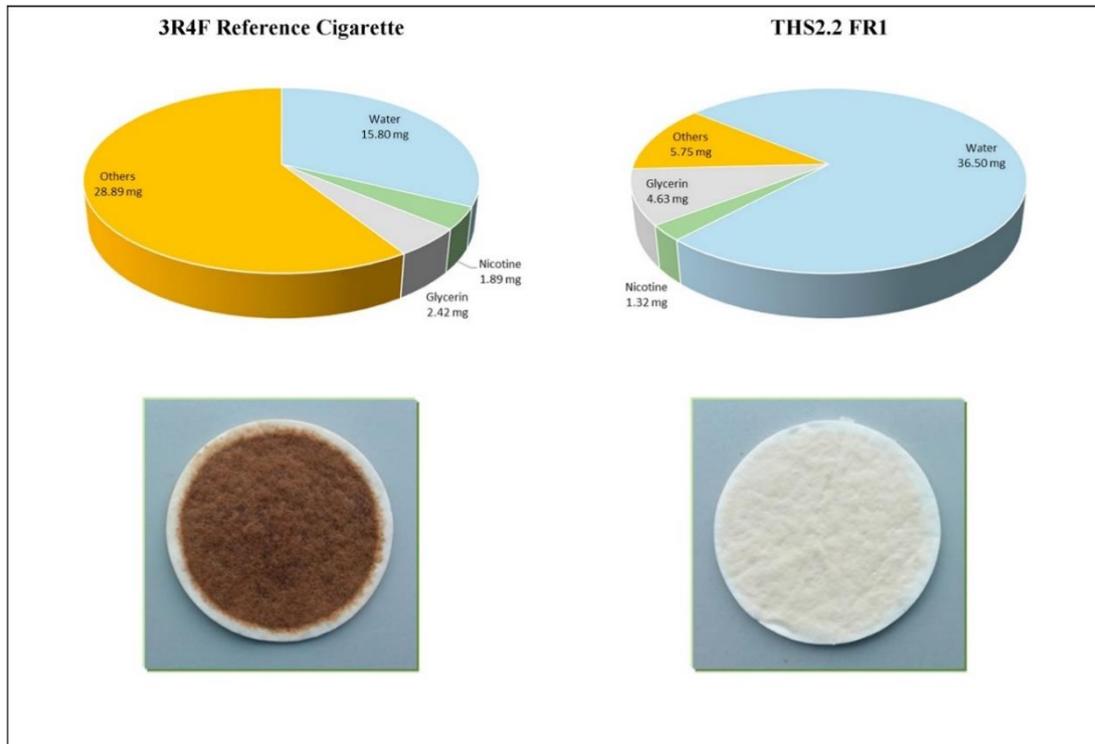
Figure 10. Cigarette TPM compared with EHTP aerosol (referred to as THS 2.2 FR1 in the figure, where FR1 is a tobacco blend) composition generated according to the HCI machine-smoking conditions for measuring emissions. The photographs of the Cambridge glass-fiber filter pads after the collection of cigarette smoke (left) and EHTP aerosol (right) are also shown [4].

As mentioned in Section 5.2.2, the mere presence of low amounts of pyrolysates in an aerosol does not imply that the aerosol is smoke, as the EHTS aerosol droplets are formed by glycerol vapors (evaporated from the tobacco material) nucleating when being cooled down and becoming supersaturated, in the same way as for the aerosol formation from EVPs.

6.4   Experiments in air (oxidative) and nitrogen (non-oxidative) atmospheres

As an additional verification that there is no combustion in the EHTP during use, the EHTS was operated in a chamber filled with air and in a chamber filled only with nitrogen, where one of the essential elements needed for combustion (oxygen) was absent [31]. Table 1 reports the composition of the EHTS mainstream aerosol [1]. The EHTS mainstream aerosol generated is composed mainly of water and of products derived from the vaporization/direct transfer of species present in the multicomponent EHTP (up to 97.8% by weight). The total amount of water reported in Table 1 is comparable under both oxidative and non-oxidative atmospheres,

suggesting that no or very limited oxidation phenomena are responsible for the presence of water in EHTS aerosol. As glycerol is added to the EHTP tobacco material to act as an aerosol former, and nicotine is naturally present in tobacco, the presence of these species in the EHTS aerosol is due to vaporization processes. While CO and CO_2 are not unambiguous markers of combustion reactions (due to their formation also from lower-temperature thermal decomposition processes [12, 17, 28, 58, 87] as described in Section 5.1.3), the levels of NO_x in the gas emissions can be used to provide evidence of the occurrence of exothermic reactions in the gas phase or in the tobacco material. Operating the EHTS under both oxidative (air) and non-oxidative (nitrogen) atmospheres had very limited influence on the aerosol constituents detected in the EHTS aerosol, as shown in Table 1. The amounts of NO_x formed under both oxidative (air) and non-oxidative (nitrogen) atmospheres under the HCI puffing regimen were 19.5 ± 1.0 µg/EHTP and 20.8 ± 1.4 µg/EHTP, respectively. These yields are comparable and are small in absolute terms with respect to the total amount of nitrogen in the tobacco material (2.02% w/w) and the level of nitrates (approximately 0.1% w/w) naturally present in the EHTP tobacco material, as shown in Table 2. The quantity of NO_x detected is also limited with respect to the amount of CO and CO_2 detected (0.5 mg/EHTP and 6.1 mg/EHTP, respectively (Table 3)) in the EHTS aerosol, suggesting that exothermic reactions in the gas phase due to the oxidation of released compounds with air can be excluded. The low levels of NO_x reported in Table 1 also support the absence of flaming phenomena or high-temperature spots occurring in the gas phase or in the tobacco material in the EHTP, which is consistent with the observations of the measured temperatures discussed in Section 6.2.

With respect to higher-molecular weight compounds present in the EHTS aerosol, Table 1 shows that limited differences are present among the yields obtained in air and in nitrogen. This suggests that the compounds may be present in the aerosol due to stripping caused by the gas stream (this is the case of plasticizers used in, for example, mouth piece filters) or may be formed by low-temperature/mild pyrolysis processes (torrefaction).

The amount of NO_x (537 ± 43 µg/3R4F cigarette) formed via the smoldering combustion of the tobacco in the 3R4F reference cigarette [4] is approximately 25 times higher with respect to the values reported in Table 1 for the EHTP. Similar differences, or even greater, were observed for CO, aldehydes, amines, and aromatic and PAH species [4]. The lower amounts of NO_x as well as the far lower amounts of CO, CO_2, aromatic compounds, aldehydes, and amines present in the EHTS aerosol compared with the levels found in 3R4F reference cigarette smoke are consistent with the lower operational temperatures reported in Figure 8 for the tobacco material compared with the temperature at the lit end of a burning cigarette (>850°C) [50]. The presence of low levels of NO_x in the EHTS mainstream aerosol, unaffected by operating the EHTS under oxidative (air) and non-oxidative (nitrogen) atmospheres, can be attributed to the thermal decomposition of nitrate salts originally present in the tobacco material [88]. A strong correlation between the nitrate content of different tobacco blends that were heated and the resulting NO_x determined in the aerosol was reported by [3]. This finding, coupled with the absence of self-sustaining exothermic processes occurring in the tobacco material of the EHTP during use, confirms the absence of combustion processes in the EHTP. The low levels of volatile organic compounds in EHTS aerosol, the presence of unconverted fixed carbon in the EHTP after heating (Table 2), and the absence of visual ash residue provide additional evidence

of the lower-temperature processes occurring in the EHTP tobacco material compared with tobacco undergoing smoldering combustion in a burning cigarette.

Table 1. Levels of aerosol constituents[1] obtained when operating the EHTP in air and in nitrogen [1, 89].

Constituents	unit	EHTP in Nitrogen		EHTP in Synthetic Air		Blank	
		Average	Std. Dev.	Average	Std. Dev.	Average	Std. Dev.
TPM*	mg/unit	54.0	1.5	55.2	1.6	0.006	0.015
Water	mg/unit	34.7	4.3	37.3	3.8	BDL	BDL
NFDPM†	mg/unit	17.9	3.1	16.5	3.4	BDL	BDL
Glycerin	mg/unit	4.38	0.24	4.39	0.40	BDL	BDL
Nicotine	mg/unit	1.38	0.10	1.37	0.09	BDL	BDL
CO	mg/unit	<0.53 but ≥0.16	----	0.54	0.16	BDL	BDL
NO	µg/unit	18.8	0.9	19.9	1.3	BDL	BDL
NOx (NO + NO$_2$)	µg/unit	19.5	1.0	20.8	1.4	BDL	BDL
Benzo[a]pyrene	ng/unit	0.60	0.09	0.61	0.11	BDL	BDL
1_3_butadiene	µg/unit	0.3	0.03	0.3	0.02	BDL	BDL
Isoprene	µg/unit	2.6	0.4	2.3	0.1	BDL	BDL
Acrylonitrile	µg/unit	0.2	0.02	0.2	0.02	BDL	BDL
Benzene	µg/unit	0.5	0.07	0.6	0.06	BDL	BDL
Toluene	µg/unit	1.9	0.3	2.0	0.2	NQ	NQ
Pyridine	µg/unit	7.4	0.6	7.8	1.4	0.2	0.1
Quinoline	µg/unit	<0.011 but ≥0.003	----	<0.011 but ≥0.003	----	BDL	BDL
Styrene	µg/unit	0.8	0.3	0.7	0.2	0.1	0.01
Hydroquinone	µg/unit	7.4	0.7	7.0	0.2	BDL	BDL
Resorcinol	µg/unit	<0.055 but ≥0.016	----	<0.055 but ≥0.016	----	BDL	BDL
Catechol	µg/unit	14.7	1.1	14.3	0.5	BDL	BDL
Phenol	µg/unit	1.3	0.1	1.4	0.1	NQ	NQ
p-cresol	µg/unit	0.07	0.01	0.07	0.01	BDL	BDL
m-cresol	µg/unit	0.03	0.01	0.03	0.01	BDL	BDL
o-cresol	µg/unit	0.07	0.01	0.06	0.01	BDL	BDL
Formaldehyde	µg/unit	6.1	1.2	9.1	1.4	1.7	0.2
Acetaldehyde	µg/unit	211	16	230	21	NQ	NQ
Acetone	µg/unit	31.0	2.3	35.9	4.3	NQ	NQ
Acrolein	µg/unit	8.4	1.3	10.7	1.7	BDL	BDL
Propionaldehyde	µg/unit	13.7	1.1	14.9	1.9	BDL	BDL
Crotonaldehyde	µg/unit	<3.29 but ≥0.988	----	<3.29 but ≥0.988	----	BDL	BDL
Methyl Ethyl Ketone	µg/unit	7.0	0.6	7.6	0.8	BDL	BDL
Butyraldehyde	µg/unit	22.5	1.9	23.1	1.9	NQ	NQ
1_aminonaphthalene	ng/unit	0.07	0.01	0.07	0.01	NQ	NQ
2_aminonaphthalene	ng/unit	0.04	0.01	0.04	0.01	0.02	0.00
3_aminobiphenyl	ng/unit	0.01	0.00	0.01	0.00	NQ	NQ
4_aminobiphenyl	ng/unit	0.02	0.00	0.02	0.00	0.01	0.01

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[1] External laboratory analysis results - *Labstat report number NS201*. *Total particulate matter. HCI regime. ISO 4387 method. †Nicotine-free dry particulate matter. BDL: Below the limit of detection (LOD), NQ: Below the limit of quantification (LOQ) - above LOD but below LOQ. HCI regime. LOQ for CO, quinoline, resorcinol, and crotonaldehyde = 0.53 mg/unit, 0.011 µg/unit, 0.055 µg/unit, and 3.29 µg/unit, respectively. Unit = EHTP.

Table 2. Elemental and water content analysis of the EHTP tobacco material and of the 3R4F reference cigarette cut filler tobacco before and after simulated use under the HCI puffing regimen in air [31].

		Tobacco weight [mg]	C [% w/w]	H [% w/w]	N [% w/w]	Water Content [% w/w]
EHTP	Before Use	323.6 ± 14.5	38.4 ± 0.3	6.1 ± 0.2	2.0	14.3 ± 0.5
	After Use	212.0 ± 10.5	46.6 ± 1.2	5.4 ± 0.3	2.6 ± 0.2	2.8 ± 0.4
3R4F Cigarette	Before Use	817.0 ± 19.6	38.2 ± 0.8	5.9 ± 0.2	1.8 ± 0.3	13.2 ± 0.4
	After Use	381.4 ± 13.7	7.6 ± 0.9	0.8 ± 0.1	1.0 ± 0.2	1.9 ± 0.2

The percentage difference in weight lost by the tobacco of the EHTP and tobacco cut filler of the 3R4F reference cigarette was 34.5% and 53.4%, respectively. The nitrate content of the tobacco material in the EHTP ranged from 0.06 – 0.11% on a dry weight basis. Average and standard deviation reported are for 8 and 3 replicates, respectively, of the weight and elemental / water content analysis.

Table 3. CO and CO_2 levels in EHTS mainstream aerosol and 3R4F reference cigarette mainstream smoke when operating in air. *Standard deviation. HCI puffing regimen. Yields of CO and CO_2 are reported by unit EHTP and 34RF reference cigarette. Average and standard deviations reported are for 9 and 19 replicates, respectively, of the 3R4F reference cigarette and EHTP [31].

	EHTP		3R4F Cigarette	
	(mg/unit)	(µg/mg of tobacco)	(mg/unit)	(µg/mg of tobacco)
CO	0.5 ± 0.1	1.7 ± 0.3	31.4 ± 1.5	38.4 ± 1.9
CO_2	6.1 ± 0.6	18.7 ± 1.7	86.2 ± 2.9	105.5 ± 3.6

On behalf of the New Zealand Ministry of Health, CRL Energy Ltd., New Zealand carried out a complimentary test to determine whether or not combustion occurs in the EHTP during EHTS operation in both air and nitrogen atmospheres [74]. As shown in Table 4, the results from the independent study show that the inhalable gas profiles are nearly identical for the EHTP when operated in the EHTS Holder under both air and nitrogen atmospheres, confirming the results presented in Table 1 [1]. This strongly indicates that there is no significant interaction with oxygen (oxidation) occurring in the EHTP during operation (i.e., there is no combustion occurring in the EHTP).

Table 4. Mainstream gaseous sample analysis for EHTS Holder (referred to as iQOS device in the table headers) with EHTP (referred to as Heet stick in the table headers) operating in air and in nitrogen, and for a cigarette operating in air [74].

Gas componet	unit	iQOS device with Heet stick operating in air	iQOS device with Heet stick operating in nitrogen	Traditional Pall Mall cigarette operating in air
Methane	(% mole/mole)	<0.0020	<0.0020	0.3752
Carbon dioxide	(% mole/mole)	0.71	0.69	5.46
Ethylene	(% mole/mole)	<0.002	<0.002	0.042
Ethane	(% mole/mole)	<0.002	<0.002	0.069
Hydrogen	(% mole/mole)	<0.002	<0.002	0.862
Oxygen	(% mole/mole)	20.42	<0.50	15.43
Nitrogen	(% mole/mole)	77.29	>99	73.36
Carbon monoxide	(% mole/mole)	0.097	0.096	2.701

6.5   Absence of solid particles

Combustion of tobacco in cigarettes generates ultrafine solid particles with a median diameter below 100 nm [90], which have been shown to be cytotoxic [91] and can cause cardiovascular and respiratory diseases as well as cancer [92]. The mainstream EHTS aerosol and 3R4F reference cigarette smoke were analyzed for the presence of solid particles by removing their volatile constituents by Pratte et al. (2017) [90]. The EHTS aerosol and 3R4F reference cigarette smoke were passed separately through a Dekati thermodenuder operating at 300°C to remove the majority of the volatile content from the EHTS aerosol and smoke particles. A thermodenuder vaporizes volatile compounds in the aerosol particulate matter to gas phase and removes the vaporized volatiles with a gas stripper, capturing most of the volatiles on an active carbon-based replaceable cartridge to largely avoid re-nucleation downstream of the instrument. Despite this, sub-30 nm nuclei particles have been reported to survive thermodenuder treatment [93, 94]. According to [95], it is difficult to distinguish whether the remaining particles are non-volatile at that temperature or are a sampling artifact, such as re-nucleation of semi-volatile compounds, without further analysis.

To avoid mischaracterization of any potential remaining semi-volatile, non-volatile compounds, or sampling artifacts as solid particles, the remaining thermally treated EHTS aerosol and 3R4F reference cigarette smoke were transferred into an impactor to collect any remaining particulate matter in the study by Pratte et al. (2017) [90]. The collected particulate matter was subsequently analyzed with an electron-dispersive x-ray examination technique using a scanning electron microscope (SEM). This analysis method enables measurements of the number of solid particles (if any), visualization of their sizes, and determination of their elemental composition. As shown in Figure 11, the analysis of the collected particulate matter revealed that the EHTP, when used as intended in the EHTS Holder, does not generate solid particles, as no solid particles were accumulated from the thermodenuded mainstream EHTS

aerosol (Figure 11(b)) in comparison with the blank Figure 11 (left), whereas 3R4F reference cigarette smoke does (Figure 11(a)). Approximately 10^{12} solid particles per 3R4F cigarette were quantified in the mainstream smoke [90]. The elemental composition of the deposited solid particles from the 3R4F reference cigarette mainstream smoke consisted mainly of carbon-based material with oxygen, but potassium and chlorine were also found, as well as traces of aluminum, sulfur, and silicon, likely released from the experimental setup.
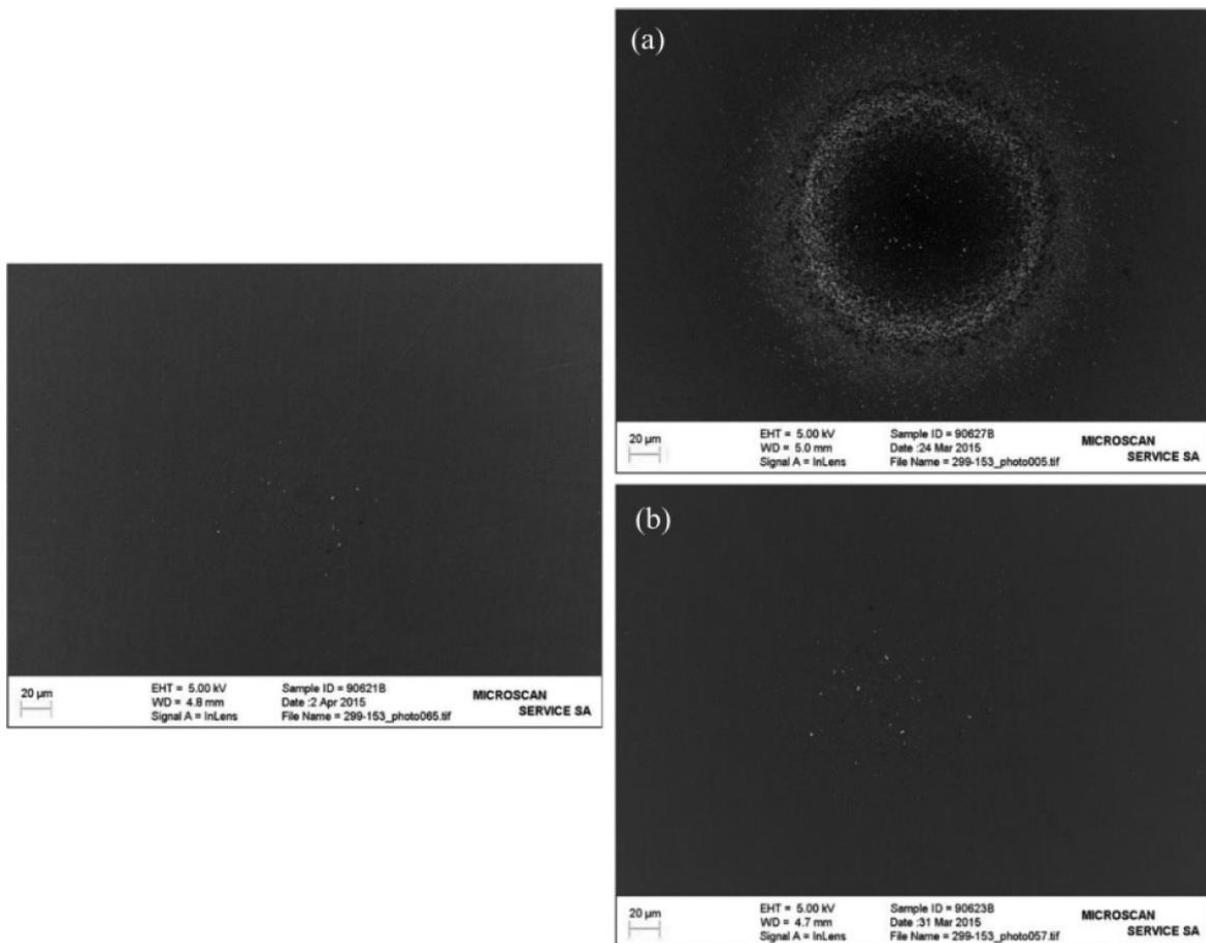
Figure 11. Pictures showing accumulated submicron particles from (a) mainstream 3R4F smoke and (b) EHTS aerosol. The picture on the left side is the related blank prior to the start of aerosol collection [90].

In a follow-up study [96], the thermodenuder technique described by [90] was used to further analyze the mainstream 3R4F reference cigarette smoke and EHTS aerosol. The EHTS aerosol and cigarette smoke were generated in separate experiments using a programmable dual syringe pump (under the HCl puffing regimen). The results from the experiments are shown in Figure 12, where the first series of peaks is attributed to the number of particulate matter objects counted for 3R4F reference cigarette smoke (black curve) and EHTS aerosol (black-dotted) when the thermodenuder was set at 30°C. The second series of peaks corresponds to

similar experiments performed when the thermodenuder was operated at 300°C. For the 3R4F reference cigarette smoke, the remaining peaks after the thermal treatment (the second series of peaks in Figure 12) were well above the lower limit of quantification (LLOQ), meaning that solid particles or high-boiling point droplets were still present in the thermally treated smoke. In the case of the EHTS aerosol, the remaining peaks overlapped with the LLOQ ± 10%. These peaks may be associated with semi-volatile compounds contained in sub-30 nm particles surviving the thermodenuder treatment, as reported by [93-95] as a limitation of the thermodenuder technique. According to [95], it is difficult to distinguish whether the remaining peaks represent non-volatile particles at that operating temperature of the thermodenuder or are a sampling artifact, such as re-nucleation of semi-volatile compounds.
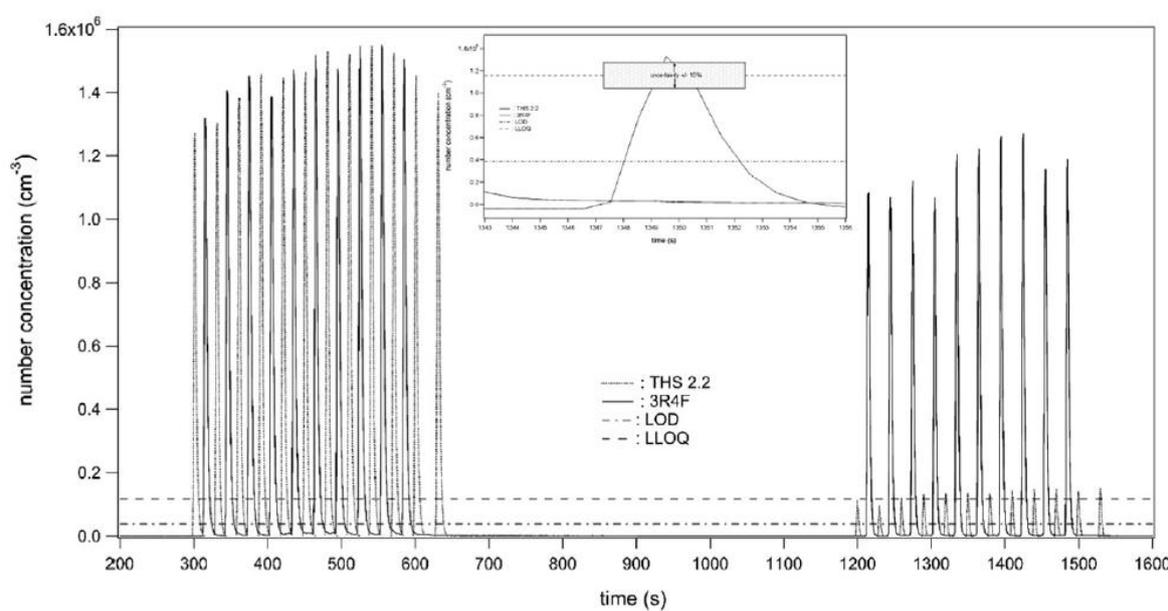
Figure 12. Typical example of measured puff profile for mainstream aerosol number concentration. The black curve corresponds to the particle number concentration for 3R4F mainstream smoke over 10 puffs, and the grey-dotted curve corresponds to the mainstream EHTS aerosol over 12 puffs. The dashed-dotted and dashed horizontal lines represent the limit of detection and the LLOQ, respectively. The insert represents a zoom of an EHTS peak, where the grey box corresponds to the uncertainty of the Condensation Particle Counter (10% according to the supplier of the instrument, for aerosol concentrations $<5 \times 10^4$ particles cm^{-3}) [96].

An external study aiming at evaluating the volatile and non-volatile fraction of particles in EHTS mainstream aerosol using a thermodilution technique was carried out by Pacitto et al. (2018) [97]. Data on particle number distribution mode (Figure 13(a)) and particle number concentration (Figure 13(b)) measured in the mainstream EHTS aerosol, as a function of the different thermodilution temperatures (from 37°C to 300°C), are shown in Figure 13. The size of the aerosol droplets decreased as the thermodilution temperature increased. Figure 13(a) clearly shows that the mode (representing the droplet size) decreased from roughly 100 nm at

37°C to less than 20 nm at 300°C. According to [95], thermodilution is considered to be efficient in removing volatile and semi-volatile particles larger than 23 nm but has been reported to be inefficient for smaller particulates. As thermodilution does not comprise a gas stripper to remove the gaseous phase after the thermal treatment (as a thermodenuder does [90, 95]), it has been reported by several research teams ([98-101]) that this leads to remaining particulate matter smaller than 23 nm in size. The gas stripper in the thermodenuder technique is known to increase the sensitivity of the measurements over techniques such as thermodilution [95], as re-nucleation of semi-volatile particles downstream in thermodilution is reduced.

A similar thermodilution system as the one used by [97] was also applied to analyze the volatility of aerosols generated by EVPs [102]. As shown in Figure 14, the mainstream aerosol emitted from EVPs are composed by a large number of volatile droplets. When the EVP aerosols passed through the thermodilution system (from 37°C to 300°C), the mode of the mainstream EVP aerosol decreased by about 24 nm to a final mode of about 10 nm, while the particle concentration decreased about 30%. Similarly to the results from the study by Pacitto et al. (2018) [97] on the thermodilution of the EHTS aerosol and by Pratte et al. (2018) [96] using the thermodenuder technique, a residual peak was visible (at about 10 nm).

As concluded by [95], it is difficult to distinguish whether the remaining particulate matter in studies like [96, 97, 102] (using thermodenuder or thermodilution techniques alone) is actually non-volatile at the thermal treatment temperature or a sampling artifact, such as re-nucleation of semi-volatile compounds. Moreover, the droplets of the EHTS aerosol contain organic acids naturally found in plants, such as palmitic acid (Hexadecanoic acid CAS: 628-97-7) [78] (boiling point 341°C for the pure compound), which is most likely a liquid phase constituent of the remaining particulate matter even after thermal treatment. As the particulate matter is a complex multicomponent mixture including many trace compounds of very low quantities, determining the physical state of specific trace compounds in the mixture based on their individual thermophysical properties is impossible, especially as these compounds may dissolve in the liquid droplets. Therefore, to ultimately determine the presence of solid particles and the content of the thermally treated EHTS aerosol, these thermal treatment techniques need to be complemented by, for example, SEM analysis, as was carried out by Pratte et al. (2017) [90] to demonstrate that the EHTP, when used as intended in the EHTS, does not generate solid particles.
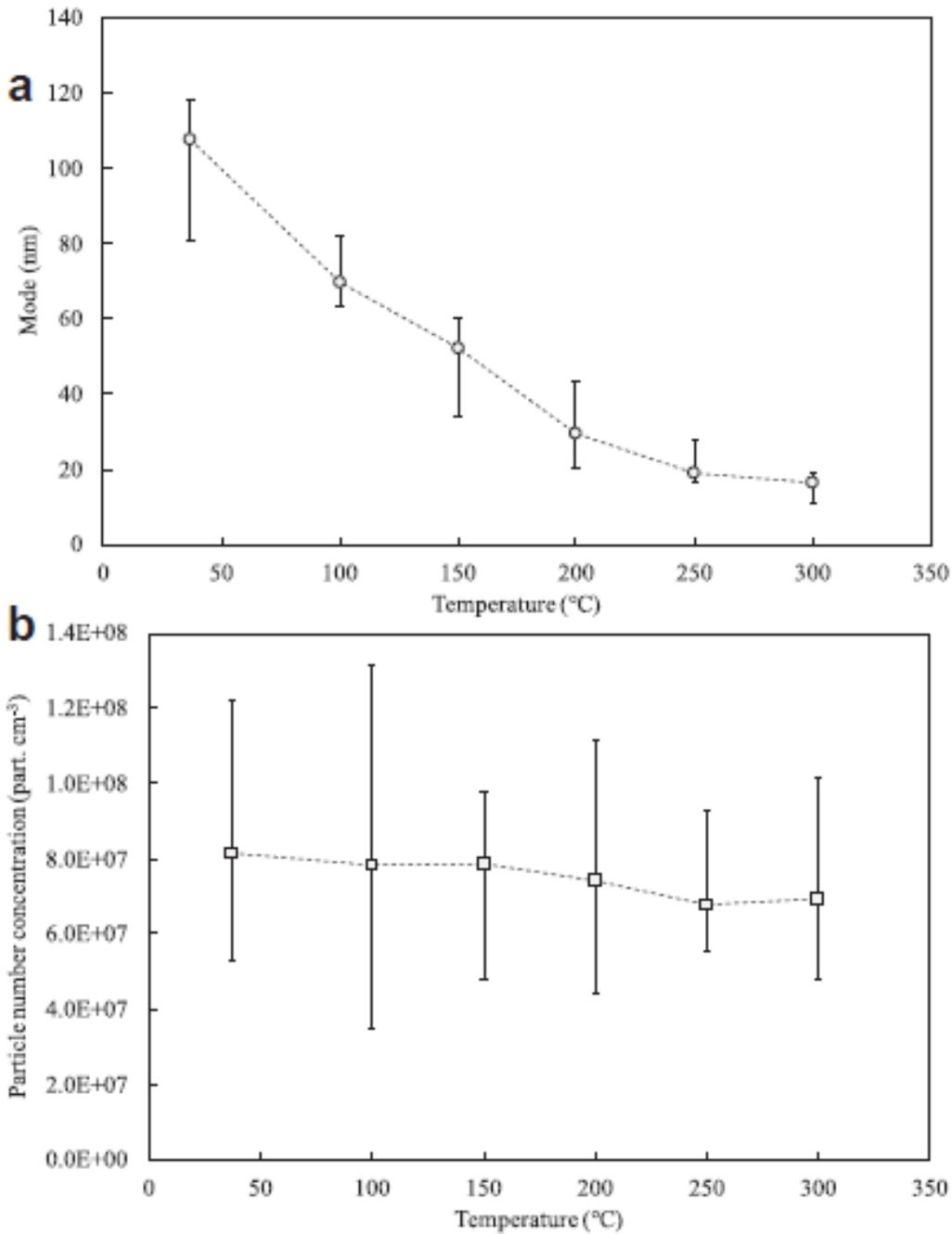
Figure 13. Trends of (a) particle number distribution mode and (b) particle number concentrations as a function of the thermodilution temperature (from 37°C to 300°C) for all EHTPs tested. Data are reported as median, 5th, and 95th percentile values [97].
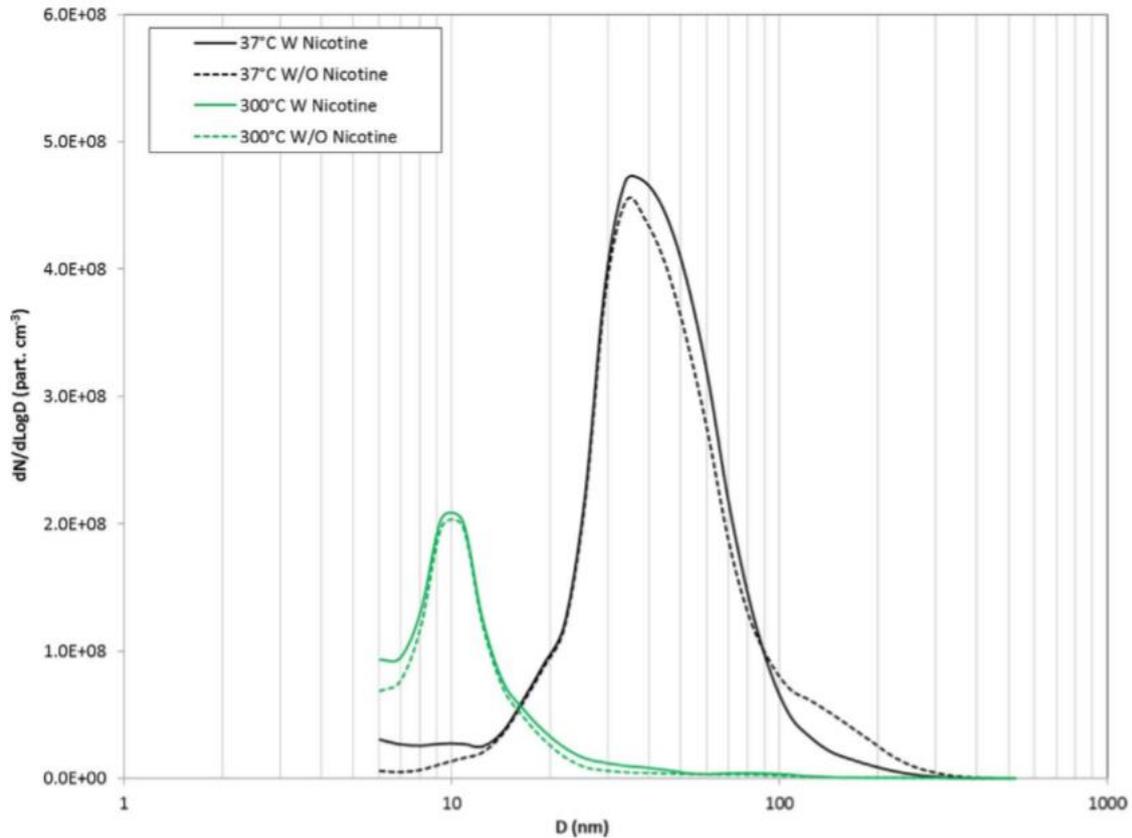
Figure 14. Particle size distributions measured in the mainstream aerosol of an EVP with and without nicotine at 37°C and 300°C [102].

6.6   Absence of ash residue

When EHTPs are heated in the EHTS Holder, they are not consumed by combustion processes, no ash residue is visible, and they retain their structural integrity, as shown in Figure 15. A visible darkening of the tobacco material is only observed in the vicinity of the Heater (heating blade) as a result torrefaction and low-temperature pyrolysis of the tobacco material at temperatures below 320°C, well below the ignition temperature [31]. The observed change in color of the tobacco material close to the Heater is similar to the color change of the tobacco material observed in the TGA experiments in both the nitrogen atmosphere (Figure 5(a)) (where no combustion can occur) and an air atmosphere (Figure 6(a)). The observed darkening of the heated tobacco material in the vicinity of the Heater is typical of the color change resulting from torrefaction (mild pyrolysis) processes [10, 24, 31]) and is also observed in other torrefaction processes, such as the roasting of coffee beans or toasting bread.
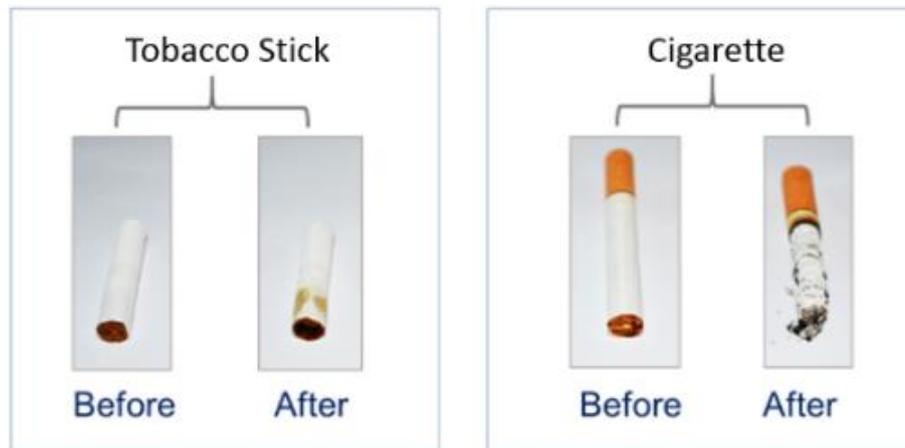
Figure 15. Photographs of EHTPs (Tobacco Stick) and cigarettes before and after use.

7   Third-party reports validating the absence of combustion and smoke

The absence of combustion in the EHTP tobacco material during EHTS use and that the aerosol generated is not smoke have been validated by leading scientific experts in the fields of combustion, fire safety, and thermochemistry from numerous countries, including Italy, the United Kingdom, Japan, Poland, the United States, Australia, Germany, and Switzerland, as well as by an independent research organization in New Zealand. In this section, their expert opinions and conclusions are summarized.

7.1   Professor Valerio Cozzani, Italy.

Dr. Valerio Cozzani, Professor of Chemical Engineering at the University of Bologna, Italy, is an expert in combustion (burning) and pyrolysis (thermal breakdown) of biomass (materials obtained from plants).

PMI retained Professor Cozzani in an independent capacity to review and analyze scientific data on the thermal processes taking place in the EHTS during aerosol generation and to propose additional experiments he considered essential to be able to conclude whether or not there is combustion in the EHTP during intended use and if the aerosol generated is smoke [103].

Professor Cozzani concluded that: "On the basis of the analysis of the technical details and of the experimental data provided in the PMI report, I can exclude that a combustion process (including self-sustained combustion) takes place in the tobacco substrate of the EHTP during EHTS operation" [103]. The report referred to in the cited conclusion is the PMI report [104], which includes assessment data and analysis on the EHTP. His conclusion was based on the following findings from his assessment:

- "temperature profiles reported for the EHTP tobacco substrate during EHTS operation, that suggest that temperatures are far below those of self-sustained combustion of tobacco" [103]
- "no clear evidence of an exothermic process in the EHTP, as may be deduced by the temperature decrease during simulated puffing" [103]

- "limited amounts of NO_x formed during EHTS operation in comparison with CO. The amounts of NO_x formed are independent of the presence of oxygen, suggesting that their formation is governed by nitrate decomposition" [103]
- "Several other evidences of low-temperature processes during operation of the EHTS (very low amounts of high-molecular weight polyaromatic compounds formed, presence of unconverted fixed carbon in the EHTP residue, etc.)" [103]

Professor Cozzani further concluded that: "Even applying the more comprehensive definitions of smoke […], I can conclude that EHTS operation does not produce smoke" [103]. His conclusion was based on the following findings from his assessment:

- "the absence of combustion processes in the EHTP tobacco substrate" [103]
- "the very limited pyrolysis phenomena present in the EHTP tobacco substrate during EHTS operation, that do not have a relevant influence on the aerosol formed. Less than 2% by weight of the aerosol components may derive from the pyrolysis of the EHTP tobacco substrate" [103]
- "the aerosol produced by EHTS operation is formed mostly by vaporization, as proven by the experimental data showing its chemical characterization" [103]
- "The aerosol generated in EHTS operation is very different in the chemical composition from the smoke formed in the self-sustained combustion of tobacco and more in general from smoke formed in combustion processes. Therefore, such aerosol should not be classified as smoke" [103]

7.2 Professor Guillermo Rein, United Kingdom

Dr. Guillermo Rein is Professor of Fire Science at the Department of Mechanical Engineering at Imperial College London, United Kingdom. He is an internationally recognized leader in fire safety and combustion research, Editor-in-Chief of the peer-reviewed scientific journal *Fire Technology* (the official research journal of the U.S. National Fire Protection Association (NFPA) and the Society of Fire Protection Engineers), and an expert in the field of heat and mass transfer. His work covers a wide area of combustion research in built and natural environments, including ignition, fire spreading, wildfires, structural fire resistance, and smoldering combustion of biofuels.

PMI retained Professor Rein to review and analyze scientific data on the thermal processes taking place in the EHTS during aerosol generation and to conduct additional experiments he considered essential to be able to conclude whether or not there is combustion in the EHTP during intended use and if the aerosol generated is smoke [105].

Combining the experimental evidence he obtained in his own laboratory on how the EHTS device functions and operates [106] with his expert knowledge of combustion science, Professor Rein concluded that:

- "the EHTS can be considered to operate in a non-combustion regime: it does not ignite or burn the tobacco. It operates in a thermal regime that only allows for drying, evaporation and low temperature pyrolysis reactions of the tobacco" [105]
- "the airborne substances released by EHTS are not smoke because there is no combustion" [105]

His conclusions were based on the following findings from his assessment:

- "the peak temperature in the EHTS tobacco is too low to ignite the combustion process" [105]
- "there is no propagation or sign of any exothermic reaction in the tobacco during EHTS normal operation" [105]
- "ash is not formed during EHTS normal operation, and therefore combustion reactions are not occurring" [105]

7.3   Professor Osamu Fujita, Japan

Professor Fujita is a faculty member of the Division of Mechanical and Space Engineering at Hokkaido University, Japan. He is an expert in the field of combustion and an international leader in microgravity combustion research. His work covers a wide area of combustion research, including ignition and flame spreading, catalytic combustion, soot formation, high-temperature combustion, combustion in microgravity, electric and magnetic field effects on combustion, biofuels, and combustion diagnostics. He is the President of the Combustion Society of Japan and a member of the board of directors of the Combustion Institute.

PMI retained Professor Fujita to review and analyze scientific data on the thermal processes taking place in the EHTS during aerosol generation and to propose additional experiments he considered essential to be able to conclude whether or not there is combustion in the EHTP during intended use and if the aerosol generated is smoke [107].

Professor Fujita concluded that:

- "Based on the product data given by PMI, combustion phenomena do not take place in the Electrically Heated Tobacco Product" [107]
- "Based on the data given by PMI, we can conclude that EHTP generates no smoke but just aerosol as long as EHTP is used as intended and heated in the Holder" [107]

The data given by PMI relates to the data presented in the PMI report [104]. Professor Fujita's conclusions were based on the following findings from his assessment:

- "There is no ignition of the tobacco material in the EHTP" [107]
- "Results from aerosol formation simulations [70] which show that aerosol droplets are formed only in the presence of an aerosol former being mainly glycerol, which is evaporated from the tobacco and is not a product of pyrolysis or combustion."

7.4   Professor Pawel Gierycz, Poland

Professor Pawel Gierycz is Professor of Chemical and Process Engineering at the Warsaw University of Technology, Poland. He is an expert in the thermal analysis and characterization of materials, including chemical thermodynamics and thermochemistry; chemical, process, and environmental engineering; and physical chemistry.

PMI retained Professor Gierycz to review and analyze scientific data on the thermal processes taking place in the EHTS during aerosol generation and to propose additional experiments he considered essential to be able to conclude whether or not there is combustion in the EHTP during intended use and if the aerosol generated is smoke [108].

Professor Gierycz concluded that:

- "there is no combustion of tobacco during the heating of tobacco in the new EHTS device" [108]
- "the aerosol is fundamentally different from the tobacco smoke" [108]

His conclusions were based on the following findings from his assessment:

- "Maximum temperature obtained in this device is so low that the process of burning of tobacco cannot be initiated, and the more it cannot be self-sustained" [108]
- "During the heating of tobacco inside the EHTS device (EHTP) the main process is a distillation of constituents along with their thermal decomposition (on a much smaller scale than in the case of burning of cigarettes)" [108]
- "The products (aerosol, dry residue) resulting from this process are almost identical (to within the accuracy of experimental error) to products of heating of tobacco carried out under atmosphere of nitrogen" [108]
- "Product of the heating of tobacco in new EHTS device is aerosol and a solid residue structurally similar to original reconstituted tobacco. These products are fundamentally different from products of "classical" burning of tobacco in cigarettes, i.e. smoke and ash" [108]
- "For the aerosol produced as a result of using the new EHTS device, more than 98% of its constituents come from the EHTP and only approx. 1.7% of substances might be a result of chemical processes occurring during the heating process" [108]

7.5   Advanced Fuel Research, Inc., USA

Advanced Fuel Research, Inc. is a privately owned business, founded in 1980, that supplies contract research, product development, and analytical services to corporate research centers, research institutes, and government laboratories. The company provides expertise in a number of areas, including combustion monitoring and control and clean energy.

PMI retained scientific experts in combustion, pyrolysis, and torrefaction of biomass (materials obtained from plants) at Advanced Fuel Research, Inc. to review and analyze scientific data on the thermal processes taking place in the EHTS during aerosol generation and to propose additional experiments they considered essential to be able to conclude whether or not there is combustion in the EHTP during intended use and if the aerosol generated is smoke [109].

The scientific experts concluded that: "combustion does not take place in the tobacco material of the EHTP when the device is used as intended" [109], based on the following findings from their assessment:

- "The heating process is neither rapid nor self-sustaining; in fact, it is self-extinguishing, if the power to the heating device is turned off" [109]
- "The heating of the tobacco in the EHTP occurs at temperatures typical for drying/vaporization, up to the temperature range of torrefaction (200–300°C), i.e., at temperatures lower than those required for pyrolysis (> 300°C) and combustion (> 400°C)" [109]
- "the use of the EHTP does not involve the formation of appreciable amounts of high-temperature combustion markers, such as nitrogen oxides or polycyclic aromatic

hydrocarbons (PAHs), e.g., benzo[a]pyrene. Neither is ash formed when the EHTP is used as intended" [109].

They also concluded that: "there is no smoke formed in the case of EHTP use" [109], based on the following findings from their assessment:

- "since the use of the EHTS does not involve combustion or pyrolysis, it follows that there is no smoke formed in the case of EHTP use" [109]. The tobacco in the EHTP is heated to temperatures typical of torrefaction reactions (i.e., < 300°C).

- "In the case of the EHTP under the ISO puffing regime, water and glycerin constitute 91% of the aerosol, whereas the corresponding percentage for the regular cigarette is much lower (19%)" [109]

- "the smoke/aerosol constituents of the regular cigarette include appreciable amounts of such species as butadiene, isoprene, and aromatics, whereas the aerosol generated from the EHTP contains only small or trace amounts of such species. Consequently, the use of conventional cigarettes is associated with smoke formation, whereas the corresponding product of the EHTS is aerosol" [109]

- "the EHTP, when used as intended and heated in the holder, does not produce any solid particles as part of the aerosol" [109]

7.6   Professor Jose Torero, Australia

Professor José L. Torero, Australia, is an internationally recognized leader in the field of fire safety and combustion. He specializes in non-premixed combustion processes and fire behavior in complex environments, such as forests, tall buildings, novel architectures, tunnels, aircraft, and spacecraft. Professor Torero was selected as an expert witness to the Grenfell Tower Inquiry's examination of the circumstances leading up to and surrounding the fire at Grenfell Tower in London on June 14, 2017.

PMI retained Professor Torero in an independent capacity between February 2017 and October 2017 to assess whether combustion of the EHTP tobacco material occurs during product use and if discarding a used EHTP in a waste bin or inappropriately discarding an EHTP in grassland or forest pose a fire risk. Professor Torero was also asked to identify additional experiments he considered essential to be able to opine on the topics [15].

Based on his expert knowledge of combustion science and fire safety, his analysis of the detailed thermochemical data for the EHTP tobacco material, and the experimental and numerical evidence on how the EHTS operates, Professor Torero concluded that:

- "in the present configuration of the PMI-EHTP, when operated as intended in the PMI-EHTS, the energy balance is negative and self-sustained smouldering propagation (smouldering combustion) cannot occur" [15]

- "Within the heating regimes used for the PMI-EHTS the main exothermic step of the smouldering reaction is fully avoided, thus preventing the majority of energy release and the generation of combustion products" [15]

- "the PMI-EHTS does not represent a fire risk under any circumstance" [15] as "the maximum operation temperature reported does not exceed 320°C" [15] and

"[m]aterials susceptible to fire initiation will represent significant heat sinks and their ignition temperatures are always higher than 320°C" [15]

7.7 Technical University of Munich, Germany

The Wood Research Laboratory at the Technical University of Munich (TUM), Germany, is accredited and holds state recognition as a testing center for the combustion properties of building materials. Mr. Rupert Ehrlenspiel, Dipl.-Ing., is the Director of Fire Testing and Research and of the Testing, Monitoring, and Certification Office at the TUM. Mr. Ehrlenspiel is an expert for the fire properties of building materials. Mr. Christoph Gruber, Dipl.-Ing., was a staff member in the Testing, Monitoring, and Certification Office at the TUM until 2017 and provided services in fire testing and product certification.

PMI retained Mr. Ehrlenspiel and Mr. Gruber to review and analyze scientific data on the thermal processes taking place in the EHTS during aerosol generation and to propose additional experiments they considered essential to be able to conclude whether or not there is combustion in the EHTP during intended use and if the aerosol generated is smoke [110].

Mr. Ehrlenspiel and Mr. Gruber concluded that:

- "In the case of EHTP, no combustion occurs" [110], as "neither ignition nor a self-sustaining process take place, i.e. no exothermic reaction in aggregate. However, the closer the tobacco is to the heating unit, the more drying, thermal decomposition, and embrittlement take place" [110]
- "the absence of solid particles in the EHTS aerosol was demonstrated. Thus, based on the fact that no combustion was found to be taking place in the tobacco, the authors infer that the EHTP does not release smoke, but rather a mixture of gas and liquid droplets (vapor)" [110].

7.8 Professor Thomas Nussbaumer, Switzerland

Professor Dr. Thomas Nussbaumer, Verenum Research, Zurich, Switzerland, is an expert in biomass combustion, thermochemical processes for biomass utilization, and pollutant formation and emission reductions from biomass combustion. He has been a representative in the International Energy Agency's Bioenergy Task 32 (Biomass Combustion) for the Swiss Federal Office of Energy since 1991 and he is the vice president of the Wood Energy Switzerland association (Holzenergie Schweiz).

PMI retained Professor Nussbaumer to review and analyze scientific data on the thermal processes taking place in the EHTS during aerosol generation and to provide his expert opinion on whether or not there is combustion in the EHTP during intended use and if the aerosol generated is smoke [111].

Based on his expert knowledge of biomass combustion and related emissions, Professor Dr. Nussbaumer concluded that:

- "Based on the scientific and technical attributes that define "combustion" as a thermo-chemical reaction and as a technical process, combustion can be excluded in IQOS." [111]

- "According to frequently-used definitions, smoke requires combustion as a source. Since no combustion takes place in IQOS, the formation of smoke is excluded according to these definitions." [111]
- "A central requirement for smoke is the presence of solids in the aerosol due to thermo-chemical formation. Analyses of the IQOS product stream show that this requirement is not met, and accordingly the product stream is not smoke." [111]

7.9   Professor Rainer Marutzky, Germany

Professor Dr. Rainer Marutzky, Germany, is an honored expert in wood science, including biomass processes and emissions. During his active professional career, Professor Marutzky was a former director of the Fraunhofer Institute for Wood Research, Wilhelm-Klauditz-Institut, and a professor at the Technical University of Braunschweig. During his active working career, he was heavily involvement in national, European, and international standardization committees.

PMI retained Professor Marutzky to review and analyze scientific data on the thermal processes taking place in the EHTS during aerosol generation and to provide his expert opinion on whether or not there is combustion in the EHTP during intended use and if the aerosol generated is smoke [20].

Professor Marutsky concluded that:

- "Swift and self-sustaining combustion of the Heat stick tobacco-containing material in the HEETS stick is to be excluded in IQOS operation." [20]
- "The aerosol-containing gas consumed from the IQOS system is briefly visible, but does not contain any solid particles, especially not any that are attributable to combustion or pyrolysis of the tobacco in the HEETS Stick." [20]
- "[P]yrolysis exclusively takes place in the bottom range of low-temperature pyrolysis." [20]
- "It is the opinion of this Expert that the term smoke is clearly linked with the term combustion." [20]
- "It has been evidenced that combustion processes do not take place when operating IQOS with HEETS Sticks." [20]

7.10 CRL Energy Ltd., New Zealand

CRL Energy Ltd. is an energy, minerals, and environmental research and consulting company. The Solid Fuel Laboratory of CRL Energy Ltd. is International Accreditation New Zealand-accredited for a number of typical fuel sampling and analysis methods and routinely analyzes a variety of samples, such as coal, coke, charcoal, and biomass, for a wide range of properties. The Solid Fuels Laboratory participates in a range of international round robin programs, has extensive quality control systems, and is accredited to ISO9001.

CRL Energy Ltd. was approached by the New Zealand Ministry of Health to investigate supplied EHTS Holders (referred to as iQOS devices in the report [74]) and EHTP (referred to as HEETS or tobacco stick in the report [74]) and to provide their opinion on a number of topics, including

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the physical, thermal, and combustion properties of the EHTP and emissions of CO and CO_2 (both in air and inert atmospheres) from the EHTP during use [74].

Based on their experimental assessments and analyses, CRL Energy Ltd. concluded that: "no combustion is taking place during normal operation of the iQOS system" [74] and "the HEETS tobacco product does not ignite" [74], based on the following findings from their assessments:

- "No evidence of exothermic reaction was evident in the iQOS device under normal operating conditions" [74]
- "temperature is not sufficient to cause combustion to occur in the tobacco stick" [74]
- "At the iQOS operating temperature of 350°C no combustion or significant oxidation was seen to occur in the HEETS tobacco" [74]
- "Gas profiles of the iQOS device operating under air and nitrogen atmospheres are very similar indicating no significant interaction with oxygen in air is occurring during operation" [74]

8   Conclusions

The scientific evidence summarized in this report substantiates the absence of combustion of the tobacco material in the EHTP, when used as intended in the EHTS Holder, and that the aerosol generated is not smoke. Physical and chemical processes occurring in tobacco as a function of temperature, the fundamentals behind aerosol formation in general and smoke formation in particular, and available technical and scientific definitions of smoke and combustion were reviewed. Based on these reviews, the scientific evidence comprehensively demonstrates that no combustion of the tobacco material occurs during the intended use of the EHTS and that the aerosol generated from the EHTP tobacco material is not smoke.

Key takeaways of this report include the following:

- All definitions of smoke listed in the Appendix involve combustion or burning, and in most definitions, smoke is defined as a product of combustion. A more comprehensive definition of smoke is given by the NFPA, who defines smoke as "solid and liquid particulates and gases evolved when a material undergoes pyrolysis or combustion" [18].

- Although smoke may be formed when a material undergoes pyrolysis according to the definition by [18], the mere occurrence of pyrolysis processes does not imply that smoke particulate matter is formed.

- Smoke particulate matter is formed when products of combustion and high-temperature pyrolysis (e.g., hydrocarbons) reach supersaturation and either condense to form droplets or react together to form particles, or by the nucleation and growth of positively charged hydrocarbon ions to form soot particles (non-existent in EHTS aerosol droplets).

- While smoke is an aerosol, not all aerosols are smoke. The EHTS aerosol liquid particulate matter (droplets) is not formed from condensation of byproducts of combustion or pyrolysis. The droplets are instead generated when glycerol (added to the tobacco material during processing as an aerosol former) is vaporized and reaches

supersaturation and condenses on cooling, forming nuclei, onto which more glycerol, water, nicotine, and other constituents can condense to form droplets.

- Aerosols formed from the condensation of water vapor, vaporized e-liquids, or vaporized glycerol from the EHTP tobacco material are not smoke and are very different in terms of origin and chemical and physical composition compared with smoke aerosols formed from the combustion and associated high-temperature pyrolysis products generated from the burning of tobacco.

- The aerosol generation process in the EHTS is equivalent to the aerosol generation process in most EVPs, for which aerosol formers (glycerol and propylene glycol) in the e-liquid are vaporized during heating and are subsequently cooled down to form liquid aerosol droplets.

- The EHTP, when used as intended in the EHTS Holder, does not generate solid particles, as demonstrated by detailed experiments involving SEM analysis. Thermodenuder and thermodilution techniques are not able to determine the presence of solid particles alone without being combined with microscopy analysis, such as SEM.

- The EHTP is designed to generate a nicotine-containing aerosol that is not smoke by heating tobacco to temperatures sufficient to release nicotine and flavors from the tobacco but low enough to prevent the tobacco from burning. As the tobacco material in the EHTP is heated and not burned, the levels of the majority of the analyzed HPHCs in the EHTS aerosol were reduced by >90% compared with the mainstream smoke of a 3R4F reference cigarette.

- More than 97.8% w/w of the EHTS mainstream aerosol constituents originate from the multicomponent EHTP and transfer to the aerosol via vaporization/direct transfer processes. The other components (<2.2% w/w) present in the aerosol are likely to be formed by torrefaction and low-temperature pyrolysis, as suggested by the limited influence of oxygen on the aerosol composition.

- For combustion (burning) of the EHTP tobacco material to occur, the temperature needs to exceed about 400°C. The maximum temperature measured in the EHTP during use in the EHTS Holder is 320°C. As the EHTP is heated to temperatures below the ignition temperature of the EHTP tobacco material, the tobacco material undergoes processes such as drying, vaporization, and thermal decomposition (torrefaction and low-temperature pyrolysis), but no combustion (neither incomplete nor complete).

- The comparison of the chemical composition of the EHTS aerosol generated in oxidative (air) and non-oxidative (nitrogen) environments indicated that oxygen (necessary for combustion to happen) does not play a major role in the thermal decomposition of the EHTP tobacco or the aerosol formation. If combustion were to occur in the tobacco, the amount of emissions of compounds such as CO, CO_2, and NO_x would be significantly different between the two atmospheres, which is not the case. Gaseous compounds, such as CO, CO_2, NO_x, etc. (usually associated with combustion) are also generated during thermal decomposition of tobacco components during heating, independent of the oxygen availability. The presence of low levels of gaseous compounds in the EHTS

aerosol is therefore not evidence that combustion (neither incomplete nor complete) has occurred.

- The presence of low amounts of pyrolysates in an aerosol does not imply that the aerosol is smoke, as the EHTS aerosol droplets are instead generated by glycerol vapors (evaporated from the tobacco material) nucleating when being cooled down and becoming supersaturated, in the same way as for the aerosol formation from EVPs, where the aerosol generated by EVPs also contains low amounts of pyrolysates.

- The absence of any form of combustion (incomplete and complete) in the EHTP, when used as intended in the EHTS Holder, and that the aerosol generated is not smoke have been demonstrated by scientifically substantiated evidence and have been verified by scientific experts in numerous countries as well as by independent research organizations.

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Appendix: Technical and scientific definitions

AEROSOL	
Definition	Reference
"An intimate mixture of a liquid or a solid in a gas; the liquid or solid, called the dispersed phase, is uniformly distributed in a finely divided state throughout the gas, which is the continuous phase or dispersing medium"	National Fire Protection Association. (2019) [18]
"A suspension of solid or liquid particles in a gas. […] The term aerosol includes both the particles and the suspending gas, which is usually air."	Hinds (1999) [59]
"A fluid where solid or liquid particles are dispersed in a gas phase."	Cozzani (2014) [103]
"a suspension of fine solid or liquid particles in gas"	Merriam-Webster dictionary (2020) [53]
General information	
Aerosols are generally defined as liquid or solid particles suspended in a gas according to the definitions listed above. As smoke is generally defined as solid and liquid particles suspended in a gaseous phase, smoke is, according to the definitions, an aerosol. However, while smoke is an aerosol, not all aerosols are smoke. Smoke is generally considered to be a product of combustion (and by one definition also a result of pyrolysis), whereas aerosols formed from condensation of vapors not originating from a pyrolysis or combustion process are considered to be aerosols that are not smoke [54, 56]. For example, aerosols formed from condensation of water vapor or vaporized e-liquids from EVPs (commonly known as e-vapor or vape) are very different in terms of origin as well as chemical and physical composition compared with smoke aerosols formed from combustion and associated high-temperature pyrolysis products generated from the burning of tobacco. Other examples of aerosols that are not smoke include clouds in the atmosphere and aerosols generated from humidifiers. Examples of smoke include aerosols generated from the burning of fossil fuels, biomass combustion, and the burning of tobacco in a lit cigarette.	

CARBONIZATION	
Definition	Reference
"Carbonization is a slow pyrolysis process in which biomass is converted into a highly carbonaceous, charcoal-like material"	Ronsse (2015) [112]
"Carbonization is a process in which a fuel is heated without air to leave solid porous carbon"	Collins dictionary (2020) [51]

COMBUSTION	
Definition	**Reference**
"A chemical process of oxidation that occurs at a rate fast enough to produce heat and usually light in the form of either a glow or flame."	National Fire Protection Association. (2019) [18]
"rapid oxidation generating heat, or both light and heat."	Turns (2012) [41]
"The reduction-oxidation chemical reaction where the reactant molecules, namely the fuel and the oxidant, are mixed and rearranged to become product molecules with the simultaneous release of heat"	Cozzani (2014) [103]
"The process of burning something: the combustion of fossil fuels." or "Rapid chemical combination of a substance with oxygen, involving the production of heat and light."	Oxford dictionaries (2018) – Combustion [113]
"The process of burning." or "Any process in which a substance reacts with oxygen to produce a significant rise in temperature and the emission of light."	Collins dictionary (2020) [51]
"an act or instance of burning." or "a usually rapid chemical process (such as oxidation) that produces heat and usually light."	Merriam-Webster dictionary (2020) [53]
General information	

Technical and scientific definitions define combustion as a chemical reaction between oxygen and a material (oxidation) happening at rate fast enough to produce heat (increase temperature) and usually light. Combustion (burning) is initiated by ignition and is self-sustained as long as the exothermic (heat-generating) oxidation reaction is sufficiently strong to overcome heat losses and the heat required for endothermic (heat-consuming) processes, such as vaporization and endothermic thermal decomposition. Combustion includes both complete and incomplete (partial) combustion processes [40, 41]. For complete combustion to occur, there needs to be sufficient oxygen available so that when oxygen reacts with a fuel in a complete oxidation reaction, only CO_2 and water are produced [40]. Complete combustion rarely occurs in natural combustion situations, and almost all combustion processes found in nature are incomplete combustion processes, including smoldering combustion [38, 42, 43] and most flaming combustions [40, 41]. In incomplete combustion (occurring when there is an insufficient supply of oxygen), the combustion of carbonic fuel produces incompletely oxidized carbon compounds. Combustion products produced during

incomplete combustion include water, CO, CO_2, PAHs, NO_x, polychlorinated aromatics, hexachlorobenzene, hydrogen cyanide, alcohols, and ketones [6, 38, 40-47]. Some of these products contribute to the formation of fine solid and liquid particulate matter, which together with all emitted gaseous compounds constitute smoke [6, 38, 40-47].

COMPLETE COMBUSTION	
Definition	Reference
See combustion.	
General information	

For complete combustion to occur, there needs to be sufficient oxygen available so that when oxygen reacts with a fuel in a complete oxidation reaction, only CO_2 and water are produced [40]. Complete combustion rarely occurs in natural combustion situations, and almost all combustion processes found in nature are incomplete combustion processes, including smoldering combustion [38, 42, 43] and most flaming combustions [40, 41].

DEVOLATILIZATION	
Definition	Reference
"The overall process in which solid carbonaceous substances undergo a primary pyrolysis leading to the formation of lower molecular weight products and of a solid char residue. The lower molecular weight products formed in primary pyrolysis vaporize and are transferred from the solid to the gas phase (thus these species are named "volatiles")."	Cozzani (2014) [103]
"Devolatilization is the removal of volatile substances from a solid."	Collins dictionary (2020) [51]

EVAPORATION	
Definition	Reference
"Evaporation, the process by which an element or compound transitions from its liquid state to its gaseous state below the temperature at which it boils"	Encyclopaedia Britannica (2020) [114]
"Evaporation is when a liquid changes to a vapor, caused by an increase in temperature and/or a decrease in pressure."	Collins dictionary (2020) [51]

IGNITION	
Definition	Reference

"the process of initiating self-sustained combustion" or "The initiation of combustion evidenced by glow, flame, detonation, or explosion, either sustained or transient."	National Fire Protection Association. (2019) [18]
"the act or process of initiating combustion"	Collins dictionary (2020) [51]

INCOMPLETE COMBUSTION	
Definition	**Reference**
See combustion.	
General information	
Almost all combustion processes found in nature are incomplete combustion processes, including smoldering combustion [38, 42, 43] and most flaming combustions [40, 41]. In incomplete combustion (occurring when there is an insufficient supply of oxygen), the combustion of carbonic fuel produces incompletely oxidized carbon compounds. Combustion products produced during incomplete combustion include water, CO, CO_2, PAHs, NO_x, polychlorinated aromatics, hexachlorobenzene, hydrogen cyanide, alcohols, and ketones [6, 38, 40-47]. Some of these products contribute to the formation of fine solid and liquid particulate matter, which together with all emitted gaseous compounds constitute smoke [6, 38, 40-47].	

OXIDATION	
Definition	**Reference**
"Reaction with oxygen either in the form of the element or in the form of one of its compounds."	National Fire Protection Association. (2019) [18]
"A chemical reaction in which the removal of electrons from a chemical species takes place. The reducing agent (or reductant) is the electron donor; the oxidizing agent (or oxidant) is the electron acceptor. The electron transfer may be accompanied by other events, such as atom or ion transfer, but the net effect is the electron transfer and hence a change in oxidation number of an element. Oxidation is usually an exothermic reaction."	Cozzani (2014) [103]
"any process in which oxygen combines with an element or substance, either slowly, as in the rusting of iron, or rapidly, as in the burning of wood"	Collins dictionary (2020) [51]

PYROLYSIS	

Definition	Reference
"The destructive distillation of organic compounds in an oxygen-free environment that converts the organic matter into gases, liquids, and char." or "A process in which material is decomposed, or broken down, into simpler molecular compounds by the effects of heat alone; pyrolysis often precedes combustion."	National Fire Protection Association. (2019) [18]
"The thermo-chemical decomposition of solid, carbonaceous substances into a range of products, either in the total absence of oxidizing agents or with a limited supply that does not permit gasification to an appreciable extent."	Cozzani (2014) [103]
"chemical decomposition of a substance by heat."	Collins dictionary (2020) [51]
General information	

Pyrolysis is an irreversible thermal decomposition of complex solid or fluid chemical substances at elevated temperatures in an inert or oxygen-free atmosphere. The rate of pyrolysis is temperature-dependent, and it increases with increasing temperature [19]. During pyrolysis, molecules are subjected to elevated temperatures, leading to molecular vibrations at which the molecules are stretched and shaken to such an extent that they start breaking down into smaller molecules [19]. Pyrolysis often precedes other processes, such as gasification and combustion, where partial or total oxidation of the treated material occurs. Pyrolysis processes can be both endothermic and exothermic [10, 16, 17].

SMOLDERING	
Definition	Reference
"Smouldering is a slow, low-temperature, flameless form of combustion, sustained by the heat evolved when oxygen directly attacks the surface of a condensed-phase fuel"	Rein (2009) [45]
"The slow, low-temperature, flameless form of combustion, sustained by the heat evolved when oxygen directly attacks the surface of a condensed-phase fuel or a char formed in a solid fuel devolatilization process."	Cozzani (2014) [103]
"burning slowly without flame, usually emitting smoke"	Collins dictionary (2020) [51]

SMOKE	
Definition	Reference

"the gaseous products of burning organic materials in which small solid and liquid particles are also dispersed."	Gross (1967) [52]
"the smoke aerosol or condensed phase component of the products of combustion."	Mulholland (2002) [54]
"The airborne solid and liquid particulates and gases evolved when a material undergoes pyrolysis or combustion, together with the quantity of air that is entrained or otherwise mixed into the mass."	National Fire Protection Association. (2019) [18]
"A visible suspension of carbon or other particles in air, typically one emitted from a burning substance."	Oxford dictionaries (2018) – Smoke [55]
"The product of combustion, consisting of fine particles of carbon carried by hot gases and air."	Collins dictionary (2020) [51]
"the gaseous products of burning materials especially of organic origin made visible by the presence of small particles of carbon."	Merriam-Webster dictionary (2019) [53]
General information	

Numerous technical, scientific, and dictionary definitions of smoke exist, and as seen above, all definitions involve combustion or burning. In most definitions, smoke is defined as a product of combustion. However, the U.S. NFPA mentions that smoke can also be emitted when a material undergoes pyrolysis or combustion [18]. As smoke is generally defined as solid and liquid particles suspended in a gaseous phase, smoke is an aerosol. Smoke is formed from combustion and associated high-temperature pyrolysis products generated from the burning of tobacco. Examples of smoke include aerosols generated from the burning of fossil fuels, biomass combustion, and the burning of tobacco in a lit cigarette.

THERMAL DECOMPOSITION	
Definition	Reference
"The production of gaseous substances (volatiles) from solids due to pyrolysis. The chemical decomposition of molecules at elevated temperatures."	Cozzani (2014) [103]
"Thermal decomposition is the process in which a chemical species breaks down when its temperature is increased."	Collins dictionary (2020) [51]

TORREFACTION	
Definition	Reference
"Torrefaction of biomass can be described as a mild form of pyrolysis at temperatures typically ranging between 200 and 300°C in an inert and reduced environment"	Shankar Tumuluru (2011) [10]

"Torrefaction is a thermal treatment at a temperature of 200 to 300 °C, at near atmospheric pressure and in the absence of oxygen."	Bergman (2005) [24]
"Torrefaction, also known as mild pyrolysis, is an example of a slow pyrolysis process. Torrefaction of biomass is a mild form of pyrolysis carried out under atmospheric conditions and at temperatures typically ranging between 200 and 320°C, where the onset of primary pyrolysis occurs at 200°C"	Al-Haj Ibrahim (2020) [19]

VAPOR	
Definition	Reference
"The gas phase of a substance, particularly of those that are normally liquids or solids at ordinary temperatures"	National Fire Protection Association. (2019) [18]
"A gas at a temperature below the critical temperature, so that it can be liquefied by compression, without lowering the temperature."	McGraw-Hill (2003) [115]

VAPORIZATION	
Definition	Reference
See volatilization.	

VOLATILIZATION	
Definition	Reference
"The conversion of a chemical substance from a liquid or solid state to a gaseous or vapor state by the application of heat, by reducing pressure, or by a combination of these processes. Also known as vaporization."	McGraw-Hill (2003) [115]
"to change or cause to change from a solid or liquid to a vapour"	Collins dictionary (2020) [51]