# The Role of Glycerol during Aerosol Formation in an Electrically Heated Tobacco Product

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## Introduction and Objective

Philip Morris International (PMI) is developing a range of novel tobacco products with the potential to present less risk of individual and population harm in comparison to continued smoking of cigarettes. One of these products is the Electrically Heated Tobacco System (EHTS) (also referred to as the Tobacco Heating System 2.2 (THS2.2)) (Figure 1a), which is currently commercialized in over 25 markets.

Using a patented *HeatControl* technology, the EHTS heats a specially designed, multicomponent tobacco stick (Electrically Heated Tobacco Product (EHTP)), that is inserted into the Holder (Heating Device) that operates at temperatures lower than that necessary for the tobacco to combust, to generate a nicotine containing aerosol.

The EHTP contains a specially manufactured porous tobacco plug that undergoes a controlled heating process to release chemical compounds (present in the tobacco material) into vapors that cool down to form a respirable aerosol (Figure 1b). As there is no combustion of the tobacco substrate (Cozzani et al. 2016), no smoke is formed. To generate a nicotine containing respirable aerosol under the controlled heating conditions of the EHTS, glycerol (due to its vapor pressure characteristics) was added to the tobacco substrate to act as an aerosol former.

The aim of this work was to investigate the role of glycerol during aerosol formation, under realistic product operating conditions of the EHTS, as well as for a relevant gas-vapor mixture composition measured in the generated EHTP aerosol.

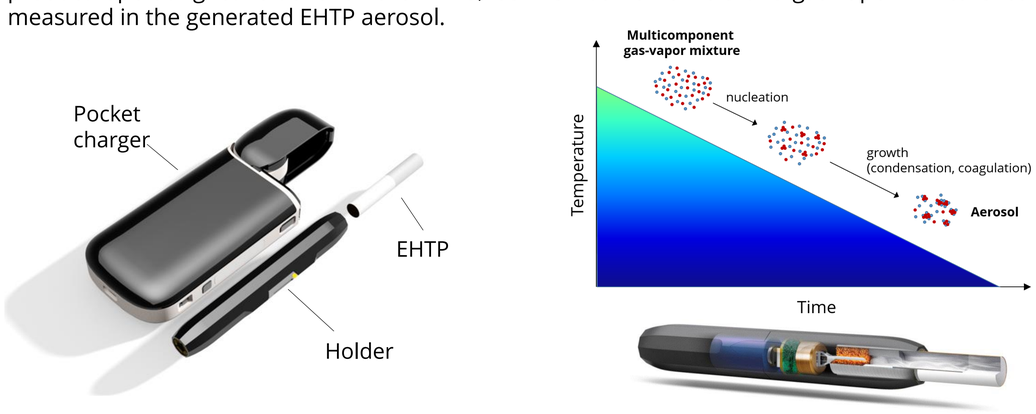


*Figure 1: a) The three components of the Electrically Heated Tobacco System, and b) a slice-through view of the EHTP inserted into the Holder during use and the processes behind aerosol formation from supersaturated, multicomponent gas-vapor mixtures.*

## Modeling aerosol formation

- An extended Classical Nucleation Theory (CNT) for multicomponent gas-vapor mixtures (Winkelmann, et al., 2017) was used to numerically simulate the aerosol formation processes (Figure 1b) taking place in the EHTP during use when operated in the Holder for the cases with and without glycerol present in the tobacco substrate.
- In the extended CNT, a multicomponent aerosol consisting of $N$ compounds is described by gas-vapor ($Y_i$) and liquid ($Z_i$) mass fractions and the droplet number density ($M$). $M$ is modeled by a two-moment approach (McGraw, 1997) with a log-normal distribution having fixed width ($\sigma$ = 1.33). The fluid (gas and/or droplet) mixture density ($\rho$) is assumed to follow the equation-of-state of an ideal gas mixture with mixture compressibility ratio ($\Psi$), depending on the temperature ($T$), pressure ($p$), and mixture composition according to Frederix (2016). $S_\rho$ is the net rate of change of density. Considering only the temporal evolution (with $t$ being the time) in a spatially homogeneous system, the governing equations can be written as:

Gas-vapor phase mass fraction equations: $$\partial_t(\rho Y_i) + S_\rho Y_i = -S_i^{nuc} + S_i^{e-c} \quad ; \quad i = 1,\ldots,N$$

Liquid phase mass fraction equations: $$\partial_t(\rho Z_i) + S_\rho Z_i = S_i^{nuc} - S_i^{e-c}; \quad i = 1,\ldots,N$$

Droplet number density equation: $$\partial_t(\rho M) + S_\rho M = J_{nuc} - J_{e-c} - J_{coa}$$

Mass fraction constraint: $$\sum_{i=1}^{N}(Y_i + Z_i) = 1$$

Equation-of-state: $$\rho = \psi(p,T)\,p$$

- The nucleation rates ($S_i^{nuc}$, $J_{nuc}$), the evaporation-condensation rates ($S_i^{e\text{-}c}$, $J_{e\text{-}c}$), and the coagulation rate ($J_{coa}$) were modeled in the same way as described by Winkelmann, et al. (2017).
- The governing equations and the models for nucleation, evaporation, condensation, and coagulation were implemented into a segregated, finite-volume based Computational Fluid Dynamics (CFD) code using the Open source Field Operations and Manipulations (OpenFOAM®) software package as described by Frederix (2016).

## Simulation set-up

- The time evolutions of gas-vapor and liquid phase compounds and the droplet number density were computed in a single computational cell for a constant cooling of two predefined multicomponent gas-vapor mixtures representing the aerosol composition analytically measured by Schaller et al., (2016). The compounds of the two mixtures were:
  - **Mixture A:** *air, water, glycerol, nicotine, carbon monoxide, nitric oxide, nitrogen dioxide, 1,3-butadiene, isoprene, acrylonitrile, benzene, toluene, pyridine, quinolone, styrene, resorcinol, catechol, phenol, p-cresol, m-cresol, o-cresol, formaldehyde, acetaldehyde, acetone, acrolein, propionaldehyde, crotonaldehyde, methyl ethyl ketone, butyraldehyde*
  - **Mixture B:** all the compounds mentioned above for mixture A, except for *glycerol*.
- The gas-vapor mixtures were cooled down from 623.15 K (350 °C) to 303.15 K (30 °C) with a constant rate of $-3{\cdot}10^5$ K/s (representative of the conditions in the EHTP (Nordlund and Kuczaj, 2016)) under isobaric conditions (pressure p = 101325 Pa).
- The temperature dependent thermophysical properties of the compounds were extracted from external databases and used in the same way as described by Nordlund and Kuczaj (2016).
- The time derivative was discretized by a second-order implicit backward differencing scheme and a constant time step of $10^{-8}$ s was used.

## Results and discussion

**Mixture A:**

- As the multicomponent gas-vapor **mixture A** was cooled down, aerosol droplets were formed (sharp increase in $M$) after ~0.8 ms when the temperature of the mixture reached ~106 °C (Figure 2a). As shown in Figure 2b, glycerol was the compound triggering aerosol formation under the simulated operating conditions of the EHTS and EHTP. Water and nicotine thereafter condensed onto the initial glycerol droplets, resulting in an increase of the diameter of average mass ($d_m$) of the droplets (Figure 2a). After the aerosol droplet formation (triggered by glycerol), $M$ was shown to decrease with time due to coagulation towards $1.1{\cdot}10^{16}$ kg$^{-1}$. The coagulation together with continued condensation onto the aerosol droplets resulted in a $d_m$ of 0.18 µm after 100 ms.

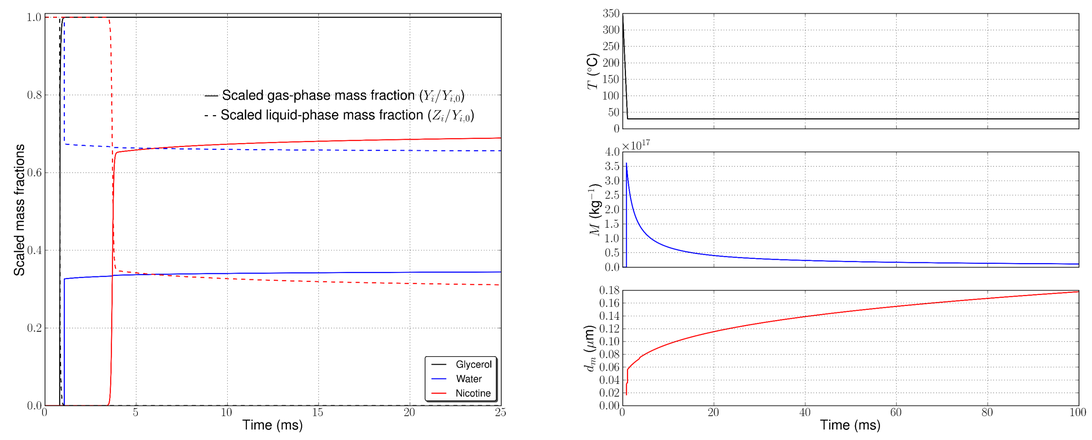


*Figure 2: a) Evolutions of the temperature (top), droplet number concentration (middle), and droplet diameter of average mass (bottom), and b) the gas-vapor and liquid phases (scaled by their initial values ($Y_{i,0}$={5.83·10$^{-3}$, 4.04·10$^{-2}$, 1.74·10$^{-3}$} for glycerol, water, and nicotine, respectively) during the cooling of the multicomponent gas-vapor **mixture A**.*

**Mixture B:**

- As the aerosol former glycerol was absent in **mixture B**, no aerosol droplets ($M$ = 0) were formed when the multicomponent gas-vapor mixture was cooled down, as shown in Figure 3a. Water and other compounds in the gas-vapor mixture were not able to reach supersaturation and therefore could not generate aerosol droplets from the multicomponent gas-vapor mixture at the simulated operating conditions of the EHTS and EHTP. This implies that without the presence of glycerol in the aerosol mixture, the compounds of the gas-vapor mixture remained in their gas-vapor phase even after having been cooled down, i.e. no phase transition occurred, as was also observed by Nordlund and Kuczaj (2016).
- This implies that according to the extended CNT, glycerol (which is evaporated from the EHTP tobacco substrate) is the compound triggering the aerosol formation from the multicomponent gas-vapor mixture under the tested operating conditions of the EHTS and EHTP.

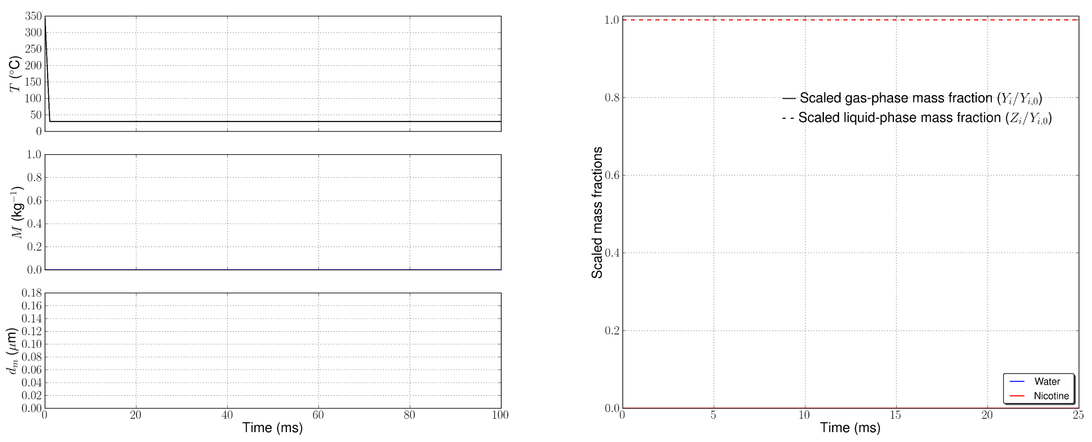


*Figure 3: a) Evolutions of the temperature (top), droplet number concentration (middle), and droplet diameter of average mass (bottom), and b) the gas-vapor and liquid phases (scaled by their initial values ($Y_{i,0}$={4.04·10$^{-2}$, 1.74·10$^{-3}$} for water and nicotine, respectively) during the cooling of the multicomponent gas-vapor **mixture B**.*

## Conclusions

- According to the extended CNT, an aerosol former, such as glycerol, is required to be present in the gas-vapor mixture for an aerosol to form under the tested operating conditions of the EHTS and EHTP.
- Aerosol droplets were generated from **mixture A** (representing the EHTP aerosol composition) when the glycerol (being evaporated from the EHTP tobacco substrate when heated) was cooled down. Water and nicotine, were found to condensate onto the already generated droplets, contributing to the increase of the droplet size in addition to the increase by coagulation.
- For **mixture B**, in which glycerol was absent, water and other compounds in the gas-vapor mixture were not able to reach supersaturation and therefore could not generate aerosol droplets from the multicomponent gas-vapor mixture at the tested operating conditions of the EHTS and EHTP.

## References

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**July 3-6, 2017**

**Competing Financial Interest**

The author is employed by Philip Morris Products S.A. (part of Philip Morris International group of companies), which is the sole source of funding and sponsor of the research described in this poster.