The combined chemical and mechanical modifications of cigarette: a novel methodology to reduce harmful effects

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Received: 20 January 2021 / Accepted: 27 May 2021 / Published online: 10 July 2021
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Abstract
Without hindering the taste, making a cigarette less harmful by reducing the percentage of toxic and carcinogenic compounds in the smoke of the cigarette is a challenging task for the current generation of researchers. In the current work, by implementing mechanical, chemical and combined modification techniques, the above stated is tried to mitigate. In addition to the above, the optimum suction pressure, burning time and the number of puffing are also determined. Mechanical modification technique considers filter to cigarette ratio and filter design as the controlling parameters. The mathematical calculation reveals that puffing should stop when the cigarette length reaches 0.15 times of its original length. Furthermore, it is also identified that the concentrations of suspended solids and droplets in the smoke decrease significantly (separation efficiency = 56.81%) if the cigarette to filter ratio is maintained at 2.32. In case of chemical modification, by using various types of adsorbents such as charcoal and Zeolite 13X, the harmful effects are further reduced. These processes depict significant reduction in harmful effect (separation efficiency up to 62.1%) by showing the decrement in the suspended solids and droplets in the smoke due to the adsorption on the active sites of adsorbents. In case of combined modification, the achieved separation efficiency is 66.51%. For the experimentation, an experimental setup fitted with artificial lungs was used.

Keywords Cigarette · Carcinogenic effect · Suspended solids · Filter

Introduction
Cigarette smoking is one of the most plausible reasons for the increment in the number of deaths worldwide. Cigarette smoke is a mixture which contains approximately 5000 chemicals. As per the information reported by the World Health Organization (WHO), 5.4 million premature deaths occur worldwide due to smoking. Talhout et al. 2011 predicted that by 2025, around 10 million smokers could die annually. In addition to the above, cigarette smoke also causes air pollution. Among all the components of cigarette smoke, air pollution is mainly caused by polyaromatic hydrocarbons (PAHs). The neural network (RNN) model developed by Zhao et al. 2019 theoretically predicts the quantity of polyaromatic hydrocarbons present in the air (Fu et al. 2020).

Cigarette smoke is divided into mainstream (MS) and side stream smokes (SS). MS consists of liquid droplets and fine solids suspended in the gas-vapour phase. These are produced due to the incomplete combustion, sublimation, pyrolysis, and pyro synthesis of gold leaf flakes and distillation, and condensation of smoke (Piadé et al. 2013). In MS, carcinogenic compounds such as acetaldehyde, benzene, 1,3-butadiene, formaldehyde, ethylene oxide, and cadmium are detected (Borgerding and Klus 2005; Burns et al. 2008; Hecht et al. 2010; Sureda et al. 2013).

SS smoke contains high amount of CO and tar and these contaminates significantly indoor air (Goniewicz et al. 2009; Al-Sayed and Ibrahim 2014). The above-stated compounds lead to respiratory illness, otitis media, asthma, sudden infant death syndrome, vascular dysfunction, chronic obstructive pulmonary disease (COPD) and lung cancer (Schick and Glantz 2005; Flouris et al. 2010; Jefferis et al. 2010; Goldklang et al. 2013; Valenti et al. 2013). It is also reported that fresh SS is approximately four times more toxic than MS.
and its percentage in the total smoke depends on the combustion efficiency and methodology (Russell et al. 1980; Schick and Glantz 2006; Benowitz et al. 2010; Schick et al. 2012, and Yu et al. 2019).

The above stated harmful chemicals present in the smoke either in the form of droplets or in the form of suspended solids which settle at various locations of the lungs, mouth and throat due to the sudden changing in the direction during the flow of smoke. The settlement mechanism is described with the help of a schematic diagram (Fig. 1). The rapid expansion of smoke in the mouth and lungs and sudden changes in flow direction at the throat and in the lungs enhance the pressure drop. Due to this, the suspended solids settle. Furthermore, the droplets agglomerate due to the temperature variation.

To reduce the harmful effect, the suspended solids, droplets settling rate and condensation rate of smoke must be minimum. The phenomena described above depend on the residence time, quality of tobacco, the efficiency of the filter and the methodology of smoking. The residence time depends on the flow pressure, and it is proportional to velocity. For the attainment of minimum settling, the pressure drop must be low.

To attain enhanced separation efficiency, chemical, mechanical or combined modification methodologies are used. In the absence of any information in the open literature regarding the above stated modification processes, in the current work, an attempt has been made to develop an efficient modification process to achieve the maximum separation efficiency.

In the mechanical modification, the filter’s design, packing density and cigarette to filter ratio are altered. The improper selection of any parameter discussed above augments the carcinogenic effect by decreasing the separation efficiency. Therefore, proper selection of the parameters mentioned above is required to minimize the carcinogenic effect without hampering the taste and feeling of the smoker. In this regard, the literature does not disclose any information in open literature.

The chemical modification includes the use of various adsorbents as a filter to enhance the separation efficiency. It is also be noticed from the previous work that various additives are used in the tobacco mixture to reduce the carcinogenic effect of smoking; However, the harmful effect of burning of additives and catalyst are not considered in the modifications (Bombick et al. 1997; Muscat et al. 2005; Miyamoto et al. 2012; Firas et al. 2014; Martins et al. 2015; Campo et al. 2016; Moura et al. 2016; Abdul Kareem et al. 2018). In the case of commercially available electronic cigarette, the disadvantages reported in the literature had been tried to eliminate. However, the survey conducted by UNICEF and WHO reveals that the use of electronic cigarette does not mitigate the requirement of the consumers (Badea et al. 2018).

For the chemical modification, charcoal, 13X-Zeolite and the mixture of both were used to separate the significant number of suspended solids and droplets (Zhang et al. 2010; Mopoung et al. 2015). However, these study were not carried out at the optimum conditions. In the current manuscript, at the optimum conditions, the removal efficiencies of the various carcinogenic elements by the adsorbents and filters have been reported. In addition to the above, the effect of both chemical and mechanical modifications on removal efficiency has also been tried to disclose.

Experimental setup and procedure

Procedure

In the current work, an indigenously designed and fabricated experimental setup and lungs were used. The schematic diagram of the experimental setup is depicted in Fig. 2. The experimental setup contains a suction chamber, filtration unit, data recording subsection, artificial lungs and an imaging unit with an illumination system. The suction chamber mimics the role of the mouth.

Experiments were performed at room temperature (−25°C) and atmospheric conditions (1 atm). For all the experiments, King’s Gold Flake brand cigarettes were used. The cigarette before ignition was fitted with the suction chamber (63 cm³) which is connected with a suction pump (capacity = 30 kPa). Thermistor 1 is attached to the mouth cavity and it measures the temperature of the inlet smoke to the mouth. At the suction chamber’s entry point, the smoke samples were collected as the inlet smoke to the cavity for the further analysis. To identify the quality of outlet smoke, smoke from the lungs was collected. During the experimentation, the pressure and the flow rate of the inflow were measured by using the pressure gauge and rotameter, respectively. The measuring instruments associated with the experimental setup have the following specifications:

1. Rotameter: 10–1000 ml/min, 2. Pressure Gauge: 0.1–0.2 bar and 3. Thermistors: 25–150 °C.

Particulate collection methodology

The schematic diagram of the fabricated separator is presented in Fig. 3. After collection of the smoke, it is sent to the separator for the determination of solid and droplet concentrations in the smoke. The separator has two sections: (1) Retentate and (2) Permeate. Both sections have equal volume that is 40 ml. The length and diameter of each section are 2.38 cm and 46.2 mm, respectively. Both the sections are separated by a
1. Sudden increment in volume leads to the decrement in pressure of smoke and as a result the kinetic energies of the suspended droplets and solids decrease. This phenomenon promotes settling of suspended solids and droplets.

2. Around the neck, sudden bending in the flow path promotes the formation of vortices and as a consequence further enhancement in pressure drop is expected. This favours settling.

3. Sudden bending once again in the flow path leads to the settling of suspended solids.

4. Augmentation in sudden volume rises the pressure drop further and settling of suspended solids and droplets as well also.

Fig. 1 Schematic depicting problem associated with cigarette smoke flow.
Fig. 2 Proposed schematic diagram of the experimental setup

Fig. 3 Schematic of the setup used to separate the suspended solids and droplets
filter paper (2 μm PTFE 46.2 mm filter) and the sections are joined by using a sealant. The stereo image and the photograph of the used filter papers are presented in Fig. 4a and b, respectively.

Materials

In the current work, the following raw materials were used (Table 10). For the experimentation, standard cigarettes available in Indian open markets were used. In addition to the above, various adsorbents such as Charcoal and Zeolite-13X were also used and the details of the manufacturer have also been provided in Table 10.

Characterization of raw materials

In the raw material characterization, moisture analysis was performed to identify the expected amount of condensate presents in the smoke. In addition to the above analysis, SEM, XRD, FESEM and EDS analysis were also performed to determine the carcinogenic compounds present in the cigarette. Furthermore, analysis of the filter was also performed. Finally, the thermal and flow behaviour were modeled to map the various process controlling the harmful effects.

Moisture analysis

For the moisture analysis, one gram of raw tobacco was taken in a known weight crucible. Then, the sample was kept in an oven for 1.5 h at 110 °C. Finally, the sample was brought out from the oven and kept in a desiccator. After the sample cooled down to room temperature in the desiccator, the weight of the sample was measured. Finally, the percentage of moisture is determined by following Eq. 1.

\[
M = \frac{W_w - W_d}{W_w} \times 100 = 9.72\%
\]  

Packing density

For the calculation of the above stated parameter, cylindrical shape is assumed for the cigarette. Without filter, the diameter
and length of the cigarette are 0.7 cm and 5.45 cm, respectively and the calculated weight is 0.7012 gm. Based on the above information, the packing densities of cigarette and filter are calculated using Eq. 2 and the obtained values are 333.9 kg/m$^3$ and 180 kg/m$^3$, respectively.

$$\text{Packing density} = \frac{W_w}{V}$$  \hspace{1cm} (2)

**Characterization of the filter**

In the current work, the commercially available filters were used. The fibres arrangement and its porosity are determined using the images captured by a Leica microscope. This characterization helps to define the flow pattern of the smoke. Furthermore, in Table 1, the physical properties and the compositions of the filter have also been presented. The optical and FESEM images of the cigarette filter after different modifications are depicted in Fig. 5.

### Results

**Composition analysis**

**In terms of compound**

For the raw cigarette tobacco, charred part and ash collected after burning, XRD analysis was performed and obtained results are presented in Fig. 6(a–c). From the XRD analysis of the cigarette ash and raw material, it is identified that both ash and raw material contain carcinogenic compounds. However, for the quantification of the above stated, elemental analyses were performed.

**In terms of elements**

As the process deals with combustion, the proximate and ultimate analyses are initially performed (Table 2). From the analysis, it is identified that the raw tobacco contains very high amount of moisture and therefore, the smoke contains fine droplets in the form of aerosol. During the flow of smoke

| Properties of filter                  |         |
|--------------------------------------|---------|
| Length of the filter                 | 1.6 cm  |
| Pore diameter                        | 50µm    |
| Packing density                      | 180 kg/m$^3$ |
| Cellulose Acetate fibre (C$_{10}$H$_{16}$O$_8$) | Chemical composition: C= 49%, O=45% and H= 6% |

| Images (Top view of the exposed part) | Cross-sectional view by Leica Microscope |
|--------------------------------------|-----------------------------------------|
| 1.6 cm                               |                                         |
| 0.7 cm                               |                                         |

Schematic of filter
inside our body, these are expected to agglomerate and deposit. Furthermore, the ultimate analysis indicates that flakes of gold leaf contain very small amount of sulphur. For the elemental analysis in detail, EDS analysis was performed for each sample and the obtained results are presented in Fig. 7(a–c).

For the detection of traced components in the composition, chemical analysis was performed. In this case, the dried and pulverized samples were chemically digested in the mineral acids such as HCl and HNO₃. The digested sample was then filtered using filter paper (Whatman filter paper 42) to separate any insoluble residues. Furthermore, to separate all the heavy metal ions from the residue, the filter paper was at least washed 3 times. Finally, the collected mother liquor was analysed using ICP-OES (Perkin Elmer 8300). The primary wavelength for each element was selected during the analysis work. The instrument’s standards were purchased from Merk (India) Ltd and used after suitable dilution. The obtained compositions are reported in Table 3.

From the above stated information, the smoke and ash compositions are calculated by using Eqs. (3)–(6).

\[\text{Raw tobacco} \rightarrow \text{Δ Ash} + \text{smoke} \rightarrow \Delta H \]

\[C_s = C_r - C_a \quad (4)\]

\[C_r = W_{dr} X_{er} \quad (5)\]

\[C_a = W_{ea} X_{ea} \quad (6)\]

The compositions of raw tobacco, smoke and ash indicate that the significant percentage of the carbon present in the raw material goes to the gaseous phase after burning (Table 4). The C in the gaseous stream as CO and CO₂ are very much harmful to human health. In addition to the above, elements

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**Fig. 5** Cigarette filter optical images: (a) Without catalyst. (b) With Activated Carbon. (c) With Zeolite-13X. (d) With Activated Carbon+Zeolite-13X; FESEM images with 500-μm scale: (A) Without catalyst. (B) With Activated Carbon. (C) With Zeolite-13X. (D) With Activated Carbon+Zeolite-13X

**Fig. 6** XRD plot for (a) cigarette tobacco, (b) charred part and (c) cigarette ash
such as Pb, Cr, Hg and Cd also transform to smoke. Therefore, along with the carcinogenic compounds, the above stated compound or elements intensities also need to reduce. The transformation rate of elements enlisted in Table 4 depends on the surface texture.

### Surface morphology and texture

SEM analysis of raw tobacco, charred part and ash is presented in Fig. 8(a), (b) and (c), respectively. These depict that after combustion, the coarse solids present in the raw material convert to comparatively finer solids. The raw tobacco pores are not visible as these are filled by volatile matter and moisture. With the progress of burning, the moisture and volatile matters become smoke and as a result, the pores are visible (Fig. 8(b)). In case of complete burning of tobacco (Fig. 8(c)), the agglomerated form of the ash is noticed. Furthermore, it also observed that tobacco after burning converts to finer particles. In addition to the cigarette part, the filter’s properties also play a significant role and decide the separation efficiency.

### Role of suction velocity and pressure drop

The following are the assumptions for the establishment of a relationship between pressure drop and velocity:

- Uniform smoke and air distribution
- The composition of tobacco is uniform
- Cigarette and filter are considered as packed bed

### Table 2 Proximate and ultimate analysis

| Proximate analysis | Ultimate analysis |
|--------------------|-------------------|
| FC 11.00%          | C 50.60%          |
| VM 63.28%          | H 5.60%           |
| M 14.20%           | O 43.09%          |
| Ash 11.52%         | N 0.21%           |
| Calorific value 9.3 MJ/kg | S 0.02% |

### Table 3 Trace element analysis

| Component | Quantity (ng/l) | Percentage (%) × 10⁻⁴ |
|-----------|-----------------|-----------------------|
| Cd        | 37.9            | 1.5                   |
| Pb        | 1753            | 10                    |
| Ni        | 10              | 50                    |
| Cr        | 753             | 1                     |

![Fig. 7 Elemental analysis of (a) raw tobacco, (b) charcoal and (c) ash parts](image)
Flakes of tobacco are assumed to be cylindrical. The density of packed bed is constant. The diameter and length of the packed bed are 0.7 and 7 cm, respectively.

Before modelling, few terms which are used in the modelling need to describe and these are sphericity and void fraction. For the calculation of void volume, the required data are the diameter of a tobacco particle (0.1 cm), length of a tobacco particle (0.31 cm) and the calculated volume ($V_p$) of one cigarette particle ($9.82 \times 10^{-3}$ cm$^3$). The surface area ($A_{ps}$) of one tobacco particle is 0.41 cm$^2$ and the equivalent radius ($r_o$) is 0.133 cm. With the help of the above information, sphericity of a tobacco particle is determined using Eq. 7.

\[
\text{Sphericity } (\phi) = \frac{A_{es}}{A_{ps}} = 0.54
\]  

(7)

Without filter, the volume of a cigarette is 2.097 cm$^3$ and the volume of water displaced is 1 ml when a cigarette without filter is dipped in water. With the help of the above information, the void fraction is calculated by using the following formula:

\[
\text{Void fraction } (\varepsilon_c) = \frac{\text{Volume of water displaced}}{\text{Volume of a cigarette without filter}} = \frac{1}{2.097} = 0.48
\]  

(8)

From the filtration unit, the volume is found to be 0.596 cm$^3$ and the volume of water displaced is 0.2 ml. Hence, the void fraction is:

\[
\text{Void fraction } (\varepsilon_{fu}) = \frac{0.2}{2.5957} = 0.34
\]  

(9)

Finally, with the help of information represented in Eqs. (8) and (9) and velocities reported in Table 5, the pressure drop is calculated using Eq. 10 (30).

\[
\Delta P = \frac{150 \mu_f U_o (1-\varepsilon)^2}{L \varepsilon^3 \phi_s^2 d_p^2}
\]  

(10)

The enhancement in velocity is observed with the rise in pressure drop (Table 5) due to the alteration of flow pattern from laminar to transition. In addition to the above, the
augmentation in residence time is also noticed and this promotes step-up in combustion efficiency, which is a desirable criterion to achieve minimum harmful effect. The following modelling work needs to be performed to identify the role of pressure drop in inspiration and expiration during smoking. Equation 10 is modified to Eq. (11) for simplicity,

\[ \Delta P = KL \] (11)

\[ P_{(in \, mouth)} = 101.325 \, kPa = KL \] (12)

\[ P_{(in \, mouth)} = (KL + 101.325) \, kPa \] (13)

According to Boyle’s law,

\[ (KL + 101.325) V_{suction} = 101.325 \times 63 \, kPa \] (14)

\[ V_{suction} = \frac{6403}{KL + 101.325} \, ml \] (15)

\[ V_{inspiration} = V_{suction} + 500 \, ml \] (16)

During expiration, 500 ml is discharged. The inspiration and expiration volumes during cigarette smoking are presented through a P-V diagram (Fig. 9) using Eqs. (11)–(15) (Kim et al. 2005; Brown 2015). \( V_{suction} \) amount retains in the lungs after expiration and releases during subsequent breathing. It is the reason for the getting cigarette smoke smell even after smoking completes. Due to the additional residence time in the lungs, the settling of droplets and suspended solid augments. Higher suction pressure indirectly increases the residence time and settling velocity. The above stated is consistent with Eq. (17) according to the information reported in literature (Kim et al. 2005).

\[ V_{settling \, in} = 5 \times 10^4 V_{settling \, outside} \] (17)

In addition to the P-V relationship describing the harmful effect of cigarette, combustion, condensation and distillation effects also need to be determined (MacIntyre and Branson 2001). For this, the thermal modelling is performed:

\[
\text{Heat released during combustion reaction} = \text{Heat absorbed by the gold leaf flakes} + \text{Heat loss due to radiation} + \text{Heat absorbed by filter} \tag{18}
\]

The heat generated by tobacco during combustion is 9.75 MJ/kg, and therefore the total heat generated by one cigarette is 9.75 MJ/kg × 0.7012 × 10⁻³ kJ heat.

\[
\text{Heat absorbed by filter unit} = m \times C_P \times \Delta T = 0.0358 \times 1.27 \times (T-25) \tag{19}
\]

\[
\text{Heat loss due to radiation from cigarette,} = \sigma \times A_c \times (T_1^4-T_2^4) = 2.182 \times 10^{-12} \times (973^4-298^4) \tag{21}
\]

Heat loss due to convection from cigarette smoke to flakes is:

\[
\frac{n \times h \times A \times \Delta T}{L} = n \times L \times 0.0332 \times R_e^{0.5} \times F_r^{0.334} \times \Delta T \times t \tag{24}
\]

Using Fig. 10 and the above stated equations (Eqs. (18)–(24)), at any time ‘t’, it can be written that
\[ \rho A \left( L - L' \right) C_p = 9.37 \times 10^6 \times L' \times \rho \times A \times -1.938t - n \]
\[ \times L \times 0.0332 \times R_e^{0.5} \times P_r^{0.34} \times \Delta T \]
\[ \times t \]  
(25)

\[
\left( L - L' \right) \times 2250 \times 750 = 9.37 \times 10^6 \times L' - 1.938t - n
\]
\[ \times L \times 0.0332 \times R_e^{0.5} \times P_r^{0.34} \times \Delta T \times t \]  
(26)

\[
\left( L - L' \right) 1687500 = 9370000L' - 1.938t - n \times L \times 0.0332
\]
\[ \times R_e^{0.5} \times P_r^{0.34} \times \Delta T \times t \]
(27)

\[
\frac{L'}{L} = 0.15
\]
(30)

\[
L' = 0.15 \times 5.7
\]
(31)

\[
L' = 0.855 \text{ cm}
\]
(32)

\[ \Delta T = 850 \degree C - T(x, t) \text{ and } T(x, t) \text{ is determined by solving the following partial differential equations:}
\]
\[ \frac{\partial^2 T(x, t)}{\partial x^2} = \frac{\partial T(x, t)}{\partial t} \]  
(33)

\[
\alpha \frac{T_{n+1}^{i+1} - 2T^n_i + T^{n+1}_{i-1}}{\Delta x^2} = \frac{T_{n+1}^{i-1} - T^n_i}{\Delta t}
\]
(34)

\[ aT^n_{i+1} + bT^n_i + cT^{n+1}_{i+1} = fT^n_i \]  
(35)

Where \( a = -\alpha \frac{\Delta t}{\Delta x^2} = c, b = 1 + 2\alpha \frac{\Delta t}{\Delta x^2} = f = 1 \)  
(36)

\[
\begin{bmatrix}
  b & a & c & \ldots \\
  a & b & c & \ldots \\
  \ldots & \ldots & \ldots & \ldots \\
  \ldots & \ldots & \ldots & \ldots \\
  \ldots & \ldots & \ldots & a & b & c \\
  \ldots & \ldots & \ldots & a & c & b
\end{bmatrix}
\begin{bmatrix}
  T_1^{n+1} \\
  \ldots \\
  T_i^{n+1} \\
  \ldots \\
  T_N^{n+1}
\end{bmatrix}
= \begin{bmatrix}
  T_1^n \\
  \ldots \\
  T_i^n \\
  \ldots \\
  T_N^n
\end{bmatrix}
\]
(37)

For the calculation of temperature distribution along the cigarette’s length, the cigarette’s geometry is initially discretized. For this, Crank-Nicolson and Fully Implicit formula are utilized. The discretized forms and the original equations are mentioned above. In addition to the above, the solution is converted into a tridiagonal matrix. For the calculation, the following assumptions are used:

1. After the grid independency test, it is found that the values of \( \Delta x \) and \( \Delta t \) are 0.006 m and 0.1 s, respectively.
2. The temperature variation along the radial direction is assumed to be insignificant.
3. At \( t = \text{any time} \); \( x = L \) and \( T = 850 \degree C \).
4. At \( t = 180 \text{ sec} \); \( x = L = \text{filter} = 0 \text{ cm} \) and \( T = 120 \degree C \).
5. At \( t = 90 \text{ sec} \); \( x = L = 0.855 \text{ cm} \) and \( T = 850 \degree C \).
6. The thermo-physical properties of gold leaf are given as the input.
7. \( L = 5.7 = n\Delta x; t = 180 = n\Delta t \) and \( \frac{L}{T} = \frac{n\Delta x}{n\Delta T} \).
8. The tri-diagonal matrix is solved for the known heat flux, and from that, the temperature at different locations is determined by following the methodology presented in INTEMP software.

For the different time intervals, by using information reported above (Eqs. (18)–(22)), the absorption zone, distillation and condensation zone lengths are also determined (Table 6).

### Optimization of the process parameters

For this investigation, the cigarette was burnt at various suction pressures. The suction was provided at an interval of 0.1 s and it continues for 3 s. During the burning of cigarette, the

| Time (sec) | Ash zone+ burning zone (cm) | Absorption + distillation zone (cm) | Comment |
|-----------|-----------------------------|-----------------------------------|---------|
| 30        | 0.855                       | 4.845 cm                          | Condensate and distillate remain in the filter |
| 60        | 1.710                       | 3.99 cm available for the above mentioned in place of 4.858 cm | A minimal amount of distillate and condensate go to the human body |
| 90        | 2.565                       | 3.135 cm available for the above mentioned in place of 4.858 cm | Around 25% of the total particulate and droplet enters into the human body |
| 120       | 3.42                        | 2.28 cm available for the above mentioned in place of 4.858 cm | Around 60% of the total particulate and droplet enters into the human body |
| 150       | 4.275                       | 1.25 cm available for the above mentioned in place of 4.858 cm | Around 90% of the total particulate and droplet enters into the human body |
whole process was recorded using a high-speed motion analyser and thereafter, by using an image processing software, the transient nature of the images was determined. Finally, by using the following formula (Eq. (38)), the burning time is determined.

\[
\text{Burning time} = \frac{(F_f - I_f)}{n} \Delta t \tag{38}
\]

The variation of burning time with the suction pressure is presented in Fig. 11 and it shows a decreasing trend with the increasing suction pressure up to 10 kPa due to the rise in airflow which generally promotes the burning process. However, above 10 kPa, as the suction pressure increases, the burning time depicts accelerating trend due to the decrement in the residence time of fresh air in the cigarette. In this case, incomplete combustion is expected. In addition to the above, Table 7 depicts that the separation efficiency increases up to 10 kPa suction pressure. Further increment in suction pressure, the separation efficiency declines. The above-stated investigation discloses that the optimum burning time and suction pressure are 135 s and 10 kPa, respectively. From the information reported above, the calculated optimum number of strokes is 9. In the current work, stroke represents one cycle of puffing. To verify the above mentioned, experiments were conducted by altering the number of strokes at the optimum burning time. The obtained results are presented in Table 8. In this analysis, instead of considering separation efficiency, the unburnt carbon percentage has been considered as the controlling parameter.

### Mechanical modifications

In mechanical modifications, the ratio of filter to cigarette length (F/C) is varied. In the current work, the cigarette length of 5.4 cm is considered as the one unit and the filter of 1.55 cm length is taken as the one unit. For this study, at 10 kPa suction pressure and around 10 strokes, the cigarette was burnt. In per suction, the smoke enters the lungs, and the smoke from the lungs is collected using the methodology described earlier.

The analysis depicts that with the increasing F/C ratio from 0 to 1.5, the separation efficiency enhances significantly (Table 9). It could be due to the entrapment of significant amount of suspended solids in the filter’s pores. Although beyond 1.5, the above-stated parameters follow the same trend line; however, the cigarette’s taste deteriorates, and the consumer gets insufficient smoke due to very high resistance for

### Table 7  Role of suction pressure at various conditions

| Suction pressure (kPa) | Suspended solids at entry (μg/m³) | Suspended solids at exit (μg/m³) | Amount settled (μg/m³) | Separation efficiency (%) |
|------------------------|----------------------------------|----------------------------------|------------------------|--------------------------|
| 14                     | 1210                             | 1097                             | 113                    | 31.25                    |
| 16                     | 1060                             | 950                              | 110                    | 39.77                    |
| 20                     | 850                              | 810                              | 40                     | 51.70                    |
| 26                     | 1001                             | 860                              | 141                    | 43.12                    |
| 32                     | 1109                             | 960                              | 149                    | 36.98                    |

### Table 8  Role of number of strokes

| Number of strokes | Unburnt carbon in ash (%) |
|------------------|---------------------------|
| 4                | 45.23                     |
| 8                | 31.8                      |
| 10               | 30.82                     |
| 13               | 30.87                     |
flow created by the higher length filter. Therefore, up to 1.5 F/C ratio is acceptable.

From the XRD analysis (Fig. 12(a–d)), it is concluded that the peak intensity of certain carcinogenic compounds decreases as the filter length increases. Cellulose fibres of the filter block some part of the smoke particles from entering into the lungs (Fig. 13). For the further improvement in separation efficiency, chemical modification is performed.

Table 9  The role of mechanical modifications

| F/C | Inlet SS conc. (μg/m³) | Outlet SS conc. (μg/m³) | Condensed matter (μg/m³) | Amount settled (μg/m³) | Separation efficiency (%) | (ΔP) x L (kPa.m) |
|-----|-----------------------|------------------------|--------------------------|------------------------|--------------------------|------------------|
| 0   | 1760                  | 1632                   | 22.8                     | 128                    | 0                        | –                |
| 0.5 | 1310                  | 1197                   | 11.3                     | 113                    | 25.55                    | 20               |
| 1   | 860                   | 780                    | 7.1                      | 80                     | 51.13                    | 30               |
| 1.5 | 760                   | 700                    | 2.13                     | 60                     | 56.81                    | 30               |
| 3   | Insufficient suction pressure (ΔP) x L= minimum pressure required to maintain a flow |

**Chemical modifications**

**Zeolite-13X**

In case of chemical modification, within the filter, adsorbents such as charcoal and Zeolite-13X were used as a separate layer (Fig. 13a). The addition of adsorbent layer is expected to reduce the suspended solids and droplets from the smoke due to

Fig. 12  Intensity VS 2θ graph from XRD analysis by varying cigarette to filter ratio: (a) 1:0 (top-left); (b) 1:1 (top right); (c) 1:2 (bottom-left); (d) 1:3 (bottom-right)
adsorption on the active site. The schematic diagrams (Figs. 13a and 14) depicting the structure of cigarettes and adsorbents for the various cases are presented. In addition to the above, the adsorption mechanism is presented in Fig. 13 (b) and the adsorption capacity of adsorbents is presented in Table 11.

Zeolite is considered as the potential solid adsorbents for separation and purification, and it is cost-effective. It also depicts higher lifetime and has eco-friendly nature (Firas et al. 2014; Martins et al. 2015; Campo et al. 2016; Moura et al. 2016). In addition to the above, zeolite also illustrates higher specific surface area and smaller average pore diameter (Tables 10 and 11). Zeolite-13X has also a promising affinity towards CO$_2$ adsorption (Miyamoto et al. 2012; Abdul Kareem et al. 2018). Various amounts of Zeolite-13X ranging from 0.5 to 2 g were used to fabricate different cigarettes. Then, each cigarette’s performance is determined by measuring the suspended solids concentration in the entry and exit...
smoke. The analysis clearly illustrates that the optimum performance is obtained in the case of 0.1 g Zeolite-13X (Tables 12 and 13). Beyond the stated amount, the 10 kPa pressure is insufficient to create suction for the proper combustion.

**Charcoal**

In case of modification by charcoal, by following the methodologies described above, finite numbers of cigarettes were prepared (Fig. 15). The performance of each cigarette was determined by measuring suspended solids and the droplets concentrations at the inlet and outlet. Table 14 illustrates that the concentrations of suspended solids at the inlet and outlet decline with the increasing concentration of charcoal up to 0.75 gm and above the stated amount, the suction pressure becomes insufficient. In this case, the reduction in suspended solid, droplets and the carcinogenic compounds in the inlet and outlet streams are also observed. The maximum achieved separation efficiency is 55.68%.

From the elemental analysis (Table 15), it identified that a filter containing activated charcoal more efficiently adsorbs selected gas-phase components of MS (carcinogen 4-(methylnitrosamino)-1-(3-pyridyl)-1-butanone (Muscat et al. 2005)). The modified filter also significantly reduces the amounts of specific biologically active vapour phase compounds in the mainstream smoke (acetaldehyde, acrolein, benzene, HCN, NOX, acrylonitrile, and ammonia (Bombick et al. 1997)).

### Both chemical and mechanical modifications

In case of both chemical and mechanical modifications, the filter to cigarette ratio is maintained at 1.5 and different ratios of charcoal and Zeolite-13X mixtures were (1:1, 1:2,1:3 and 1:4 ratio) prepared. These mixtures were positioned as shown in the schematic (Fig. 16). During the experimentation, the suction pressure was maintained at 10 kPa.

Table 17 clearly ensures that both chemical and mechanical modification depict the highest separation efficiency. It is further verified by performing the EDS analysis of the solid sample collected from the smoke after filtration. The EDS analysis (Table 16) corroborates the information reported in Table 17 qualitatively.

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**Table 10** Properties of the adsorbents

| Sl. No. | Adsorbent | Manufacturer | Specific surface area (m²/kg) | Average pore dia (nm) |
|---------|-----------|--------------|-------------------------------|-----------------------|
| 1.      | Charcoal  | Charcoal Activated-250, decolourizing powder, (CAS NO: 7440-44-0), Prod.No.22395, manufactured by Fisher Scientific India Pvt. Ltd., Mumbai-400076. | 2800 | 121 |
| 2.      | Zeolite-13X | Molecular sieves, 13X (Zeolite-13X, CAS No: 63231-69-6), Prod.No. 208647, Merck Millipore and Sigma-Aldrich, Kolkata – 700046. | 165300 | 1.1 |

**Table 11** Adsorption capacity

| Pressure (kPa) | Adsorption capacity (mg/g) | Charcoal | Zeolite-13X | Zeolite-13X(Zhang et al. 2010) |
|---------------|----------------------------|----------|-------------|-------------------------------|
| 6             | 72                         | 12       | 85          |
| 8             | 89                         | 160      | 96          |
| 10            | 91                         | 173      | 120         |
| 12            | 92.5                       | 199.5    | 130.2       |
| 14            | 93.6                       | 210.6    | 140.6       |

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Consumer comfort

The word “taste” is used to indicate the user’s satisfaction level after the modification of cigarette. This was determined by taking the feedback of the consumers after smoking of modified and unmodified cigarette. The testing was conducted with the help of a group of people who consumes cigarettes regularly and according to them, the satisfaction level after the use of modified cigarette was unaltered. The weight increments of the cigarette after modifications are presented in Table 18.

Discussion

For the identification of the efficient methodologies, a comparison has been performed among all (Table 19). For this, the suspended solid removal efficiency is considered as the parameter. The comparison indicates that the combined chemical and mechanical modification methodology separates significant amount of suspended solid from the smoke. However, for the combined case, beyond 1.5 g mixture, the suction pressure becomes insufficient. In addition to the above, the economic analysis also ensures that the increment in the price of the cigarette after modification is about 0.96–6.19 % (Table 20).

Conclusions

Experiments were conducted to investigate the modification methodologies which decline the cigarette’s harmful effect without deteriorating its taste. Initially, the role of smoking parameters was disclosed, and then the chemical, mechanical and combined modification processes on the final quality of smoke were studied. Based on current experimental results, the followings are the conclusion:

| Table 13 | EDX spectra of a filter of cigarette using Zeolite-13X |
|-----------|-----------------------------------------------|
| Elements  | %     | Reduction (%) |
| Hg        | 34    | 3.34          |
| C         | 32.7  | 4             |
| O         | 30    | 21.6          |
| Ni        | 0.3   | 13            |
| Cu        | 0.3   | 40            |
| F         | 0.78  | 48.7          |
| Co        | 0.14  | 6.67          |
| Cd        | 0.15  | 2             |
| Cr        | 0.4   | 1.5           |
| Ca        | 0.22  | 20            |
| Mn        | 0.17  | 1.2           |
| N, K, As, Mg, Pb | 0.65 | –             |

| Table 15 | EDX spectra of a filter of cigarette using activated carbon |
|-----------|-------------------------------------------------------------|
| Elements  | %     | Reduction (%) |
| Hg        | 35.1  | 29            |
| C         | 37.2  | 24.55         |
| O         | 33.8  | 37.78         |
| Cu        | 0.13  | 27            |
| F         | 0.42  | 1.25          |
| Co        | 0.3   | 66.67         |
| K         | 0.6   | 25            |
| Mg        | 0.28  | –             |
| N, Ni, Cr, As, Cl, Cd, Ça, Mn, Pb | 0.09 | –             |

Table 14 The role of charcoal concentration in cigarette performance

| Charcoal concentration (g) | Inlet concentration (μg/m³) | Outlet concentration (μg/m³) | Amount of settlement (μg/m³) | Removal efficiency (%) |
|---------------------------|-----------------------------|-------------------------------|-----------------------------|------------------------|
| 0.25                      | 820                         | 730                           | 90                          | 53                     |
| 0.5                       | 803                         | 740                           | 63                          | 54.37                  |
| 1                         | 780                         | 720                           | 60                          | 55.68                  |
| 1.5                       | Insufficient suction pressure |                               |                             |                        |

Fig. 16 Schematic diagram of a cigarette with catalysts zeolite-13X and charcoal
1. Without any modification, the maximum separation efficiency is achieved if the suction pressure is maintained at 10 kPa and 135 s burning time is maintained.

2. The analysis of ash, flakes and smoke ensures that every stream contains carcinogenic elements in different forms.

3. The P-V analysis indicates that additional (63 ml) smoke goes into the lungs during cigarette smoking. However, only 500 ml comes out during expiration, and the remaining 63 ml comes out in the subsequent breathing. Due to this, the suspended solids and droplet containing 63 ml find sufficient time to settle. Furthermore, the discussed phenomenon enhances the settling velocity up to $5 \times 10^4$ times.

4. To avoid the distillation and condensation of cigarette smoke within the human body, L/U must be 0.15.

5. In mechanical modification, the optimum F/C ratio is found to be 1.5, and the achieved separation efficiency is 51.13%.

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**Table 16** EDX spectra of a filter of cigarette using zeolite-13X and activated charcoal

| Elements | % | Reduction (%) |
|----------|---|---------------|
| Hg       | 26.4 | 25            |
| C        | 39.4 | 25.12         |
| O        | 31.5 | 28.26         |
| Ni       | 0.19 | 26.9          |
| Cu       | 0.31 | 72.23         |
| F        | 0.46 | 4.1           |
| Co       | 0.13 | 13.33         |
| K        | 0.23 | 0.48          |
| Cr       | 0.2  | 1.2           |
| Ca       | 0.28 | 1.3           |
| N, As, Cd, Mg, Mn | 0.69 | –            |

**Table 17** The cigarette performance in case of combined modification

| Zeolite-13X + charcoal concentration (g) | Inlet concentration (μg/m³) | Outlet concentration (μg/m³) | Amount of settlement (μg/m³) | Removal efficiency (%) |
|----------------------------------------|-----------------------------|------------------------------|-----------------------------|------------------------|
| 1:1                                    | 615                         | 602                          | 13                          | 64.8                   |
| 1:2                                    | 605                         | 596                          | 9                           | 65.44                  |
| 1:3                                    | 591                         | 585                          | 6                           | 66.51                  |
| 1.4                                    | Insufficient suction pressure |                              |                             |                        |

**Table 18** Increased cigarette weight in filter modification

| Weight of the cigarette without modification (g) | Weight of the cigarette after modification (g) | Increase in cigarette weight (g) |
|-------------------------------------------------|-----------------------------------------------|---------------------------------|
| 0.9207                                          | 1.089407                                      | 0.168707                        |
| 0.9195                                          | 1.095915                                      | 0.176415                        |
| 0.8856                                          | 1.050940                                      | 0.165340                        |
| 0.8929                                          | 1.061031                                      | 0.168131                        |
| 0.9017                                          | 1.066660                                      | 0.164960                        |

**Table 19** Comparative study

| Name of the methodology | Reduction in harmless effect (μg/m³) | Maximum efficiency (%) | Increased price (%) |
|-------------------------|--------------------------------------|------------------------|---------------------|
| Alteration of smoking methodology | 850-810 | 45.7 | – |
| Mechanical modification | 800-760 | 51.13 | – |
| Chemical modification (Charcoal) | 765-735 | 55.68 | 0.9626 |
| Chemical modification (Zeolite-13X) | 780-771 | 62.5 | 4.7341 |
| Chemical + Mechanical modifications | 591-585 | 66.51 | 2.8484 |
6. In chemical modification, the separation efficiency of 55.68% and 62.5% are achieved in case of charcoal and zeolite 13-X, respectively.
7. The separation efficiency of 66.51% is obtained in the case of combined modifications.
8. The economic analysis indicates that 4.74% rise in the actual cost and per cigarette, the increment in average weight is 0.114 g.

**Table 20** Cost of the cigarette with and without modifications

| Unmodified cigarette ( ) | Mechanical modification ( ) | Chemical modifications |
|--------------------------|-----------------------------|------------------------|
|                          |                             | Charcoal ( )           |
|                          |                             | Zeolite-13X ( )        |
|                          |                             | Charcoal+Zeolite13X ( )|
| 15                       | 15.30                       | 15.14439               |
|                          |                             | 15.71012               |
|                          |                             | 15.92753               |

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