Effects of metal powders on the release of volatiles from electromagnetic pyrolysis of reconstituted tobacco

Yizhen Chen[1,2,3], Yimin Xiong[4], Ziwei Wang[1,2,3], Ben Liu[1,2,3], Yaowei Xu[1,2,3], Song Gao[1,2,3], Zhichang Liu[1,2,3], Qunshan Yan[1,2,3], Yi Wang[4] and Yuxing Tong[1,2,3,*]

[1]China Tobacco Hubei Industrial Co., Ltd., Wuhan, 430040, China
[2]Hubei Xinye Reconstituted Tobacco Development Co., Ltd, Wuhan, 430058, China
[3]Applied Technology Research of Reconstituted Tobacco Hubei Province Key Laboratory, Wuhan, 430040, China
[4]State Key Laboratory of Coal Combustion, School of Energy and Power Engineering, Huazhong University of Science and Technology, Wuhan, 430074, China

*tong_yx@126.com

Abstract. To investigate the effects of metal powders on the release of volatiles from electromagnetic pyrolysis of reconstituted tobacco, experiments involving varying types, proportions, and particle sizes of metal additives were conducted under different heating voltages. Results show that heating voltage plays a predominant role in the release of volatiles during pyrolysis. Increasing heating voltage significantly enhances the total release of volatiles, as well as the release of key constituents such as nicotine, glycerol, and neophytadiene. In comparison to iron powder, nickel powder exhibits a more pronounced ability to promote the release of major constituents and flavor compounds in the volatiles during pyrolysis. Reducing the particle size of nickel powder leads to a moderate decrease in the total release of volatiles but has a limited impact on the composition of the volatiles. Conversely, reducing the particle size of iron powder, while somewhat decreasing the total release of volatiles, promotes the generation of effective constituents in the volatiles. Increasing the proportion of metal powder addition enhances the release of major constituents and flavor compounds in the volatiles, albeit with diminishing returns in terms of the improvement. These findings provide valuable insights for potential applications in tobacco industry processes and product development.

Keywords: Electromagnetic heating, Pyrolysis, Metal powders.

1 Introduction

Due to its relevance to the tobacco industry and its potential implications for public health, research on the pyrolysis process in the context of tobacco reengineering has garnered significant attention [1]. Recombinant tobacco, a composite material derived from tobacco waste and extracts, plays a pivotal role in the production of various tobacco products, including Heat Not Burning (HNB) tobacco. HNB tobacco, owing to

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its lower emission of harmful constituents and reduced health impact, has emerged as a crucial avenue for development and research within the tobacco sector [2-3].

Currently, the heating source for HNB tobacco primarily employs metal resistance heating, whereby electric current passes through metal resistors, generating heat that is subsequently applied to the tobacco, facilitating thermal decomposition and volatiles release. However, the resistance heating method is limited by its relatively slow heating response. Moreover, due to the low thermal conductivity of tobacco, the rate of heat transfer within the tobacco stick is also slow. These factors collectively result in a gradual temperature rise in traditional heating methods, coupled with sluggish instrument responsiveness to temperature control, rendering it challenging to achieve a uniform and rapid tobacco stick heating and volatiles release [4].

To overcome these challenges, electromagnetic heating technology has been applied to heat biomass, solid waste, and other organics [5]. This innovative method employs a controllable electromagnetic field to induce a controlled pyrolysis process, thereby enabling precise control over reaction parameters. In comparison to resistive heating methods, it offers significant advantages, including rapid heating rates [6], precise temperature control [7], and user-friendly operation. Compared to heating with a central heat source using a sheet-like metal, utilizing dispersed metal powder as the heat source effectively ensures even heat distribution throughout the sample, thereby overcoming the issue of uneven sample heating associated with central heat source methods.

Understanding the intricate mechanisms governing tobacco pyrolysis and emissions is paramount to elucidating the complex chemical processes underpinning the formation of volatile compounds in tobacco. Therefore, this study employed dispersed metal powder as a heating medium to investigate the influence of various metal types, proportions, and particle sizes on the emission characteristics of volatile compounds during tobacco thermal decomposition. This research unveils the intricate interplay between additives and the thermal decomposition of reconstituted tobacco.

2 Materials and Methods

2.1 Electromagnetic pyrolysis of reconstituted tobacco

Specialized reconstituted tobacco samples, incorporating varying categories of metal powder types, sizes, and additive ratios, were provided by Hubei Xinye Reconstituted Tobacco Development Co., Ltd. Reagents employed in the experiments included dichloromethane (chromatographic grade, Aladdin), methanol (chromatographic grade, Aladdin), tetrahydrofuran (chromatographic grade, Aladdin), nitrogen and helium gases (purity > 99.999%, Wuhan Huaerwen Industrial Co., Ltd.).

To replicate the authentic thermal decomposition process of tobacco cartridges, we designed a reactor that adhered to the dimensions of real cartridges and carefully configured the pertinent heating parameters. This deliberate approach ensured that the thermal decomposition characteristics of the cartridges during the experimental process closely approximated those encountered in actual smoking scenarios.

The electromagnetic pyrolysis experiments on reconstituted tobacco were conducted within a custom-built fixed-bed quartz reactor, as depicted in Figure 1. The reactor possessed an inner diameter of 8 mm. The heating coil had a height of 20 mm and a diameter of 10 mm, with a total of 8 coil windings. High-purity nitrogen gas (purity > 99.999%) was employed as the carrier gas, with a gas flow rate set at 50 mL·min^{-1}. Before the experiments, approximately 1.000 ± 0.001 g of reconstituted tobacco samples were uniformly packed within the heating zone, followed by the introduction of the carrier gas to initiate the pyrolysis experiments. Based on the experimental results, the yields of char and condensed oil were calculated separately. Char yield (%) was defined as the percentage of char mass relative to the total mass of re-constituted tobacco after pyrolysis, while oil yield (%) was defined as the percentage of condensed oil mass relative to the total mass of reconstituted tobacco after pyrolysis.

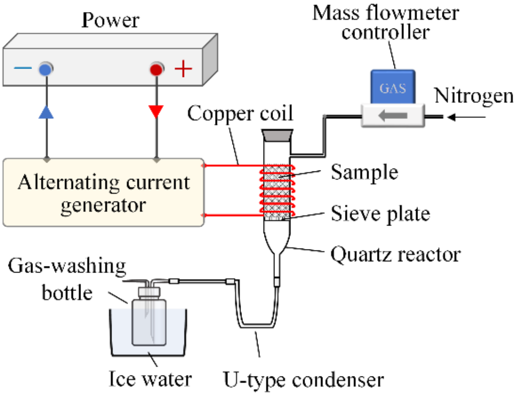

Fig. 1. Schematic diagram of the electromagnetic heating device.

2.2 Product analysis

The molecular weight distribution of components in the pyrolysis oil was determined using gel permeation chromatography with an Agilent 1260II instrument. A refractive index detector was employed, and the gel column used was OligoPore with dimensions of 7.5×300 mm. During testing, the column temperature was maintained at 40 °C, while the detector temperature was set at 45 °C. The mobile phase consisted of tetrahydrofuran (chromatographic grade), with a flow rate of 1 mL·min^{-1} [8].

Characterization analysis of the pyrolysis oil solution was conducted using gas chromatography-mass spectrometry with a Thermo Scientific Trace 1300/ISQ instrument. A capillary column HP-INNOWax (30 m×0.25 mm×0.25 mm) was employed. The injection volume was set at 1 μL, with high-purity helium gas (purity > 99.999%) used as the carrier gas at a flow rate of 1.0 ml·min^{-1} in a splitless injection mode. The mass spectrometer source temperature was 230 °C, and the quadrupole temperature was 150 °C. The mass spectrometry scan range was 15-500 m·z^{-1}, with a mass spectrometry delay time of 4.7 min. The inlet temperature was set at 250 °C. The results were compared to the database of the National Institute of Standards and Technology (NIST, 2014 edition) [9].

3 Results and discussion

3.1 Effect of metal powders on the yield of bio-oil and coke during electromagnetic pyrolysis of reconstituted tobacco

The category, size, and proportion of metal powders themselves constitute crucial process parameters in production. The pyrolysis product yields of different reconstituted tobacco at 12 V voltage are shown in Figure 2. Compared to the electromagnetic heating method that utilizes a thin iron sheet as the central heat source, the metal powder heating method employed in this study allows for the diffusion of the heat source throughout the entire tobacco stick, resulting in a more uniform heating and pyrolysis process.

As depicted in Figure 2, when switching from nickel powder to iron powder, a substantial increase in char yield (averaging 6.18%) is observed, accompanied by a significant reduction in the mass of released volatiles, indicating that nickel powder exhibits superior heat generation and heat transfer effects within the tobacco stick, resulting in a more thorough pyrolytic reaction. However, when increasing the metal additive ratio to 20%, an overall decreasing trend in char yield is observed (averaging 3.48%), suggesting that augmenting the quantity of metal powder can enhance the efficiency of the pyrolytic reaction. Nonetheless, at this higher metal powder concentration, a considerable cost-benefit balance needs to be considered. Surprisingly, reducing the powder particle size to 500 mesh results in an upward trend in char yield (averaging 2.48%), particularly noticeable at lower additive ratios (averaging 3.75%). Decreasing the powder particle size leads to an increase in the number of individual metal powder particles at the same additive ratio. However, the heating efficiency of individual particles significantly diminishes during the electromagnetic heating process, thus reducing the effectiveness of the pyrolytic reaction. The aforementioned results indicate that switching from iron powder to nickel powder can significantly enhance the yield of pyrolysis oil products. Furthermore, increasing the additive ratio and particle size can also enhance the final oil yield.

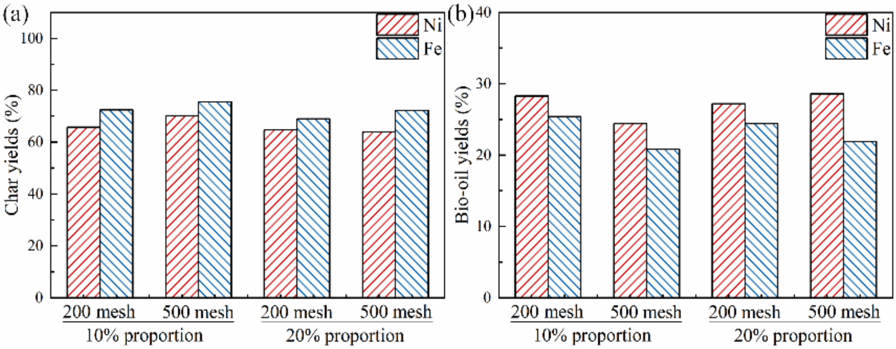

Fig. 2. The pyrolysis products yields of the reconstituted tobacco with different metal powder types, sizes, and addition ratios under 12 V.

Furthermore, the variations in pyrolysis product yields of different reconstituted tobacco under varying heating voltages (8-12 V) were investigated, and the results are presented in Figure 3. As the heating voltage increases, there is a significant augmentation in the yield of pyrolysis oil obtained from reconstituted tobacco. At 12 V voltage, the oil yield increases by more than twofold compared to 8 V, primarily attributed to the substantial temperature rise during electromagnetic pyrolysis with higher external energy input, leading to a rapid increase in the release of volatile components [10-12].

Comparing reconstituted tobacco with the addition of iron powder and those with nickel powder at different voltages, it is evident that the pyrolysis performance of reconstituted tobacco with nickel powder addition surpasses that of iron powder. This further underscores the superior electromagnetic heating capability of nickel powder when compared to iron powder. Additionally, comparing yield variations at different particle sizes reveals that electromagnetic pyrolysis at a particle size of 200 mesh yields a higher oil yield than at 500 mesh. This suggests that, even with variations in external power input, larger particle sizes still exhibit favorable cracking effects.

However, as the heating voltage decreases, the influence of metal category and particle size diminishes gradually. At 8 V voltage, the differences between these factors become less pronounced, indicating that the impact of electromagnetic heating parameters is more significant compared to the properties of the metal powder particles themselves.

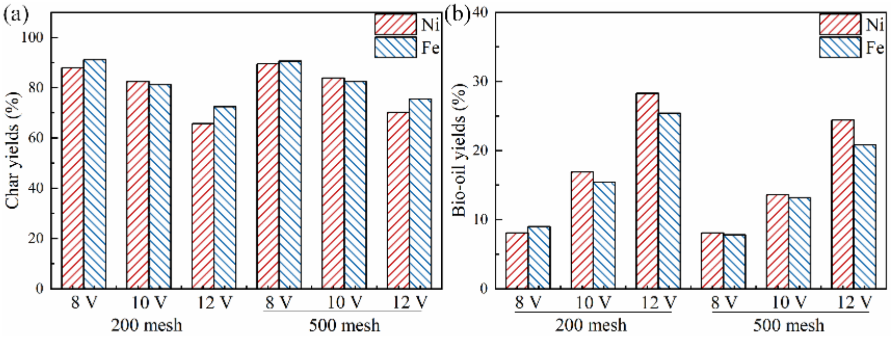

Fig. 3. The pyrolysis products yields of the reconstituted tobacco with different metal powder types and sizes under different voltages with 10% addition ratios.

3.2 Effect of metal powder on molecular weight distribution of bio-oil during electromagnetic pyrolysis of reconstituted tobacco

Molecular weight characterization analysis was conducted on the pyrolysis oils collected through condensation from specialized reconstituted tobacco containing different types of metal powders, various particle sizes, and additive ratios, as illustrated in Figure 4. For nickel powder, the molecular weight distribution peaks of the oil under 200 mesh particle size and 500 mesh particle size exhibited similar primary characteristics, with both peaking at around 280 g·mol^{-1}. However, the signal intensity for the 500 mesh particle size oil was stronger, indicating a higher proportion of molecules with a molecular weight of 280 g·mol^{-1} in the oil compared to the 200 mesh particle size oil, resulting in a lower average molecular weight for the latter.

Furthermore, for nickel powder, under the same particle size, the molecular weight distribution curves of the oils obtained from different additive ratios closely overlapped, suggesting that their molecular weight distribution patterns and average molecular weights were relatively consistent. When combined with yield information, this indicates that, for nickel powder, increasing the particle size can promote the re-lease of volatiles during the pyrolysis process, resulting in a higher presence of larger molecular weight substances in the volatiles. Conversely, increasing the additive ratio, although somewhat increasing the volatiles release, had little impact on the distribution of components in the volatiles.

For iron powder, similar to nickel powder, the molecular weight distribution peaks of the oil under 200 mesh and 500 mesh particle sizes shared comparable characteristics, both peaking at approximately 280 g·mol^{-1}. Again, the 500 mesh particle size oil exhibited stronger signals, suggesting a higher proportion of molecules with a molecular weight of 280 g·mol^{-1} in the oil compared to the 200 mesh particle size oil, resulting in a lower average molecular weight for the latter. However, unlike nickel powder, for iron powder, under the same particle size, lower additive ratios resulted in a higher proportion of molecules with a molecular weight of 280 g·mol^{-1}, signifying relatively smaller molecular weights. Further analysis in conjunction with yield information indicated that, for iron powder, increasing both the particle size and additive ratio can promote the release of volatiles during the pyrolysis process, resulting in a higher presence of larger molecular weight substances in the volatiles.

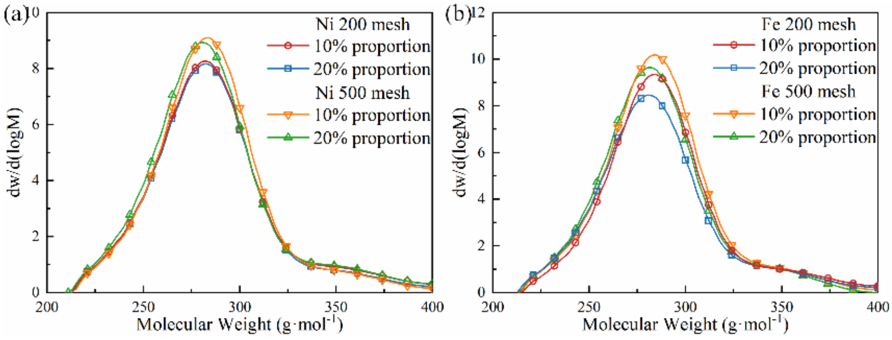

Fig. 4. The molecular weight distribution of the reconstituted tobacco pyrolytic oil with different metal powder types, sizes, and addition ratios under 12 V.

The aforementioned results suggest that during the pyrolysis process of reconstituted tobacco, as the pyrolysis progresses, smaller molecular additives and moisture in the tobacco are initially released. With increasing temperature, the primary substances in the tobacco gradually emerge. During this stage, the released molecules have smaller molecular weights. As the temperature further increases, heavier aromatic compounds also begin to volatilize, leading to an increase in the molecular weight of substances in the volatiles [13-15].

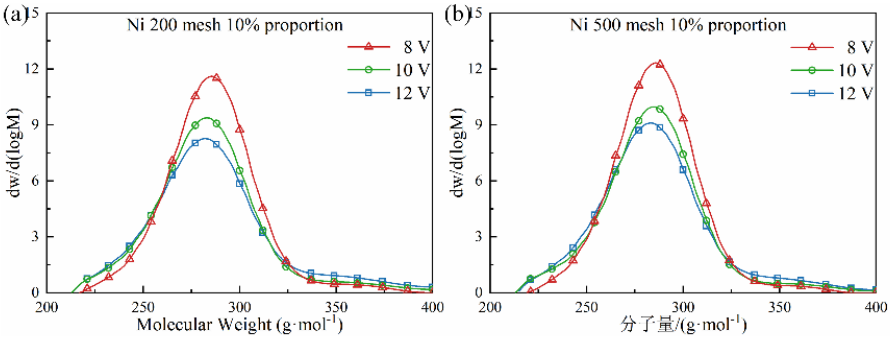

Fig. 5. The molecular weight distribution of the reconstituted tobacco pyrolytic oil with different nickel powder sizes under different voltages with 10% addition ratios.

Further investigation into the molecular weight distribution patterns of pyrolysis oils from different reconstituted tobacco under varying heating voltages was conducted, and the results are presented in Figure 5. The results indicate that, for the same nickel powder additive ratio under different voltages, the molecular weight distribution patterns of pyrolysis oils from reconstituted tobacco with different particle sizes are fundamentally similar. As the heating voltage increases, the proportion of molecules with a molecular weight of 280 g·mol^{-1} gradually decreases, signifying an increase in the average molecular weight. This suggests that with higher heating voltages, the pyrolysis process becomes more intense, leading to the precipitation of larger aromatic macromolecules in the oil. The yield results mentioned earlier demonstrate that with the increase in heating voltage from 8 V to 12 V, the oil yield collected increases by over two-fold. Combined with the molecular weight distribution analysis, it is evident that the higher heating voltage not only facilitates the release of smaller molecular additives, moisture, and effective components in the volatiles but also further promotes the release of aromatic substances in the tobacco. This results in an increased presence of aroma and flavor in the volatiles.

3.3 Effect of metal powder on the distribution of bio-oil component during electromagnetic pyrolysis of reconstituted tobacco

To gain more specific information about the distribution of components in the oil, gas chromatography-mass spectrometry (GC-MS) analysis was conducted on the pyrolysis oils. The distribution of typical components in the volatiles is shown in Figure 6. Under 200 mesh particle size, reconstituted tobacco with nickel powder additives at different ratios exhibited higher levels of major constituents such as glycerol, propylene glycol, and nicotine in the volatiles compared to reconstituted tobacco with iron powder additives. Additionally, the volatiles from reconstituted tobacco with nickel powder contained significantly higher levels of flavor compounds, including neophytadiene, hexadecanoic acid, and octadecanoic acid, indicating a higher release of flavor-related substances in the volatiles when nickel powder was used as the additive. This suggests that, under 200 mesh particle size, nickel powder is more effective than iron powder in promoting the release of various active components from tobacco.

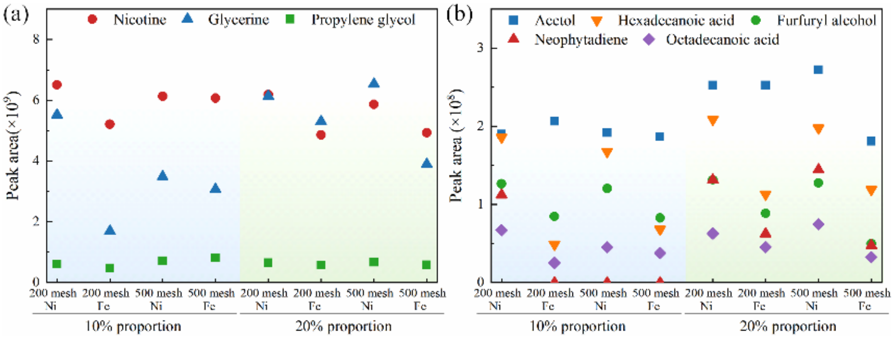

Fig. 6. The peak areas of the species in reconstituted tobacco pyrolytic oil detected by GC-MS with different metal powder types, sizes, and addition ratios under 12 V.

Furthermore, combining the yield and molecular weight distribution information, it was observed that during the pyrolysis process of reconstituted tobacco, as the pyrolysis progresses, initially, the aerosols (glycerol and propylene glycol) and moisture added to the tobacco begin to precipitate. With increasing temperature, the nicotine from the tobacco gradually precipitates. During this stage, the released molecules have smaller molecular weights. As the temperature continues to rise, larger molecular weight aromatic compounds, such as stearic acid and palmitic acid, also begin to volatilize, resulting in increased volatiles yield and an increase in the molecular weight of substances in the volatiles [16].

However, under 500 mesh particle size, reconstituted tobacco with iron powder additives at different ratios exhibited higher levels of major constituents such as nicotine and propylene glycol in the volatiles compared to reconstituted tobacco with nickel powder additives. Nevertheless, the volatiles from reconstituted tobacco with nickel powder still contained significantly higher levels of flavor compounds, including neophytadiene, hexadecanoic acid, and octadecanoic acid, indicating a superior release of flavor-related substances when nickel powder was used as the additive. This suggests that, under lower particle size conditions, iron powder is more effective than nickel powder in promoting the release of major constituents such as nicotine, but nickel powder outperforms iron powder in the release of flavor compounds such as neophytadiene. This observation aligns with the yield information and molecular weight distribution data.

Further investigation into the distribution of specific components in the pyrolysis oils from reconstituted tobacco at different heating voltages is shown in Figure 7. It can be observed that with increasing heating voltage, regardless of the particle size and metal powder additive ratio, the volatiles produced from the pyrolysis of reconstituted tobacco contain significantly higher levels of both major constituents such as nicotine and glycerol, as well as key flavor compounds like neophytadiene, octadecanoic acid, and 2-furan methanol. This result is highly consistent with the previously discussed yield and molecular weight distribution information, indicating that an increase in heating voltage not only enhances the release of aerosols, nicotine, and other effective components in the volatiles but also further promotes the release of heavier aromatic compounds from the tobacco, thereby increasing the aroma and flavor of the volatiles.

Under 200 mesh particle size conditions, reconstituted tobacco with nickel powder additives consistently exhibited higher levels of major constituents such as nicotine and glycerol, as well as flavor compounds like neophytadiene, in the volatiles at different voltages compared to reconstituted tobacco with iron powder additives. Conversely, under 500 mesh particle size conditions, although the levels of substances like nicotine in the volatiles from reconstituted tobacco with nickel powder were lower than those with iron powder additives, the levels of flavor compounds remained relatively high. This observation aligns with the conclusions drawn from the previous analyses.

Furthermore, as the heating voltage decreases, there is a significant decrease in the levels of major constituents like nicotine and flavor compounds such as neophytadiene in the volatiles. Under low heating voltage conditions, the effects attributable to the type and presence of metal powders become markedly weaker. Consequently, heating voltage plays a dominant and decisive role in determining the release of volatiles from the pyrolysis of reconstituted tobacco.

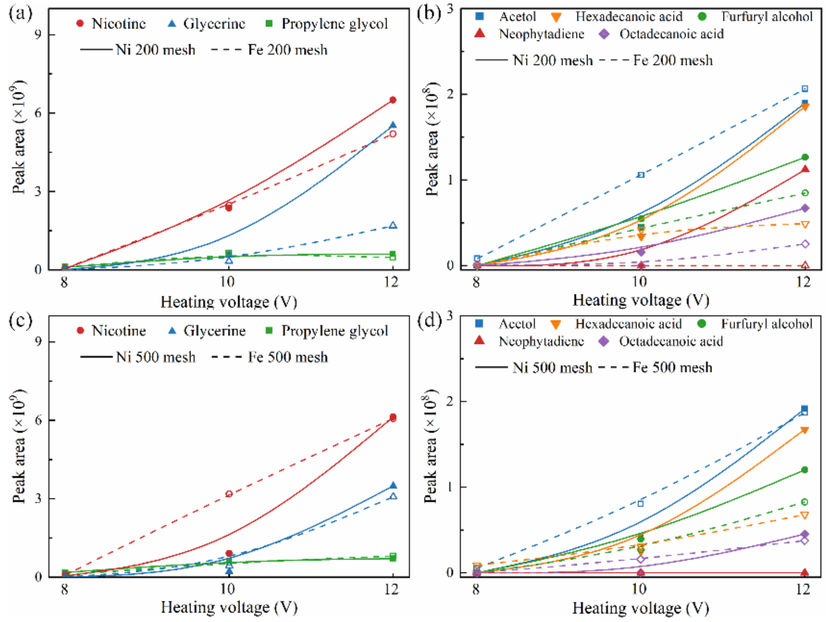

Fig. 7. The peak areas of the species in reconstituted tobacco pyrolytic oil detected by GC-MS with different metal powder types and sizes under different voltages with 10%.

3.4 Perspective

This study substantiates the feasibility of employing metal powder additives in the realm of electromagnetic heating for heat-not-burn tobacco and elucidates the influence of various metal types, proportions, and particle sizes on the release of volatile compounds during tobacco electromagnetic thermal cracking. To advance the progress in this field, we believe it is imperative to explore the following aspects:

1. The impact of the coking characteristics of the metal powder additives on the heat transfer throughout the entire heating process.
2. The effects of introducing metal powder additives on the release of tobacco aerosol.
3. Leveraging the distinctive catalytic properties of metal powder additives to reduce the emission of harmful substances from tobacco.

4 Conclusions

In summary, this research investigated the effects of different metal types, ratios, and particle sizes on the release of volatiles from the electromagnetic pyrolysis of reconstituted tobacco. As the heating voltage increases, the total volume of volatiles released from the pyrolysis of reconstituted tobacco rapidly increases. The composition and molecular weight of the volatiles also increase significantly. This includes major constituents such as nicotine, glycerol, and propylene glycol, as well as flavor compounds like myrcene, stearic acid, and palmitic acid. The addition of nickel powder to reconstituted tobacco results in overall higher levels of total volatile volume, major constituents, and flavor compounds compared to the addition of iron powder. For reconstituted tobacco with nickel powder additives, reducing the particle size of the nickel powder to some extent decreases the total volatiles release, but has little impact on volatiles composition. In the case of reconstituted tobacco with iron powder additives, reducing the particle size of the iron powder also reduces total volatiles release to some extent but promotes the generation of effective components in the volatiles. Increasing the additive ratio of metal powders slightly enhances both the total volatiles release and the content of effective components in the volatiles during the pyrolysis of reconstituted tobacco. However, due to the limited extent of this enhancement, the choice of metal powder additive content needs to be carefully balanced between cost and benefit considerations.

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