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Comparative Kinetic and Thermodynamic Analysis of Biomass-Plastic Blend and the Effect of Activated Carbon Addition Using Isoconversional Methods

SABARIKANTH T[1], RAMESH KASIMANI[2], KULANDAIVEL DURAISAMY[3], AYYAPPAN S[4], SHOBAN BABU M[5]

[1] PG Scholar, Department of Thermal Engineering, Government college of Technology, Coimbatore, Tamil Nadu,641013, India.
[2]Professor, Head of the Department, Department of Mechanical Engineering, Government college of Technology, Coimbatore,Tamil Nadu, 641013, India.
[3]Assistant Professor, Department of Mechanical Engineering, Government College of Engineering, Erode, Tamil Nadu, 638316, India.
[4]Associate Professor, Department of Mechanical Engineering, Government College of Technology, Coimbatore, Tamil Nadu, 641013, India.
[5]Project Associate, Department of Mechanical Engineering, Government College of Technology, Coimbatore, Tamil Nadu, 641013, India.
Corresponding Author:
Sabarikanth T
Email: sabarikanth567@gmail.com

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ABSTRACT

The massive generation of agricultural and post-consumer plastic waste has become a serious global environmental concern. This study presents a comparative thermo-kinetic investigation of two blended feedstocks using thermogravimetric analysis (TGA). Sample PSM was prepared in 1:1 ratio using pistachio shell and shredded surgical mask waste (polypropylene), while Sample PSMC contained the same blend incorporated with 30 wt% activated carbon. Non-isothermal thermogravimetric analysis (TGA) was performed under a nitrogen atmosphere at heating rates of 10, 15, and 25 °C/min to investigate the thermal decomposition behavior of the samples. The kinetic characteristics were analyzed using four widely adopted model-free isoconversional approaches, namely Kissinger–Akahira–Sunose (KAS), Flynn–Wall–Ozawa (FWO), Starink, and Friedman methods. In addition, important thermodynamic properties such as enthalpy change (ΔH), entropy change (ΔS), and Gibbs free energy change (ΔG) were calculated to better understand the thermal reaction mechanism.Results revealed that PSM exhibited lower average activation energy compared to PSMC. The addition of activated carbon increased char yield but slightly raised the energy barrier. The pyrolysis process was found to be endothermic and non-spontaneous in nature. The obtained kinetic and thermodynamic data provide useful insights for optimizing co-pyrolysis of biomass and plastic waste for sustainable energy recovery.

Keywords: Pistachio shell, Surgical mask waste, Polypropylene, Activated carbon, TGA, Pyrolysis kinetics, Activation energy, Isoconversional methods.

1. INTRODUCTION

Rapid urbanization and changing consumption patterns have led to a steep rise in global municipal solid waste generation, estimated at 2.3 billion tons annually in 2025 and projected to reach 3.8 billion tons by 2050. Conventional waste disposal methods such as landfilling, open dumping, and incineration cause severe environmental pollution and fail to recover the embedded energy.

Among various thermochemical methods, pyrolysis stands out as a promising technology for converting waste into valuable energy resources like bio-oil, gas, and char.Blending lignocellulosic biomass with synthetic polymers such as polypropylene can improve overall energy density and product yield due to synergistic effects. In this context, pistachio shell (an agricultural byproduct) and discarded surgical masks (primarily polypropylene) were selected as feedstocks. Activated carbon was added in one sample to study its influence on thermal degradation behavior.

The main aim of this investigation is to study the kinetic parameters and the thermal decomposition characteristics of the prepared blends by using the non-isothermal TGA at different heating rate.

1.1 OBJECTIVES

The main objectives of this research work are as follows:

- To study how pistachio shell and surgical mask plastic blends break down when heated at three different speeds (10, 15, and 25 degrees per minute).
- To measure the amount of energy required for this breakdown, we employ four straightforward comparison methods.
- To determine the thermodynamic parameters (ΔH, ΔS, and ΔG) and assess the energy feasibility of the pyrolysis process.
- To analyse the effect of 30 wt% activated carbon addition on the thermal stability, kinetic parameters, char yield, and overall pyrolysis performance of the blends.

2. REVIEW OF LITERATURE

Numerous studies have examined the pyrolysis kinetics of biomass and plastic materials through thermogravimetric analysis.

Tonbul (2008) evaluated the pyrolysis of pistachio shell using a novel thermogravimetric method and reported a two-step decomposition process with activation energy values confirming its potential as an energy feedstock.

Acikalin (2012) applied both model-fitting and model-free isoconversional methods on pistachio shell pyrolysis and determined activation energies in the range of 140–170 kJ/mol, highlighting the lignocellulosic degradation behaviour.

Vazquez-Castillo et al. (2020) employed KAS and FWO isoconversional methods to investigate the pyrolysis of pistachio shells and reported narrow activation energy ranges between 165 and 169 kJ/mol. They also evaluated thermodynamic parameters (ΔH, ΔG, and ΔS), confirming the endothermic and non-spontaneous nature of the pyrolysis process.

Aguado et al. (2008) conducted a detailed kinetic study on the pyrolysis of polyolefins, particularly polyethylene and polypropylene, and emphasized their high energy density and suitability for fuel production.

Marcilla et al. (2012) investigated the thermal degradation of commercial polypropylene using TGA and reported activation energies in the range of 180–230 kJ/mol with a single-stage decomposition mechanism.

Xue et al. (2017) studied the co-pyrolysis of biomass and polypropylene and observed synergistic effects that reduced activation energy and improved volatile yields compared to individual feedstocks.

Recent studies on COVID-19 related plastic waste have gained importance. A Monash University research team (2022) analysed the pyrolysis of surgical mask waste (polypropylene) using KAS and OFW methods and reported an activation energy of approximately 135 kJ/mol.

Hassan and Hameed (2024) examined the co-pyrolysis of sugarcane bagasse with PET using isoconversional methods and found activation energies ranging between 163 and 221 kJ/mol, demonstrating intermediate behaviour between pure biomass and plastic.

Das (2024) performed a comprehensive kinetic analysis of mixed plastic wastes using Friedman, OFW, and KAS methods and reported multi-stage degradation with activation energies up to 297 kJ/mol.

Mishra and Mohanty (2018) investigated the pyrolysis kinetics of castor seed de-oiled cake and successfully applied isoconversional methods to determine kinetic and thermodynamic parameters for bioenergy applications.

2.1 RESEARCH GAP

Several studies have independently explored the pyrolysis kinetics and thermal degradation behaviour of pistachio shells and polypropylene materials. Researchers reported activation energy values for pistachio shell in the range of 140–170 kJ/mol, while polypropylene showed higher values between 180–230 kJ/mol. Co-pyrolysis of biomass and plastics has been found to exhibit synergistic effects that alter degradation patterns, volatile release, and activation energy profiles.

Recent studies on surgical mask waste (polypropylene) and biomass-plastic blends have highlighted the importance of model-free isoconversional methods (KAS, FWO, Starink, and Friedman) for accurate kinetic analysis. However, limited literature is available on the combined effect of pistachio shell, surgical mask waste, and activated carbon. This study aims to bridge this research gap

3. MATERIALS AND METHODOLOGY

The present study involves the preparation of two different blended feedstocks using pistachio shell and surgical mask-derived polypropylene, with and without activated carbon addition. Detailed experimental procedures were carried out to investigate the thermal degradation behaviour using thermogravimetric analysis (TGA). Model-free isoconversional methods were employed to determine kinetic and thermodynamic parameters, thereby elucidating the influence of activated carbon on the co-pyrolysis process.

3.1 FEEDSTOCK PREPARATION

Pistachio shells (Pistacia vera L.) were collected as agro-waste from household sources. The collected shells were cleaned thoroughly with distilled water to remove surface impurities and dust particles. After cleaning, they were shade-dried for a few days to eliminate moisture content. The dried shells were then pulverized using a mechanical grinder, followed by sieving to obtain a uniform particle size suitable for further analysis

Discarded surgical masks, primarily composed of polypropylene (PP), were collected from a nearby scientific institution. The masks were cleaned with distilled water, disinfected using a mild detergent, air-dried, and the melt-blown polypropylene layer was separated and cut into small pieces.

Two samples were prepared:

- **PSM**: 1:1 (w/w) blend of pistachio shell powder and shredded polypropylene.
- **PSMC**: PSM blend incorporated with 30 wt% activated carbon.

The activated carbon was added to investigate its catalytic effect on pyrolysis behavior.

Figure 3.1 (a)

Figure 3.1 (b)

Figure 3.1 shows the prepared feedstock materials showing (a) pistachio shell powder, (b) shredded polypropylene from surgical masks, and (c) activated carbon used in the study.

3.2 THERMOGRAVIMETRIC ANALYSIS (TGA)

Thermal decomposition analysis was carried out using a Shimadzu DTG-60 simultaneous thermogravimetric and differential thermal analyzer (TGA-DTA). All experiments were conducted in a nitrogen environment with a gas flow rate of 200 mL/min to maintain inert conditions and avoid oxidation during pyrolysis.

Approximately 5–10 mg of each sample was loaded into alumina crucibles and heated from 30°C to 900°C at heating rates of **10, 15, and 25 °C/min** under non-isothermal conditions.

All experiments were repeated to ensure reproducibility. TGA provided mass loss data, while DTA recorded heat flow for identifying thermal transitions. The Figure 3.2 shows the TGA-DTG analyser setup used in this study.

Figure 3.2 Shimadzu DTG-60 TGA-DTA analyzer

3.3 KINETIC MODELING

The fractional conversion (α) was determined using the following relationship:

$$\alpha = \frac{m_i - m_t}{m_i - m_f}$$

where m_i, m_t, and $m_f$represent the initial sample mass, mass at time t, and final residual mass, respectively. The kinetic parameters were evaluated using four widely adopted model-free isoconversional approaches, namely Kissinger–Akahira–Sunose (KAS), Flynn–Wall–Ozawa (FWO/OFW), Starink, and Friedman methods (Vyazovkin et al., 2011; Mishra and Mohanty, 2018).

The detailed expressions and plotting methods for each are summarized in **Table 3.1**

Method	**Expression**	**Plot**	**Slope**
KAS	$\ln(\beta/T^2) = \text{const} - E_a/RT$	$\ln(\beta/T^2)$vs $1/T$	$-E_a/R$
FWO	$\ln(\beta) = \text{const} - 1.052E_a/RT$	$\ln(\beta)$vs $1/T$	$-1.052E_a/R$
Starink	$\ln(\beta/T^{1.92})$ =const – (1.0008 E_a / RT	$\ln(\beta/T^{1.92})$vs $1/T$	$-1.0008E_a/R$
Friedman	$\ln(d\alpha/dt) = \ln[Af(\alpha)] - E_a/RT$	$\ln(d\alpha/dt)$vs $1/T$	$-E_a/R$

Table 3.1 Isoconversional kinetic methods used in the study.

3.4 THERMODYNAMIC PARAMETERS

hermodynamic analysis plays a vital role in understanding the energy requirements, thermal behaviour, and feasibility of the pyrolysis reaction. In the present work, the key thermodynamic properties — change in enthalpy (ΔH), entropy (ΔS), and Gibbs free energy (ΔG) — were estimated at the peak decomposition temperature (T_m) using the activation energy (E_a) and pre-exponential factor (A) derived from the isoconversional methods.

These parameters were computed using the following standard relations:

- $\Delta H = E_a - RT_m$
- $\Delta G = E_a + RT_m \ln\left(\frac{K_B T_m}{hA}\right)$
- $\Delta S = \frac{\Delta H - \Delta G}{T_m}$

where $R$is the universal gas constant (8.314 J/mol·K), $K_B$is the Boltzmann constant, $h$is Planck's constant, $T_m$is the peak temperature in Kelvin, $E_a$is the activation energy, and $A$is the pre-exponential factor.

The calculated values of ΔH, ΔS, and ΔG helped assess whether the pyrolysis process is endothermic or exothermic, spontaneous or non-spontaneous, and the degree of disorder during thermal decomposition. These results were further used to compare the thermodynamic behaviour of PSM and PSMC samples and to evaluate the influence of activated carbon addition on the overall feasibility of the co-pyrolysis process.

4. RESULTS AND DISCUSSION

4.1 TGA and DTG Analysis

Thermogravimetric (TGA) and derivative thermogravimetric (DTG) analyses were performed on two prepared blends — PSM (1:1 ratio of pistachio shell and polypropylene) and PSMC (PSM with 30 wt% activated carbon) — at three different heating rates of 10, 15, and 25 °C/min under inert nitrogen atmosphere. The thermal degradation of both samples occurred in three distinct stages.

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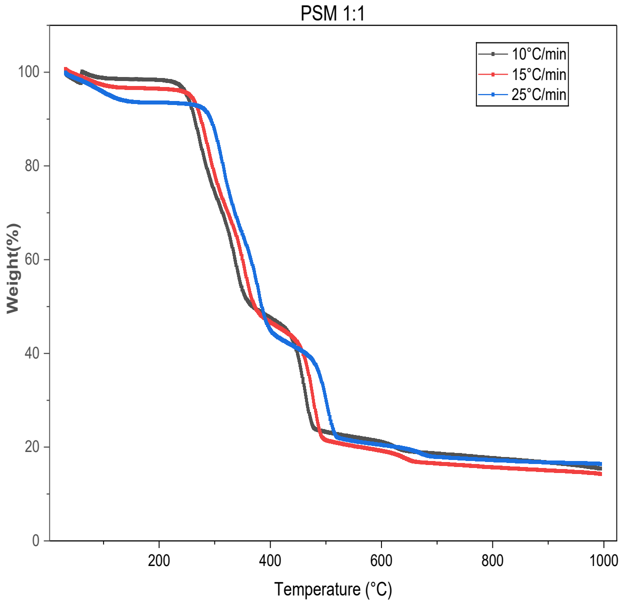

Figure 4.1 (a) TGA curves of PSM at different heating rates

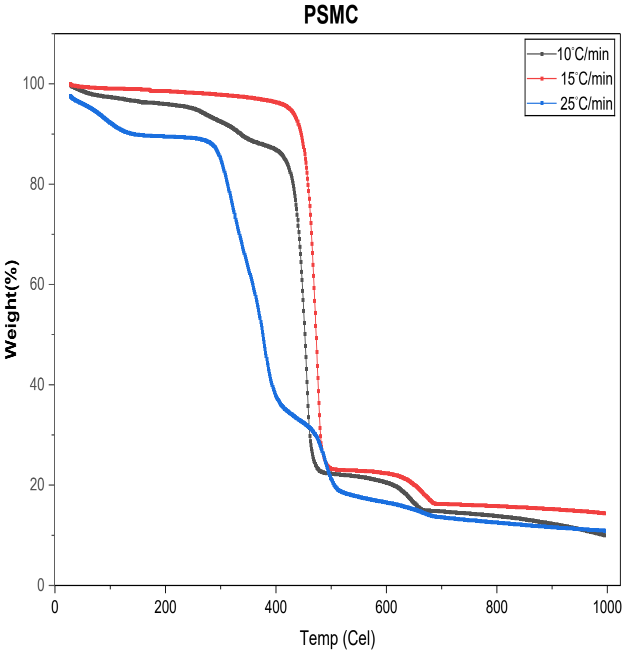

Figure 4.1 (b) TGA curves of PSMC at different heating rates

As observed from **Figure 4.1 (a)** the PSM sample exhibited rapid mass loss in the active pyrolysis zone (230–500 °C), with nearly 95% mass loss completed by 500 °C. In contrast, **Figure 4.1 (b)** shows that the PSMC sample (with 30% activated carbon) demonstrated higher thermal stability, slower decomposition rate, and higher final char residue (10–15%) compared to PSM (5–8%).

The DTG curves revealed that the peak decomposition temperature shifted to higher values with increasing heating rate due to thermal lag. The addition of activated carbon broadened the DTG peak and shifted it from ~340–350 °C (PSM) to ~360–380 °C (PSMC). This indicates that activated carbon acts primarily as a thermal sink and diffusion barrier rather than a strong catalyst under the studied conditions.

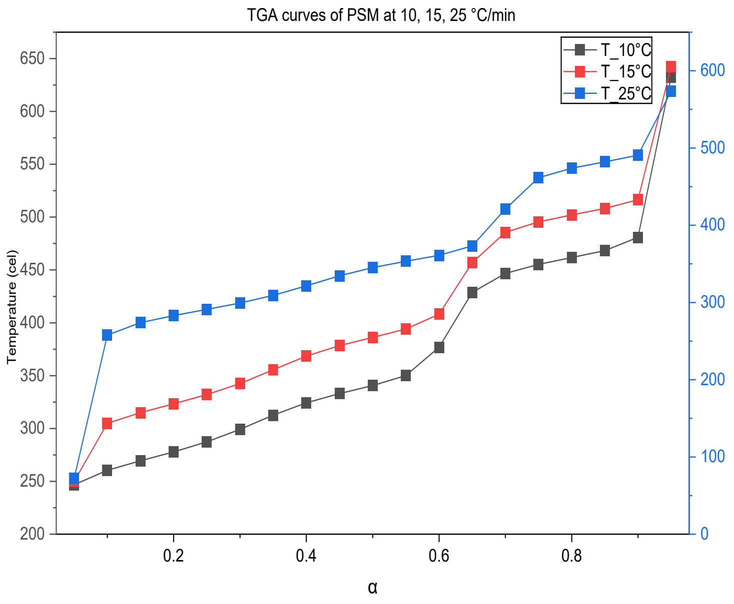

Figure 4.2 (a) Isoconversional α–T profiles for PSM

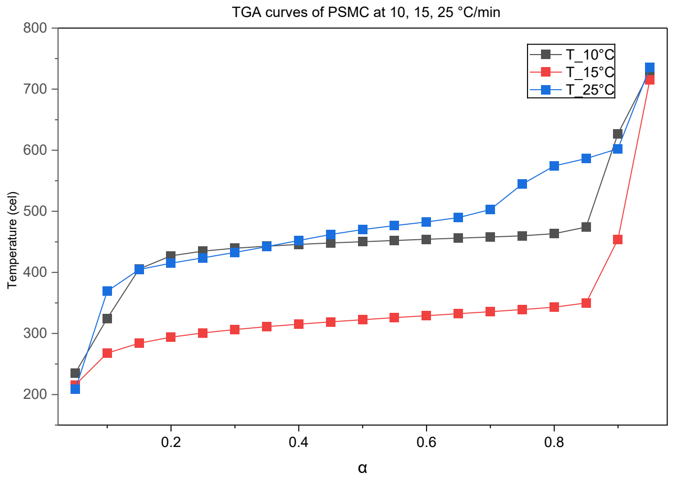

Figure 4.2 (b) Isoconversional α–T profiles for PSMC

Figure 4.2 presents the relationship between conversion (α) and temperature at different heating rates for both samples. It is observed that PSMC required higher temperatures than PSM to reach the same conversion level (especially at $\alpha = 0.1$–0.9), further confirming the increased thermal stability due to activated carbon addition. The gap between the curves reduced at the highest heating rate (25 °C/min), suggesting that the effect of activated carbon becomes less prominent under rapid heating conditions.

4.2 KINETIC ANALYSIS

The thermal degradation kinetics of PSM and PSMC samples were investigated using four model-free isoconversional methods: **Kissinger-Akahira-Sunose (KAS), Flynn-Wall-Ozawa (FWO), Starink, and Friedman.** These methods were applied over the conversion range of $\alpha = 0.05$ to 0.95 based on non-isothermal TGA data obtained at heating rates of 10, 15, and 25 °C/min.

The isoconversional plots ($\ln(\beta/T^2)$ vs. $1/T$ for KAS, $\ln(\beta)$ vs. $1/T$ for FWO, $\ln(\beta/T^{1.92})$ vs. $1/T$ for Starink, and $\ln(d\alpha/dt)$ vs. $1/T$ for Friedman) exhibited high linearity with R^2 values generally exceeding 0.95, particularly in the active conversion range ($\alpha = 0.1$–0.8). This high linearity validates the reliability of the model-free approach for studying the complex co-pyrolysis behaviour of biomass-plastic blends.

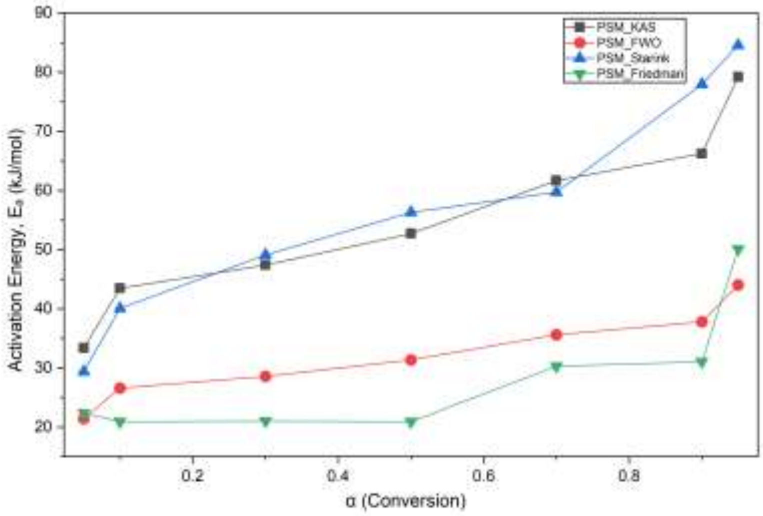

Figure 4.3 (a) Plots of activation energy versus conversion for PSM sample.

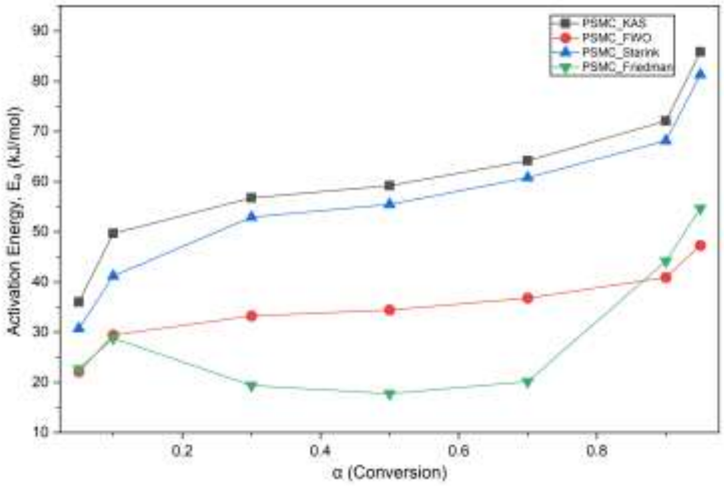

Figure 4.3 (b) Plots of activation energy versus conversion for PSMC sample.

As evident from **Figure 4.3**, the activation energy rose gradually as the conversion increased for both samples. This pattern suggests that the thermal degradation of the biomass-plastic blend follows a complex multi-step mechanism, which is typical when lignocellulosic materials such as pistachio shell are blended with polypropylene.

The average activation energy values obtained from the four methods are summarized in **Table 4.1.**

Method	**PSM (kJ/mol)**	**PSMC (kJ/mol)**	**Difference (%**
KAS	52.71	58.14	+10.3
Starink	50.30	54.96	+9.3
FWO	31.36	34.12	+8.8
Friedman	23.24	20.90	-10.1

Table 4.1 Average activation energy (kJ/mol) for PSM and PSMC using different isoconversional methods

The Starink method, widely regarded as one of the most accurate integral isoconversional approaches [Starink, 2003], yielded average activation energies of 50.30 kJ/mol for PSM and 54.96 kJ/mol for PSMC. PSM consistently exhibited lower activation energy compared to PSMC across the reliable integral methods (average increase of ~9%). The addition of 30 wt% activated carbon moderately raised the energy barrier, mainly due to physical effects such as adsorption of volatile species on the porous surface of activated carbon and increased diffusion resistance.

It is observed that the **integral methods (KAS, Starink, and FWO)** showed a consistent increase in activation energy for the PSMC sample. However, the **differential Friedman method** showed a slight decrease. This variation is common in literature because integral methods give an average value over the entire heating process, while the Friedman method is more sensitive to instantaneous reaction rates and local changes occurring at each conversion point.

The moderate increase in activation energy observed in the integral methods suggests that the addition of activated carbon raises the overall energy barrier, mainly due to physical effects such as adsorption of volatile species on its porous surface and increased diffusion resistance within the sample matrix. Nevertheless, the activated carbon also promoted higher char formation, as observed in TGA results.

4.3 THERMODYNAMIC ANALYSIS

Thermodynamic parameters such as the change in enthalpy (ΔH), entropy (ΔS), and Gibbs free energy (ΔG) were calculated at the peak decomposition temperature ($T_m \approx 778$ K). These calculations were performed using the activation energy (E_a) and pre-exponential factor (A) obtained primarily from the KAS and Starink methods. The results are summarized in **Table 4.2.**

Parameter	**PSM**	**PSMC**	**Difference**
ΔH (kJ/mol)	46.24	51.68	+11.8%
ΔS (J/mol·K)	-188.42	-187.60	Similar
ΔG (kJ/mol)	192.83	197.63	+2.5%

Table 4.2 Average thermodynamic parameters for PSM and PSMC

Positive values of ΔH confirmed the endothermic nature of the pyrolysis process, indicating the requirement of continuous external heat supply. Large negative ΔS values suggested a highly ordered transition state during decomposition. The Positive values of ΔG suggest that the pyrolysis reaction does not proceed spontaneously under the investigated experimental conditions and requires continuous external heat energy for decomposition to occur.. PSM showed slightly lower ΔH and ΔG values, suggesting marginally better thermodynamic feasibility compared to PSMC.

Overall, the kinetic and thermodynamic results reveal that the blend without activated carbon (PSM) offers lower energy barriers and better reactivity, making it more suitable for energy-efficient pyrolysis aimed at higher volatile/bio-oil yields. In contrast, the addition of 30 wt% activated carbon in PSMC enhances char formation and process stability at the cost of a moderate increase in energy demand, primarily through physical adsorption and diffusion effects.

4.4 OVERALL DISCUSSION

The comprehensive kinetic and thermodynamic analysis reveals distinct thermal degradation behaviours between the two blended feedstocks. The PSM sample (1:1 pistachio shell + polypropylene) consistently demonstrated lower activation energy, lower enthalpy change (ΔH), and marginally lower Gibbs free energy (ΔG) compared to the PSMC sample across most isoconversional methods. This indicates that the absence of activated carbon results in easier initiation of thermal decomposition and better overall reactivity.

The increasing trend of activation energy with conversion (α) observed in both samples confirms the complex, multi-step nature of the co-pyrolysis process. Different components (hemicellulose, cellulose, lignin from pistachio shell, and polypropylene chains) degrade at different temperature regimes, leading to varying energy requirements at different stages of conversion. The moderate increase in activation energy (≈ 8–10%) upon addition of 30 wt% activated carbon in PSMC is primarily attributed to physical phenomena such as adsorption of volatile fragments on the porous surface of activated carbon and increased mass transfer resistance, rather than any strong chemical catalytic effect. This is further supported by the broadened DTG peaks and higher char residue observed in PSMC.

From the thermodynamic perspective, the positive values of ΔH for both samples confirm the endothermic nature of the pyrolysis process, necessitating continuous external heat input. The large negative ΔS values suggest the formation of a highly ordered activated complex during decomposition, which is typical for solid-state reactions involving char formation and volatile release. The positive ΔG values shows pyrolysis reaction is non-spontaneous over the experimental environment and requires sustained thermal energy to proceed. Notably, PSM exhibited relatively better thermodynamic feasibility compared to PSMC.

Comparative Performance Summary

- **PSM (without activated carbon)**: Lower energy barrier, faster decomposition kinetics, sharper DTG peaks, and higher volatile release potential. This makes it more suitable for energy-efficient pyrolysis processes targeting maximum bio-oil and syngas yields.
- **PSMC (with 30% activated carbon)**: Higher thermal stability, increased char yield (10–15%), and improved process stability. Although it demands slightly higher energy input, it may offer advantages in applications requiring in-situ tar cracking, volatile adsorption, or higher biochar production.

The synergistic interaction between lignocellulosic biomass and polypropylene was evident, as the blends exhibited degradation behaviour intermediate between pure biomass and pure plastic. However, the addition of activated carbon did not show strong catalytic enhancement under the studied non-isothermal conditions. Instead, its role appears predominantly physical, influencing heat and mass transfer within the sample matrix.

The results of this study hold significant practical value for waste-to-energy systems. The kinetic and thermodynamic data generated here can be effectively utilized for designing, optimizing, and scaling up co-pyrolysis reactors to convert agricultural residues and plastic wastes (including used surgical masks) into useful energy products.Future studies may explore the effect of lower activated carbon dosages, different catalyst types, or isothermal conditions to further enhance product selectivity and process efficiency.

5. CONCLUSION

This study successfully demonstrated the thermal degradation behaviour of pistachio shell–polypropylene blends with and without activated carbon using non-isothermal TGA. The PSM blend exhibited lower activation energy and better thermodynamic feasibility, making it more suitable for energy-efficient pyrolysis. The addition of 30 wt% activated carbon improved char yield but increased the energy barrier moderately. The kinetic and thermodynamic parameters obtained in this work provide valuable insights for the design and optimization of co-pyrolysis reactors for sustainable valorization of agricultural and plastic waste.

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