

XYLOGLUCAN AS A SUSTAINABLE BIO-BASED BINDER FOR CONDENSED AEROSOL FIRE SUPPRESSION

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ABSTRACT

Condensed aerosol fire extinguishing systems (CAFES) are a leading halon-free option for protecting enclosed spaces, but the binder that holds their pyrotechnic charge together remains largely petrochemical. This work tests whether an unmodified plant polysaccharide can replace such a binder without loss of firefighting performance. Dextrin and xyloglucan were each loaded at 3 wt.% into a gallic acid-KNO₃-KClO₃ aerosol-forming composite and compared, under identical conditions, with carboxymethyl cellulose sodium salt (CMC) and hydroxyethyl cellulose (HEC). Thermogravimetric analysis (TGA), differential scanning calorimetry (DSC), bomb calorimetry, uniaxial compression testing, and 4.57 kW n-heptane pool-fire trials were used to rank the formulations. Xyloglucan gave the best overall performance: burn rate 5.72 mm s⁻¹, ignition at 217 °C, flame temperature 418 °C, compressive modulus 184 MPa (31% above the CMC benchmark), and extinction of the pool fire in 35 s, whereas the CMC-bound composite failed to extinguish it. Its favourable oxygen balance (−117.7%) contributes to its fast burn rate, though suppression performance ultimately tracked extinction time and mechanical strength more closely than oxygen balance alone. Because xyloglucan is recovered without derivatisation from tamarind kernel powder, an agro-processing residue, its use removes hazardous synthesis reagents from the binder supply chain. The study supplies comparative evidence for substituting renewable feedstocks for synthetic binders in energetic formulations and identifies compressive modulus as a practical selection criterion alongside extinction time. The work is aligned with SDG-12: Responsible Consumption and Production.

Keywords: Xyloglucan, Bio-based Binder, Aerosol-forming Composite, Condensed Aerosol Fire Extinguishing System, Gallic Acid Pyrotechnics, SDG-12.

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INTRODUCTION

Firefighting technologies have evolved substantially over the past century, driven by the need for effective, rapid, and versatile fire suppression methods across different environments and hazards. Early firefighting relied primarily on water and rudimentary mechanical systems, which were largely inadequate for electrical or chemical fires.¹ A transformative advancement in fire suppression technology emerged during the mid-twentieth century with the introduction of halon-based extinguishing systems, which came to be widely adopted across high-risk sectors including industrial facilities, aerospace engineering, and military applications.² These compounds gained particular recognition for their exceptional efficacy in combating Class C fires—a critical consideration for protecting sensitive electronic infrastructure—while simultaneously demonstrating remarkable performance against Class A and B fire scenarios.³ This represented a substantial technological evolution in proactive fire mitigation strategies. Halon compounds—typically Halon 1211 (bromochlorodifluoromethane) and Halon 1301 (bromotrifluoromethane) are halogenated hydrocarbons containing bromine, fluorine, chlorine, and carbon. These compounds suppress fire primarily by chemically disrupting the free radical chain reaction occurring during combustion.⁴ Despite their efficacy, concerns regarding the environmental consequences of halon usage began to emerge in the 1970s. Groundbreaking work by Molina and Rowland (1973) identified that halogenated compounds, including halons, contribute significantly to ozone layer depletion.⁵ Subsequent atmospheric studies confirmed the long atmospheric lifetimes and high ozone depletion potentials (ODPs) of halons, making them some of the most environmentally hazardous fire suppression agents.^{6,7} Confronted with mounting ecological dangers, the 1987 Montreal Protocol emerged as a pivotal international accord, designed to systematically eliminate the manufacture and use of

substances responsible for ozone layer degradation, notably halons.^{8,9} Under its provisions, industrialized nations were mandated to halt all halon production by 1994, with developing countries granted an extended compliance period until 2010. This decisive policy transformation consequently precipitated a critical demand for alternative fire suppression agents that could match the efficacy of halons without their detrimental environmental impact. One such innovative technology is the Condensed Aerosol Fire Extinguishing System (CAFES), developed as an eco-friendly replacement and notably adopted in high-stakes applications like the Soyuz space program.¹⁰ CAFES operates by dispersing fine aerosol particles that interfere with the combustion process, without depleting the ozone layer or producing toxic residues.^{11,12} These systems are based on Aerosol Forming Composites (AFCs)-pyrotechnic formulations composed of a fuel (reducing agent), an oxidizer, a binder, and other performance-enhancing additives.^{13,14} The operation of AFCs is grounded in redox chemistry: the reducing agent donates electrons during combustion, while the oxidizer decomposes thermally to supply oxygen. The reaction produces heat and forms non-combustible gases and micron-scale solid particles (aerosols), which suppress flames by cooling and chemically interrupting the fire process.^{15,16} The binder component plays a critical role by ensuring homogeneity, structural integrity, and shelf stability of the formulation.¹⁷ Furthermore, binders influence parameters like ignition sensitivity, combustion rate, and mechanical robustness.^{18,19} Historically, the binder function has been met by synthetic macromolecules such as carboxymethyl cellulose sodium salt (CMC), hydroxyethyl cellulose (HEC), phenol-formaldehyde resin, and ethylcellulose, which are valued for their adhesive strength, ready availability, and processing convenience. Their drawbacks are, however, well documented: the feedstocks are petrochemical, the polymers resist biodegradation, and their manufacture depends on hazardous reagents such as chloroacetic acid and ethylene oxide.^{20,21} Combustion of such binders has been linked to the release of volatile organic compounds and to elevated particulate loading in the discharged aerosol.²⁵ These concerns have redirected attention towards binders of biological origin. Pathak and co-workers first established the viability of this route by substituting tannic acid into a gallic acid-based aerosol-forming composite and recording simultaneous gains in suppression efficiency, hygroscopic stability and grain integrity.²⁰ The same group subsequently extended the concept to guar gum²² and, more recently, to inulin³², while maltose³¹ and stearic-acid/dry-heat-modified potato starch³³ have been reported by others as combustible binders for the same class of composite. Naturally occurring polysaccharides, including dextrin, starch, gum arabic and xyloglucan, are attractive in this context because they are renewable, biodegradable and oxygen-rich, and because their viscosity and film-forming behaviour can be tuned without recourse to aggressive chemistry.^{23,24} They are also chemically compatible with gallic acid, a plant-derived phenolic fuel whose exothermic decarboxylation supplies the carbon skeleton for the suppressing aerosol.^{20,25} Dextrin has a long record of use in star pellets, crackling compositions and display fireworks, where its low cost, water solubility and grain-consolidating ability are well established.^{26,27} Xyloglucan, the storage hemicellulose of tamarind (*Tamarindus indica*) kernel, is of particular interest here: it is recovered from a by-product of the tamarind pulp industry, requires no chemical derivatisation, and shows adhesive and film-forming behaviour that has already been exploited in firecracker and flash-powder compositions.^{28, 29, 30} Two gaps remain in this body of work. First, the bio-based binders reported so far have each been assessed in isolation, so no study places a set of natural and synthetic binders on a common experimental footing within the same gallic acid-KNO₃-KClO₃ matrix, using identical specimen geometry and an identical fire load. Comparisons drawn across separate publications are therefore confounded by differences in oxidiser ratio, grain dimensions and pan size. Second, xyloglucan has not previously been evaluated in an aerosol-forming composite at all, despite its documented adhesive performance in unrelated fields; consequently, nothing is known about its oxygen balance, ignition behaviour or suppression efficiency in an energetic system. A third, more practical gap cuts across both: compressive integrity, which determines whether a pyrotechnic grain survives storage, transport and handling, is seldom reported alongside extinction data, leaving binder selection without a mechanical criterion. The broader significance of this work lies in demonstrating that sustainability and performance need not be traded against one another in energetic formulations. If a renewable, unmodified binder can match or exceed a synthetic cellulose ether on burn rate, mechanical strength and extinction time, the case for retaining petrochemical binders in CAFES devices weakens considerably. The

comparative dataset reported here is intended to give formulators, device manufacturers and standards bodies a defensible basis for that substitution, and to open a route for subsequent work on functionalised polysaccharide binders. The work is aligned with Sustainable Development Goal 12 (SDG-12): Responsible Consumption and Production.³⁴ Target 12.4 seeks the environmentally sound management of chemicals throughout their life cycle and a substantial reduction in their release, while Target 12.5 calls for waste generation to be reduced through prevention, recycling and reuse. Replacing a petrochemical binder synthesised by way of chloroacetic acid or ethylene oxide with a polysaccharide recovered from tamarind kernel powder - an agro-processing residue with little competing commercial value - addresses both targets at once, since it removes hazardous reagents from the binder supply chain while converting a waste stream into a functional material. By improving the effectiveness of ozone-benign fire protection for enclosed occupancies, the study also contributes indirectly to SDG-9 (Industry, Innovation and Infrastructure) and SDG-11 (Sustainable Cities and Communities). This study holds significant implications for the development of more sustainable and environmentally friendly fire suppression technologies. By exploring the potential of natural binders, the work contributes to the reduction of synthetic materials and promotes the adoption of eco-friendly practices in the field of pyrotechnics. Additionally, successful implementation of natural binder-based PyFs could lead to improved fire safety and reduced environmental impact.

EXPERIMENTAL

Chemical Reagents and Sources

The reagents used were of appropriate purity, with details regarding their source, grade, and characteristics provided in Table-1.

Table-1: Grade, Source, and Physicochemical Characteristics of the Raw Materials

Chemical Reagents	Grade	Molecular Formula	Melting Point (°C)	Density (g/cm ³)	ΔH _f (kJ/mol)	Source
Gallic Acid (GA)	Analytical Reagent	C ₇ H ₆ O ₅	260	1.70	-769.29	Hi Media Chemicals
Potassium Nitrate (PN)	Analytical Reagent	KNO ₃	334	2.11	-494	Hi Media Chemicals
Potassium Chlorate (PC)	ACS Reagent	KClO ₃	356	2.34	-391.20	Sigma Aldrich
Carboxymethyl cellulose sodium salt (CMC)	High Viscosity	(C ₈ H ₁₁ NaO ₇) _{n=1}	274	1.6	-2573	Hi Media Chemicals
Hydroxyethyl cellulose (HEC)	Low Viscosity (145 mPa s)	(C ₁₂ H ₂₁ O ₈) _{n=1}	140	0.60	-1435	Hi Media Chemicals
Dextrin (DEX)	Analytical Reagent	(C ₆ H ₁₀ O ₅) _{n=1}	192	1.5	-1332	Nice
Xyloglucan (XYL)	Analytical Reagent	(C ₅₁ H ₈₆ O ₄₃) _{n=1}	176.47	1.5	-990	Megazyme Ltd

Preparation of Formulation

The preparation began with accurately weighing the required ingredients in accordance with the mass percentages listed in Table-2. These components were carefully ground using a mortar and pestle to achieve a fine particle size. To ensure uniformity in particle distribution, the ground mixture was then passed through standardized 400-micron sieves.²⁰

Following this, the sieved powder was gradually combined with a suitable solvent and mixed thoroughly to form a homogeneous dough, with consistent mixing and chopping to maintain uniform texture. The resulting dough was cast into three distinct shapes-cuboid (25 mm × 25 mm × 6 mm), strand (65 mm × 8 mm × 5 mm), and cube (50 mm × 50 mm × 50 mm)-using precision glass molds to ensure accurate dimensions. After shaping, the specimens underwent drying in a vacuum oven to eliminate residual moisture. Once dried, they were stored in a desiccator to prevent reabsorption of moisture, thereby ensuring their long-term stability.

Table-2: Composition of the Pyrotechnic Formulations, PyF-1 to PyF-5 (mass %)

Ingredient		GA	PN	PC	CMC	DEX	HEC	XYL
Component Content (mass %)	PyF-1	50	25	25	—	—	—	—
	PyF-2	49	24	24	3	—	—	—
	PyF-3	49	24	24	—	3	—	—
	PyF-4	49	24	24	—	—	3	—
	PyF-5	49	24	24	—	—	—	3

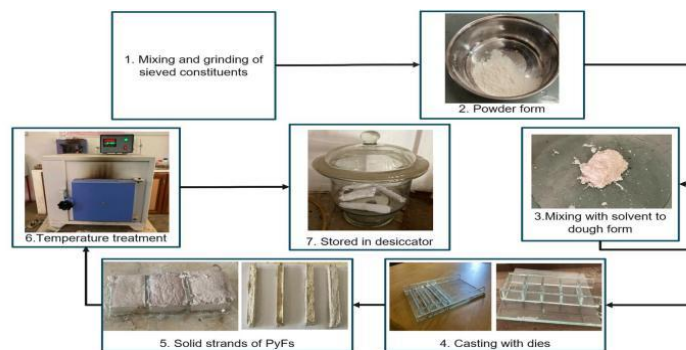


Fig.-1: Preparation of Pyrotechnic Formulation

Characterization and Experimental Analysis

To comprehensively evaluate the pyrotechnic formulations (PyFs), a series of characterization techniques and experimental procedures were conducted.

Thermal Analysis

To characterize the thermal degradation behavior and stability profiles of the prepared formulations, thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were employed. All measurements were conducted using a PerkinElmer STA 8000 analytical instrument under a controlled nitrogen environment. Samples underwent heating from 50 °C to 800 °C at a constant ramp rate of 10 °C min⁻¹. For the DSC investigations, aliquots of 2–4 mg were analyzed with a 50 mL min⁻¹ nitrogen purge, maintaining the identical heating rate to accurately determine ignition temperatures.

Calorific Value Determination

Energy content quantification for all specimens was conducted through bomb calorimetry employing a Mahler instrument. To establish measurement reliability, triplicate trials were executed for every formulation. Each experimental run consisted of precisely weighing a 1 g sample into a stainless-steel crucible, accompanied by an ignition filament for combustion initiation. The sealed calorimeter vessel was pressurized with oxygen to 300 Pa and submerged within a hydrated enclosure. Following ignition, the liberated thermal energy produced a quantifiable temperature increase in the surrounding aqueous medium, enabling subsequent determination of the material's energy density expressed in J g⁻¹.

Burn Rate Analysis

The burn rate of each pyrotechnic formulation (PyF) was determined using the strand-burning procedure described by Pathak *et al.*²⁰ Solid specimens of standardised dimensions (65 mm length × 8 mm width × 5 mm thickness) were used throughout. The time required for complete combustion of each strand was recorded manually and independently verified from video footage (Fig.-3). Three strands were burned per formulation and the mean value is reported. The linear burn rate was calculated as:

$$\text{Burn Rate} = [\text{Strand Length} / \text{Combustion Time}] \quad (1)$$

Figure-2 illustrates the experimental arrangement and the measurement procedure adopted in this study. All numerical results obtained from the tests described in this section are collected in a single consolidated table (Table-3) presented in the Results and Discussion section, so that no dataset appears in both a table and a figure.

Table-3: Consolidated Combustion, Thermal and Fire-Suppression Performance of Formulations PyF-1 to PyF-5

Formulation	Burn Time (s)	Burn Rate (mm/s)	Calorific Value (J/g)	Ignition Temperature (°C)	Residual Mass at 500°C (%)	Combustion Flame Temperature (°C)	Extinction Time (s)
PyF-1	9	5.93	6838	305	28.00	140	22
PyF-2	14	5.54	6668	218	48.49	136	Not extinguished
PyF-3	16	3.30	6975	216	41.48	420	43
PyF-4	20	1.40	7338	220	57.72	487	52
PyF-5	11	5.72	7025	217	36.51	418	35

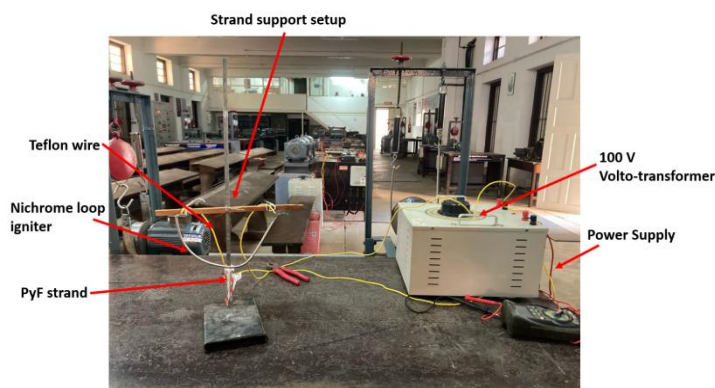


Fig.-2: Experimental Setup for Burn Rate Measurement of PyF Strands

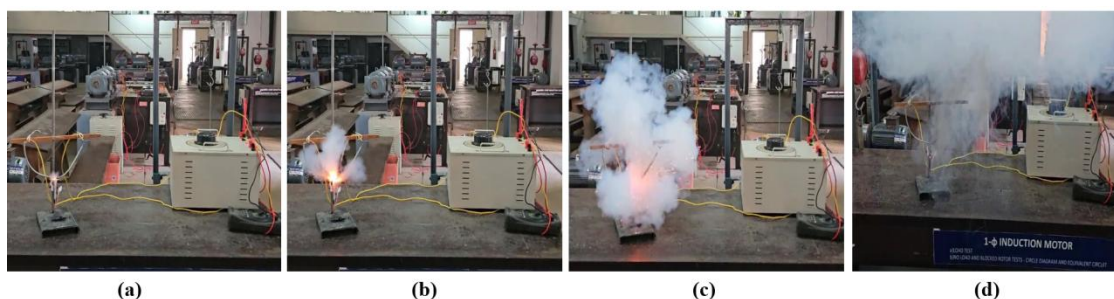


Fig.-3: Burn Rate Measurement Sequence: (a) Nichrome Loop Heating, (b) Ignition, (c) Peak Combustion, (d) Burnout

Compressive Strength Testing

The compressive characteristics of the prepared pyrotechnic formulations were evaluated using a Universal Testing Machine (Model: UTE 60, Manufacturer: Fuel Instruments and Engineers Pvt. Ltd.) equipped with a compression testing attachment. For this mechanical characterization, cubic specimens with dimensions of 50 mm × 50 mm × 50 mm were tested under compression using the Universal Testing Machine. The tests were conducted at a constant compression rate of 1.3 mm/min at ambient temperature. A maximum load of 2000 kg was applied, along with a travel limit of 3 mm. For each pyrotechnic formulation (PyF), three samples were tested to ensure accuracy and reproducibility. The compression modulus was calculated based on the initial linear portion of the stress–strain data obtained during the tests.

Combustion Efficiency and Fire Suppression

To study spontaneous combustion behavior, a combustion efficiency test was conducted using 1 g samples. The residue was weighed after ignition, and burn characteristics were recorded using a digital video recorder. The efficacy of each formulation in fire suppression scenarios, along with corresponding combustion flame temperatures, was assessed following the methodological framework developed by Pathak *et al.*²⁰ Figure-4 presents a schematic overview of the experimental configuration. The testing environment consisted of a stainless steel chamber measuring approximately 483 mm × 500 mm × 485 mm. Within this enclosure, a stainless steel fuel pan (96 mm diameter, 49 mm depth) generated a

standardized n-heptane pool fire with a nominal heat release rate of 4.57 kW. The pan contained a layered solution with 185 mL of water as the bottom phase and 92.5 mL of n-heptane as the upper phase. A cuboidal PyF sample (measuring 25 mm × 25 mm × 6 mm) housed in a designated case was positioned 250 mm away from the fire source. Ignition of the PyF was carried out using a nichrome wire loop, which was heated to incandescence by supplying 100 V alternate current (AC). Figure-5 shows the positioning of the pool fire source along with the thermocouple. A K-type thermocouple, having a 6 mm diameter and capable of measuring temperatures in the range of 0 to 1200 °C, was placed in proximity to both the fire source and the pyrotechnic sample. A cold discharge test was also conducted to measure flame temperature using a K-type thermocouple (6 mm diameter, 0–1200 °C range) in direct contact with the burning PyF sample. To test fire self-extinguishing behavior, a pool fire was ignited without intervention in a closed chamber. Observations showed sustained combustion for over 10 minutes, confirming the fire did not self-extinguish. During the fire suppression experiment, the fuel was first ignited and allowed to burn under open-door conditions for 60 seconds to ensure the development of a stable flame. After this pre-burn phase, the chamber door was shut, and the fire was allowed to burn for an additional 70 seconds. At this point, the PyF was triggered to release the aerosol for fire extinguishment. After each suppression event, the fuel pan was refilled, and the procedure was repeated. Three trials were conducted for each pyrotechnic formulation to ensure repeatability. A digital video camera was used to record the entire process, enabling accurate determination of both the aerosol discharge duration and the time taken to extinguish the fire.

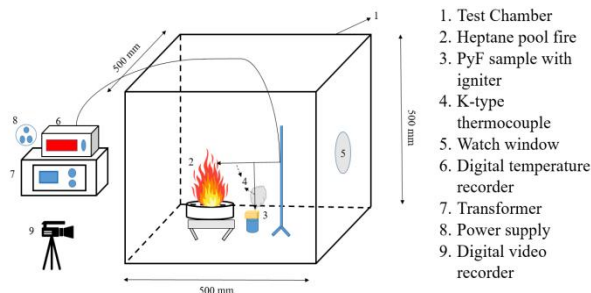


Fig.-4: Schematic Diagram of Fire Suppression Setup.

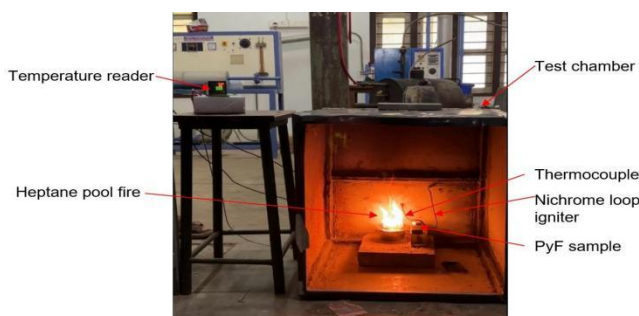


Fig.-5: Experimental Setup Showing Pool Fire and Thermocouple Placement.

RESULTS AND DISCUSSION

The selected binders exhibit significant differences in their chemical structures, which were considered to have a major influence on the variation in linear solid strand burn rate values. To analyze these differences in binder systems, oxygen balance (OB %) calculations were performed. For simplification, the polymer length of each selected binder was assumed to be $n = 1$. Oxygen balance (OB %) calculations were subsequently performed under the theoretical condition of complete combustion yielding exclusively carbon dioxide and water vapor as reaction products. These determinations employed equation-2, which incorporates several stoichiometric parameters: M represents the cumulative count of metal oxide atoms produced, while X , Y , and Z correspond to the total quantities of carbon, hydrogen, and oxygen atoms present in the molecular structure, respectively.

$$\text{OB \%} = (-1600 / M. \text{ Wt. of the compound}) [2X + (Y/2) + M - Z] \quad (2)$$

The computed OB% values were -118.4% for dextrin ($C_6H_{10}O_5$), -99.1% for carboxymethyl cellulose sodium salt ($C_8H_{11}NaO_7$, degree of substitution ≈ 1), -144.6% for hydroxyethyl cellulose ($C_{12}H_{21}O_8$) and -117.7% for xyloglucan ($C_{51}H_{86}O_{43}$). Xyloglucan and carboxymethyl cellulose therefore both carry comparatively favourable oxygen balances, while hydroxyethyl cellulose is the most oxygen-deficient of the four. Oxygen balance alone, however, does not fully account for the measured suppression performance: despite its favourable theoretical oxygen balance, the carboxymethyl cellulose-bound composite (PyF-2) failed to extinguish the pool fire, which points to a kinetic rather than thermodynamic limitation. As discussed below, rapid scission of the carboxylate ($-COO^-$) groups in carboxymethyl cellulose releases CO_2 early in the burn, cooling the reaction zone before sufficient aerosol has been generated - a mechanism that a static oxygen-balance calculation cannot capture. Xyloglucan's combination of a favourable oxygen balance and the absence of an easily-cleaved carboxylate group appears to explain its superior real-world performance more completely than oxygen balance alone. To assess combustion behaviour, which governs the safety of the gallic acid- KNO_3 - $KClO_3$ system under each binder, thermal analysis (TGA and DSC) was carried out on all five formulations; the profiles are presented in Fig.-6 to 10.

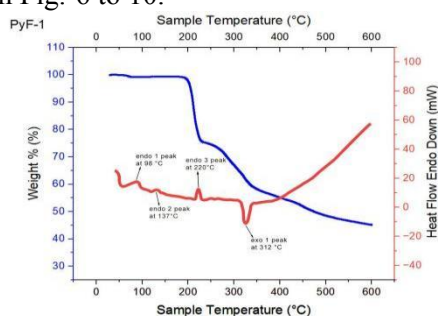


Fig.-6: TGA and DSC Profile of PyF-1

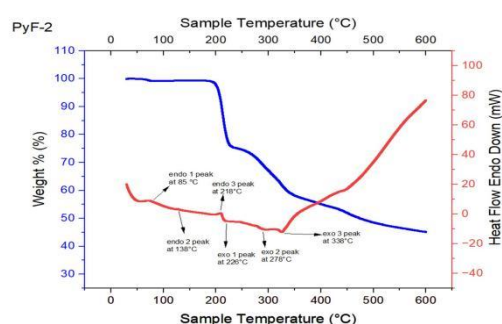


Fig.-7: TGA and DSC Profile of PyF-2

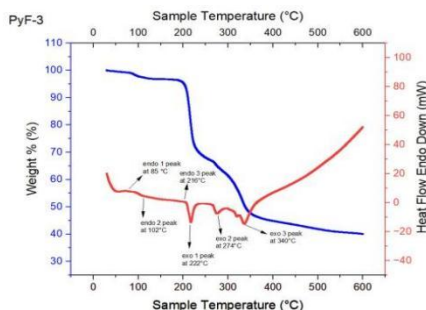


Fig.-8: TGA and DSC Profile of PyF-3

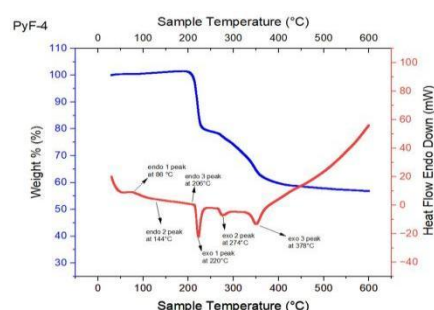


Fig.-9: TGA and DSC Profile of PyF-4

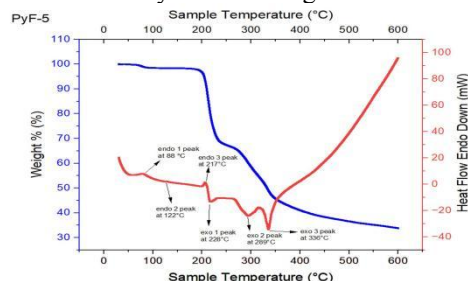


Fig.-10: TGA and DSC profile of PyF-5

The TG curves show a progressive three-stage mass loss between $50\text{ }^{\circ}\text{C}$ and $800\text{ }^{\circ}\text{C}$. The unbound ternary system already leaves an appreciable residue, and every binder addition increases it further, chiefly because the binder raises the carbon content of the charge. Ranked by residual mass at $500\text{ }^{\circ}\text{C}$ (Table-3), the xyloglucan composite leaves the least residue, followed in increasing order by dextrin, carboxymethyl cellulose and hydroxyethyl cellulose. A lower solid residue is desirable in practice: it indicates more

complete conversion of the charge into active aerosol and leaves less post-discharge deposit to be cleaned from the protected enclosure.

Thermal analysis of PyF-1 by differential scanning calorimetry (Fig.-6) produced three characteristic endothermic responses. An initial moisture-evaporation event was recorded at 98 °C, followed by the orthorhombic-to-trigonal crystalline transition of potassium nitrate at 137 °C. The third endotherm, at 220 °C, corresponds to decarboxylation of gallic acid with release of pyrogallol and CO₂. The thermogram also displayed an exothermic signature at 312 °C, indicating redox activity between pyrogallol (carbon donor) and the oxidisers (KNO₃/KClO₃), yielding potassium bicarbonate, nitrite and chloride salts. Analysis of the differential scanning calorimetry (DSC) profile for formulation PyF-2 (Fig.-7) demonstrated three distinct endothermic events followed by a series of three exothermic transitions. The initial endothermic response, observed at approximately 90 °C, is attributable to the volatilization of physisorbed water molecules. A subsequent endothermic shift at 130 °C aligns with the crystalline polymorphic transition of potassium nitrate. The third endothermic manifestation, detected at 218 °C, coincides with saccharide ring fragmentation within the carboxymethyl cellulose (CMC) component, generating glucose-derived thermal products. Concurrently, this thermal regime facilitates the decarboxylation of gallic acid, yielding pyrogallol and carbon dioxide as principal decomposition products. The first exothermic manifestation, occurring at 226 °C, originated from the scission of residual glucosidic linkages within the carboxymethyl cellulose (CMC) structure. This thermal decomposition process yielded potassium bicarbonate (KHCO₃), carbon dioxide (CO₂), and potassium nitrite (KNO₂) as primary products. A subsequent exothermic event observed at 278 °C corresponded to two simultaneous phenomena: the thermal dissociation of KHCO₃ into potassium carbonate (K₂CO₃) and potassium chloride (KCl), alongside the oxidative breakdown of pyrogallol derivatives and glucose fragments mediated by potassium chlorate (KClO₃). The final exothermic transition at 338 °C indicated the formation of potassium oxide (K₂O) via the comprehensive thermal decomposition of K₂CO₃, KNO₂, and remaining carbonaceous material. Thermal analysis of formulation PyF-3 via differential scanning calorimetry (Fig.-8) demonstrated a characteristic profile consisting of three endothermic transitions succeeded by three exothermic decomposition events. The initial endothermic response at 85 °C corresponds to the volatilization of physisorbed water and low-molecular-weight volatile constituents. A subsequent endothermic shift at 120 °C indicates the glass transition and initial thermal mobilization of the dextrin polymer framework. The third endothermic manifestation at 216 °C reflects substantial saccharide decomposition within the dextrin matrix, generating carbon-rich fragments alongside the decarboxylation of gallic acid to form pyrogallol and carbon dioxide (CO₂). The first exothermic event at 222 °C arises from redox chemistry between carbonaceous decomposition products and oxidizing components (KNO₃ and KClO₃) yielding chemical intermediates including potassium bicarbonate (KHCO₃), carbon dioxide, and potassium nitrite (KNO₂). A subsequent exothermic peak at 274 °C marks the thermal dissociation of into potassium carbonate (K₂CO₃) and potassium chloride (KCl), concurrent with oxidative consumption of residual organic material. The terminal exothermic transition at 352 °C signifies the formation of potassium oxide (K₂O) through complete decomposition of carbonate, nitrite, and carbonaceous residues. Differential scanning calorimetry of PyF-4 (Fig.-9) demonstrated three sequential endothermic peaks preceding three exothermic transformations. The initial endothermic response at 86 °C indicated the liberation of volatile species such as CO, CO₂, and H₂O. A subsequent endothermic transition at 144 °C corresponded to the crystalline phase change of KNO₃. The third endothermic event, observed at 206 °C, was attributed to the cleavage of glycosidic bonds in hydroxyethyl cellulose (HEC), resulting in the formation of low-molecular-weight carbonyl compounds, along with the decarboxylation of gallic acid, which produced pyrogallol and CO₂. The first exothermic process occurred at 220 °C and was driven by redox interactions between KNO₃, acting as the oxidant, and the reductants pyrogallol and carbonyl derivatives. This reaction pathway led to the formation of KHCO₃, CO₂ and KNO₂. At 274 °C, the second exothermic transformation involved the thermal decomposition of KHCO₃ into K₂CO₃ and KCl, accompanied by the oxidative pyrolysis of remaining organic constituents in the presence of KClO₃. Finally, the third exothermic peak, appearing at 378 °C, indicated the formation of K₂O through the complete thermal degradation of K₂CO₃, KNO₂, and residual carbon. The differential scanning calorimetry thermogram for formulation PyF-5 (Fig.-10) exhibited a

thermal profile characterized by three endothermic events preceding three exothermic decomposition processes. An initial endothermic response at 88 °C indicates the liberation of hygroscopically bound water. A subsequent endothermic transition at 152 °C reflects the solid-state polymorphic conversion of potassium nitrate from its orthorhombic to trigonal crystalline configuration. The third endothermic phenomenon at 217 °C signifies simultaneous depolymerization of xyloglucan through glycosidic bond scission and gallic acid decarboxylation, yielding pyrogallol and carbon dioxide (CO₂) as primary volatile products. The initial exothermic manifestation at 228 °C likely originates from concerted pyrolytic degradation of thermally modified xyloglucan and oxidation-reduction chemistry between phenolic compounds (pyrogallol) and carbohydrate-derived carbonyl species with the KNO₃-KClO₃ oxidizer system. These reactions generate potassium bicarbonate (KHCO₃), carbon dioxide, and potassium nitrite (KNO₂) as principal reaction intermediates. At 289 °C, the second exothermic process occurred, characterized by the thermal decomposition of KHCO₃ into K₂CO₃ and KCl, simultaneous with the KClO₃-mediated oxidative pyrolysis of dehydroxylated pyrogallol and xyloglucan derivatives. The reaction sequence culminated at 336 °C with formation through concerted pyrolytic breakdown of K₂CO₃, KNO₃, and residual carbonaceous materials.

Thermal analysis revealed that incorporating a binder system into the gallic acid-KNO₃-KClO₃ ternary pyrotechnic formulation (PyF-1) led to a reduction in ignition temperature by approximately 85 to 89 °C, as shown in Table-3. The observed reduction likely results from exothermic redox reactions occurring between the binder and potent oxidizers (KNO₃ and KClO₃). Among all binder-containing compositions, DSC results indicate that PyF-4 exhibits the highest thermal stability, remaining stable up to 220 °C.

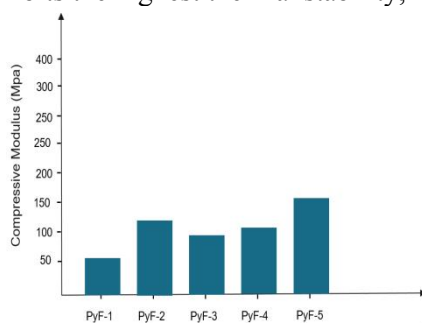


Fig.-11: Compressive Modulus (MPa) of PyF-1 to PyF-5 Formulations

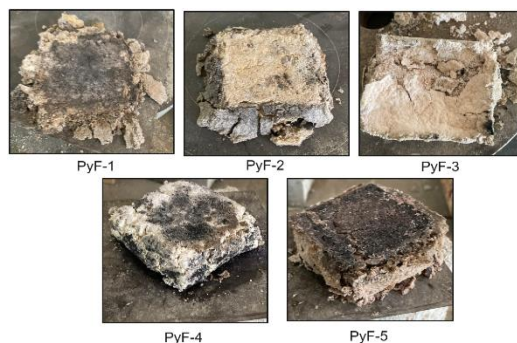


Fig.-12: Compressed PyF Samples after UTM Testing (PyF-1 to PyF-5)

All five formulations were compression-tested to establish their mechanical characteristics, and the mean compressive moduli are plotted in Fig.-11. Adding a binder to the unbound gallic acid-KNO₃-KClO₃ charge (PyF-1, 55 ± 2.1 MPa) raises compressive stiffness substantially in every case. PyF-5, containing xyloglucan, was the most rigid at 184 ± 4.2 MPa, whereas the dextrin-bound PyF-3 recorded 95 ± 3.2 MPa. PyF-2 (CMC) followed at 140 ± 3.8 MPa and PyF-4 (HEC) at 135 ± 4.1 MPa, both of which similarly exceeded PyF-3. The 31% margin of xyloglucan over the CMC benchmark is the single largest performance gap observed in this study and is attributed to the branched xyloglucan backbone, whose dense hydroxyl population forms an extensive hydrogen-bonded network bridging the oxidiser and fuel

particles. Figure-12 shows that all specimens deform progressively rather than failing abruptly under increasing stress, indicating internal structural rearrangement rather than brittle fracture - a favourable characteristic for grains that must tolerate vibration and impact in service. Fire-suppression performance was evaluated in n-heptane pool-fire tests and the results are collected in Table-3. Video analysis showed that the discharged aerosol engaged the fully developed fire and reduced it to intermittent flickers before final extinction in every case except PyF-2. The unbound control (PyF-1) extinguished the fire fastest, in 22 s, but it is not a practical candidate because it lacks the mechanical integrity required of a storable grain. Among the binder-containing compositions, the xyloglucan-based PyF-5 was clearly the most effective, extinguishing the flame in 35 s, against 43 s for the dextrin-bound PyF-3 and 52 s for the HEC-bound PyF-4; the CMC-bound PyF-2 failed to extinguish the fire at all within the test period. PyF-2 also recorded the lowest combustion flame temperature, 136 °C, and the two observations are related: rapid scission of its carboxylate ($-\text{COO}^-$) groups liberates a large volume of CO_2 early in the burn, which cools the reaction zone and suppresses the composite's own combustion before a sufficient mass of active aerosol has been generated. The superior response of PyF-5 follows from its higher burn rate, which produces a swift, directed flow of aerosol particles towards the flame front; the accelerated combustion promotes efficient contact between the discharged aerosol and the fire, giving faster heat absorption and chemical quenching. Placed against the existing literature, these results extend rather than merely repeat earlier findings. Pathak et al. established that a natural binder (tannic acid) could function in this matrix²⁰, and subsequent work confirmed the same for guar gum²², maltose³¹ and inulin³²; in each case, however, the natural binder was benchmarked against only one or two synthetic controls and mechanical data were either absent or reported separately. The present study contributes three things that were previously unavailable. First, it provides the first performance record of any kind for xyloglucan in an aerosol-forming composite, including its oxygen balance, ignition temperature and extinction time. Second, it demonstrates that a bio-based binder can simultaneously outperform both synthetic controls on mechanical strength (184 MPa versus 140 and 135 MPa) and on extinction time (35 s versus 52 s and a failure to extinguish), which removes the performance objection most often raised against natural binders. Third, it identifies oxygen balance as a simple, calculable screening parameter that correctly ranked all four binders by burn rate before any test was performed, offering formulators a way to narrow a candidate list without experimental effort. These outcomes carry a direct sustainability dimension and substantiate the alignment of this work with SDG-12: Responsible Consumption and Production.³⁵ Carboxymethyl cellulose and hydroxyethyl cellulose are manufactured by etherifying cellulose with chloroacetic acid and ethylene oxide respectively, both hazardous reagents that generate saline and organic effluent; their combustion products include volatile organic compounds. Xyloglucan requires neither reagent, being extracted from tamarind kernel powder - a residue of the tamarind pulp industry that is presently underused - and its lower residual mass at 500 °C (36.51%) points to more complete conversion and less post-discharge solid waste. The substitution therefore addresses Target 12.4 (sound management of chemicals and reduced release to air, water and soil) at the manufacturing stage and Target 12.5 (waste reduction through reuse) at the feedstock stage, while delivering superior, not merely equivalent, fire-suppression performance.

CONCLUSION

Four binder systems - two synthetic (carboxymethyl cellulose sodium salt and hydroxyethyl cellulose) and two natural (dextrin and xyloglucan) - were evaluated at 3 wt.% in a gallic acid - KNO_3 - KClO_3 aerosol-forming pyrotechnic composite against an unbound control, using a single set of combustion, mechanical and thermal test protocols. Xyloglucan gave the best overall performance: a burn rate of 5.72 mm s⁻¹, a burn time of 11 s, a combustion flame temperature of 418 °C, a compressive modulus of 184 Mpa, and extinction of a 4.57 kW n-heptane pool fire in 35 s. Its residual mass at 500 °C was 36.51%, the lowest of the binder-containing set, and its ignition temperature of 217 °C balances handling safety against responsiveness. A favourable oxygen balance of -117.70% contributes to this behaviour, although suppression performance tracked extinction time and mechanical strength more closely than oxygen balance alone. By comparison, the CMC-bound formulation failed to extinguish the pool fire, and the HEC-bound formulation required 52 s; both synthetic binders depend on energy-intensive etherification with chloroacetic acid or ethylene oxide and are associated with volatile organic emissions on combustion.

Dextrin performed creditably as a biodegradable option but was consistently behind xyloglucan on thermal response, mechanical strength and suppression efficiency. The principal conclusion is therefore that an unmodified, agro-residue-derived polysaccharide can replace a synthetic cellulose ether in a condensed aerosol fire-extinguishing composite with a net gain, rather than a loss, in performance. On this basis the following recommendations are made: (i) xyloglucan should be adopted as the reference bio-based binder for gallic acid-based aerosol-forming composites in further development work; (ii) the binder loading should be optimised across a range around 3 wt.%, since the present study fixed it at a single level; (iii) long-term storage, humidity-ageing and vibration testing should be undertaken before device-scale deployment; and (iv) the aerosol discharged by PyF-5 should be characterised by SEM, XRD and FTIR to confirm the identity and particle-size distribution of the active potassium species. Future work may also explore functionalised xyloglucan derivatives, such as oxygenated and thiolated forms, to tune thermal stability and oxidiser interaction further. In replacing a petrochemical input with a tamarind-processing residue while improving fire-suppression performance, this work contributes directly to SDG-12: Responsible Consumption and Production (Targets 12.4 and 12.5), and supports the wider transition towards greener energetic materials.

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CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

The present work is related to *SDG-12: Responsible Consumption and Production*. All the authors contributed significantly to this manuscript, participated in reviewing/editing and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

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