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Cellulose-fiber-based insulation materials with improved reaction-to-fire properties

Chao Zheng, Dongfang Li, and Monica Ek

KEYWORDS: Cellulose, Reaction-to-fire properties, Thermal insulation

ABSTRACT: The poor reaction-to-fire properties of cellulosic thermal insulation need to be improved to meet the safety regulations for building materials. In this study, cellulose-fiber-based insulation foams were prepared from formulations containing mechanical pulp and commercial fire retardants. Results of single-flame source tests showed that foams developed from the formulations with 20% expandable graphite (EG) or 25% synergetic (SY) fire retardants had substantially improved reaction-to-fire properties, and passed fire class E according to EN 13501-1. The results indicated that the foams could resist a small flame attack without serious flame spreading over a short period of time. Compared with the reference foam that contained no fire retardant, the peak heat release rate of the 20% EG and 25% SY foams decreased by 62% and 39% respectively when the samples were subjected to a radiance heat flux of 25 kW m^{-2} in a cone calorimeter, which suggested enhanced reaction-to-fire properties of these foams.

ADDRESSES OF THE AUTHORS: Chao Zheng (chaozh@kth.se), **Dongfang Li** (dongfan@kth.se), **Monica Ek** (monicaek@kth.se), Department of Fiber and Polymer Technology, KTH Royal Institute of Technology, Teknikringen 56-58, SE-100 44 Stockholm, Sweden
Corresponding author: Monica Ek

Introduction

Thermal insulation materials made from cellulose-based fibers have advantages over other types of materials from a sustainability point of view (Bribián et al 2011). To obtain a good thermal insulation property, cellulosic thermal insulation materials shall have a low density to ensure a low thermal conductivity (Xu et al 2004). However, as building components, the poor reaction-to-fire properties of cellulosic thermal insulation with low density have to be improved to ensure fire safety. Halogen and intumescent fire retardants are commercially used to improve the reaction-to-fire properties of thermal insulation. Compared with the former, the latter is more eco-friendly and safer, since no toxic gasses are produced during its combustion (Modesti, Lorenzett 2002 ;Ghanadpour et al 2015; Koklukaya et al 2015). The intumescent fire retardants have been widely utilized in thermal insulation foams developed from fossil resources (Modesti et al 2002), but such applications in cellulosic foams have rarely been reported.

In the present study, the commercial intumescent fire retardants such as an expandable graphite (EG) fire retardant and a synergetic (SY) fire retardant have here been used to improve the reaction-to-fire properties of cellulosic thermal insulation foams. In principle, EG can expand and form a “worm”-like char layer on the surface of the cellulose fiber upon heating. The char layer can suppress the flame and hinder heat and mass transfer from the material to the heat source, and thereby limit further thermal decomposition of the thermal insulation material (Camino et al 2001). The expansion of EG is initiated by a redox reaction between H_2SO_4 and the graphite that leads to the blowing:

$$C + H_2SO_4 \rightarrow CO_2 + 2H_2O + 2SO_2 \qquad [1]$$

The blowing effect can lead to an increase in the volume of the thermal insulation material by approximately 100 times when the temperature is above 200°C.

The SY fire retardants are made up of 50% ammonium sulfate, 10% ammonium polyphosphate, and 40% aluminum hydroxide. Ammonium sulfate decomposes at 284°C to generate ammonia (NH_3) that can dilute the combustible gasses in the fire, and sulfuric acid (H_2SO_4) that can catalyze the dehydration of cellulose to facilitate char formation. As a thermal barrier, the char can reduce the heat transfer from the external to the internal material as well as the speed of decomposition (Statheropoulos, Kyriakou 2000). Ammonium polyphosphate (APP) can also decompose to generate ammonia and polyphosphate that plays the same role as sulfuric acid. In addition, polyphosphoric acid and phosphoric acid produced by APP can improve the char amount by cross-linking carbonaceous structures from the decomposition of cellulosic fibers (Seefeldt et al 2012). Aluminum hydroxide decomposes into alumina and water vapor at 180°C, absorbing a large amount of heat. Water vapor can also dilute the combustible gasses. Moreover, the alumina or aluminum oxide (Al_2O_3) can form a protective layer on the surface and this obstructs the further reaction of the material with oxygen (Redwan et al 2015).

To obtain a CE-mark and be allowed to trade within the EU market, thermal insulation with low density needs to achieve fire class E (Östman, Mikkola 2006), as stated by the Construction Products Regulation (CPR), and to be certified in this case according to European Assessment Document (EAD). Different performance classes (A to F) for walls, roofing products, and floor-coverings are classified according to the standard test method (CEN ISO, 2010) “The Single Flame Source Test EN ISO 11925-2: Reaction to fire tests for building products-Part 2: Ignitability when subjected to direct impingement of flame”. If the flame tip does not reach 150 mm above the flame application point during 20 s when a single flame is applied on the sample, the tested samples can be classified as E. Fire class E materials are able to resist a small flame attack without substantial flame spreading for a short period.

To evaluate the ignition behavior, the fire performance, and simulate or predict full-scale fire behavior of the cellulosic thermal insulation materials that pass class E, the cone calorimeter can be used to determine fire response parameters, such as time to ignition (TTI), heat release rate (HRR), total heat release (THR), smoke production, mass loss, and CO production. A longer time to ignition and a lower heat release rate indicate a better reaction-to-fire properties (Schartel, Hull 2007).

In the present study, two types of commercial intumescent fire retardants were added at different weight ratios to the formulations to prepare cellulosic thermal insulation foams by the wet foaming technique. The fire performances of the foams were tested by the single-flame source test and cone calorimeter fire test.

Materials and Methods

Materials

Bleached chemithermomechanical pulp (CTMP) with an average fiber length of 2.5 mm and width of 40 µm was provided by Rottneros Company (Söderhamn, Sweden). Sodium dodecyl sulfate (≥99.0%) was purchased from Sigma-Aldrich (Stockholm, Sweden). Commercial fire retardant-expandable graphite with a particle size of 250 µm was provided by GrafTech (USA), and a commercial synergetic fire retardant with a composition of 50% ammonium sulfate, 10% ammonium polyphosphate, and 40% aluminum hydroxide was supplied by a local company.

Preparation of insulation materials with fire retardants

Foam-forming technique was used to produce the cellulose-fiber thermal insulation materials (Al-Qararah et al 2012; Lehmonen et al 2013; Jahangiri et al 2014; Lappalainen et al 2014; Mira et al 2014; Jahangiri et al 2016; Koponen et al 2016). 0.3 g of sodium dodecyl sulfate surfactant and 5%, 10%, 15%, 20%, 25%, or 30% of either expandable graphite or the synergetic intumescent fire retardant were mixed with 4.5% bleached mechanical pulp suspension. After mechanical stirring (3000 rpm) for 15 minutes in an L&W Pulp Disintegrator (ABB, Zürich, Switzerland) to form foams, the mats were dried at 90°C for 8h in a TS8000 oven (TERMAKS, Bergen, Norway). A pulp suspension without fire retardant was added to produce a reference insulation material.

Test Methods

Retention of fire retardants: The retention of fire retardants was determined as:

$$Retention = \frac{m_2 - m_1}{m_1} \times 100\% \qquad [2]$$

where m_1 and m_2 are respectively the dry weights of the pulp fibers and of the insulation foam with fire retardants.

Single-flame source test*:* A single-flame was applied to the surface or edge of the conditioned (23°C, 50% RH) samples (250 mm long by 90 mm wide), e.g., surface exposure was conducted when the flame was applied on the centerline of the sample surface, 40 mm above the bottom edge of the test sample; edge exposure was carried out when the flame was applied to the center of the width of the bottom edge of the test sample, following the method described in standard EN ISO 11925-2 (CEN ISO, 2010), and the position reached by the flame after the flame application time 15 s and duration time 20 s was recorded. The fire classification was evaluated according to the standard EN 13501-1 (CEN, 2007) (if the flame tip does not reach the 150 mm limit then the material is classified as class E).

Thermogravimetric analysis (TGA): TGA was used to study the thermal behavior of the samples that have achieved fire class E, using a TGA/SDTA 851e (Mettler Toledo, Greifensee, Zürich, Switzerland). The samples were heated from 25 to 800°C at a rate of 10°C min^{-1} in a 50-mL min^{-1} flow of nitrogen. Two repetitions of every sample were carried out to evaluate the onset decomposition temperature (the temperature at which 10% of the weight loss appears), the maximum decomposition temperature (the peak temperature at which maximum decomposition rate occurs), and the residue at 800°C.

Cone-Calorimeter fire test*:* A Cone Calorimeter (Fire Testing Technology Ltd. UK) was used to study the combustion behavior. The samples (100 mm × 100 mm × 15 mm) were subjected to a 25 kW m^{-2} of radiant heat flux following the procedure described in the standard ISO 5660-1 (ISO, 2015). The time to ignition (TTI, s), peak heat release rate (peak-HRR, kW m^{-2}), and total heat release (THR, MJ m^{-2}) were assessed. Two repetitions of each sample were performed.

Scanning electron microscopy (SEM): The surface morphology of insulation foams before and after cone calorimeter test was observed in a Scanning electron microscope (Hitachi S-4800 FE-SEM, JEOL Ltd., Japan) after gold/palladium sputtering.

Thermal conductivity:The thermal conductivity of the fire-retardant insulation foams was determined by the Hot Disk TPS 2500S instrument (Hotdisk AB, Gothenburg, Sweden), with a 20 mW output power and a 40 s measurement time in the isotropic mode. A 6.4-mm Ni wire sensor and a bulk mode were used for the thermal conductivity determination (Hot Disk Thermal Analyser 7.2). Three measurements of each sample were performed at 25°C.

Results and Discussion

Retention of Fire Retardants

In order to study the potential leaching of fire retardants during the dewatering process, the actual amounts of fire retardants retained in the final materials were calculated. The results are summarized in *Table 1*. Because of the insolubility in water of expandable graphite (EG), there was no difference between the retention of expandable graphite (EG) and the nominal fire retardant amount in the formulations, but there was a loss of the synergetic (SY) intumescent fire retardant, which could be due to the leaching of water-soluble ammonium sulfate (*Table 1*). The synergetic (SY) fire retardant retained in the foams may consist predominantly of 20% ammonium polyphosphate and 80% aluminum hydroxide.

Table 1– Retention of fire retardants of different percentages of fire retardants in the formulations.

Sample	Content of fire retardant in the formulation (%)	Retention/actual content of fire retardant in the foam sample (%)
5% SY	5	1
10% SY	10	1
15% SY	15	5
20% SY	20	6
25% SY	25	17
30% SY	30	19
5% EG	5	5
10% EG	10	10
15% EG	15	15
20% EG	20	20
25% EG	25	25
30% EG	30	30

Ignitability

Cellulosic foams with more than 20% of expandable graphite in the formulations reached class E as illustrated in *Fig 1*. For the foams with 15% and 20% SY in the formulations, they fulfilled the requirements of E class on one surface (*Fig 2*), but there were large burned areas in the edge exposure test (*Fig 3*), which could relate to the limited retention (5–6%) of the fire retardants (*Table 1*). The 20% EG, 25% EG, and 30% EG samples performed better than the 20% SY, 25% SY, and 30% SY samples, as shown in *Fig 4*. No obvious improvement in the reaction-to-fire properties could be observed when the content of fire retardant in the formulation was increased from 20 to 30%, which is probably due to an uneven distribution of fire retardant in the foams. Neither 10% EG nor 10% SY could achieve class E, indicating that this amount of fire retardant in the formulations was not enough to reduce the ignitability of the foams. Based on the results of the single-flame source test, samples 20% EG and 25% SY were subjected to testing in the cone calorimeter.

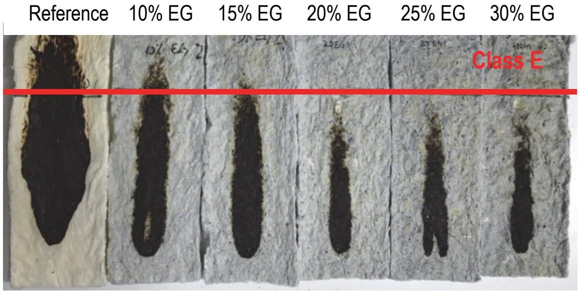

Fig 1 - Single-flame source (surface) test result for reference foam without fire retardant, and foams with 10–30% expandable graphite (EG) fire retardant in the formulation.

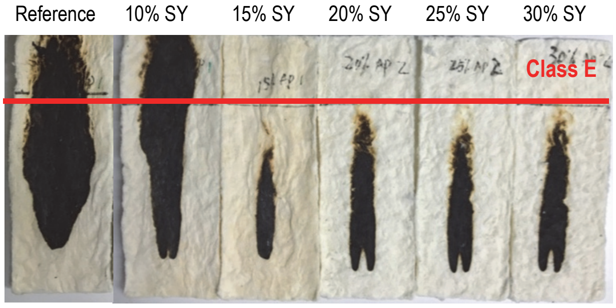

Fig 2 - Single-flame source (surface) test result for reference foam without fire retardant, and foams with 10–30% synergetic (SY) fire retardant in the formulation.

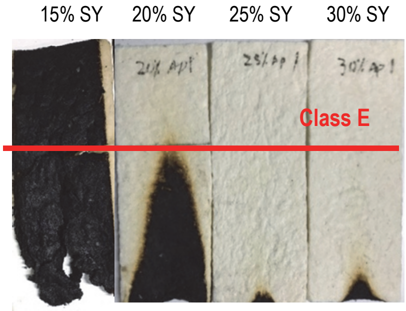

Fig 3 - Single-flame source (edge) test result for foams with 15–30% synergetic (SY) fire retardant in the formulation.

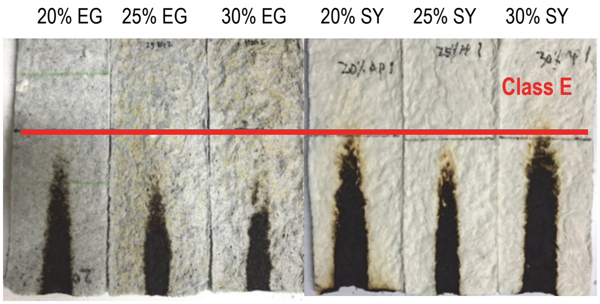

Fig 4 - Single-flame source (edge) test result for foams with 20–30% expandable graphite (EG) or synergetic (SY) fire retardant in the formulation.

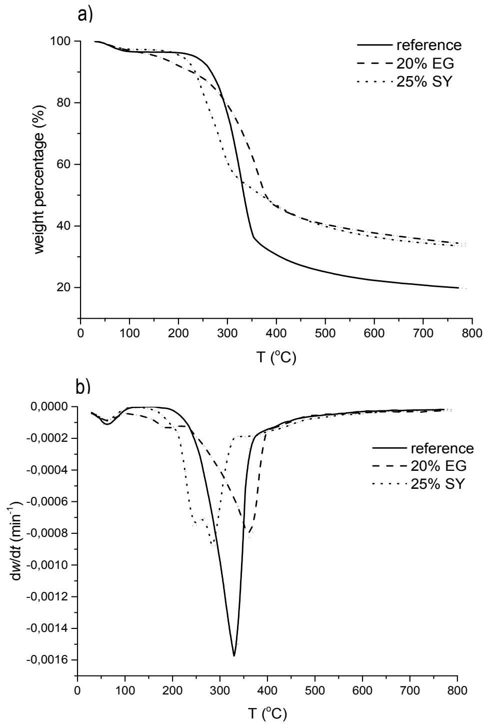

Fig 5 - a) Thermogravimetric (TG) and b) Derivative curves (DTG) for the reference and fire-retardant containing insulation foams.

Table 2 - Thermogravimetric analysis results of the reference without fire retardant, 20% EG, and 25% SY.

	Reference	**20% EG**	**25% SY**
$T_{onset\ 10\%}$ (°C)	269±1	226±4	235±0
T_{max1} (°C)	331±1	193±1	245±2
T_{max2} (°C)	-	363±1	285±2
Residue (%)	20±0	33±2	33±1

Table 3 - Cone calorimeter test results of the reference without fire retardant, 20% EG, and 25% SY (incident heat flux 25 kW m^{-2}).

	Reference	**20% EG**	**25% SY**
Time to ignition(TTI) (s)	9.0±0.0	6.0±0.0	9.5±0.7
Peak-HRR (kW m^{-2})	116.5±6.4	44.0±1.4	71.5±3.5
Time to Peak-HRR (s)	15.0±0.0	10.0±0.0	15.0±0.0
Total heat release (THR) (MJ m^{-2})	9.9±0.3	4.4±0.1	6.1±0.0

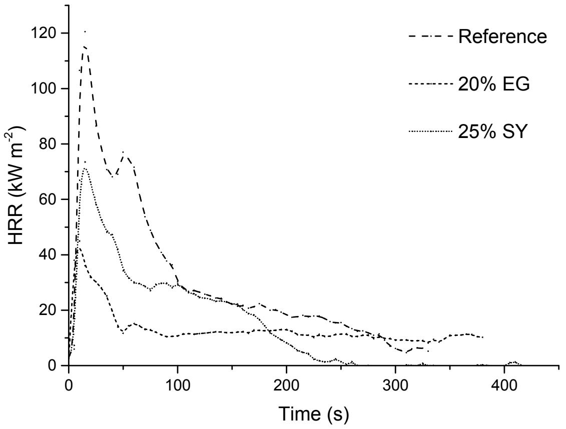

Fig 6 - Heat release rate curves of the reference without fire retardant, 20% EG, and 25% SY from the cone calorimeter fire test (incident heat flux 25 kW m^{-2}).

Thermal Stability

The thermogravimetric analysis curves are shown in *Fig 5* and the numerical results are presented in *Table 2*.

Compared with the reference foam without fire retardant, 20% EG and 25% SY had lower decomposition temperature for 10% weight loss ($T_{onset\ 10\%}$) and a lower temperature at the maximum decomposition rate (T_{max1}). The lower $T_{onset\ 10\%}$ and T_{max} values indicate a lower thermal stability. A decrease in thermal stability favors early thermal decomposition of the foams and this facilitates the formation of a char layer on the foam surface at a lower temperature. The increase in residue or char (13%) can serve as a thermal barrier to protect the material from the external heat and hinder further combustion.

T_{max1} for the 20% EG sample was much lower than that for the 25% SY, but the temperature at the maximum weight loss rate (T_{max2}) for 20% EG was obviously higher than that for 25% SY. The higher T_{max2} for 20% EG may be related to the formation of a thermally stable char due to graphite expansion at T_{max1} (Meng et al 2009). Two peaks were observed in the DTG curve of 25% SY. The first peak could result from the onset of decomposition of aluminum hydroxide at approximately 250°C giving Al_2O_3 and water (Arao et al 2014). The second peak could result from the loss of APP at 300°C due to the release of NH_3 and H_2O (Riva et al 2003).

Heat Release

The heat release rate curves are shown in *Fig 6* and the numerical data are presented in *Table 3*.

The peak heat release rate (Peak-HRR) and average HRR are crucial for explaining the rate of heat release during combustion. The HRR and the total heat release (THR) are also important in the evaluation of fire disaster. A lower heat release rate suggests better reaction-to-fire properties.

As shown in *Table 3*, compared to the reference without fire retardant, the Peak-HRR of the 20% EG and 25% SY decreased by 62% and 39% respectively, and the total heat release (THR) decreased by 56% and 38% respectively. The shorter time to ignition (TTI) of 20%

EG could be related to the black color of the foams with EG as the fire retardant, which could absorb more radiant heat from the cone heater, and thus show a faster increase in the surface temperature and an earlier initiation of thermal degradation of the cellulose (Wei et al 2013).

There are two peaks in the heat release rate for the reference in *Fig 6*. The first one may be due to the quick consumption of a large amount of oxygen after the ignition (Lindholm et al 2009), and the second peak arose when the surface char cracked (Schartel et al 2003). On the other hand, the peak heat release rate of both 20% EG and 25% SY was significantly lower, as a result of the heat absorption by the fire retardants. The expansion of EG and the formation of a char layer were initiated by a redox process between H_2SO_4 and the graphite layers (Fink 2013). Phosphoric acid produced from APP can also promote the formation of a char layer by an acid-catalyzed dehydration of cellulose (Meng et al 2009; Wu, et al 2009). The char layer formed can protect the insulation material from heat and mass transfer, and thus reduce the heat release rate (Recasens et al 2005).

As shown in *Fig 7*, only minor char residue was left at the end of the cone calorimeter test for the reference. However, char formation resulting from the combustion of 25% SY and 20% EG was obvious. A greater formation of char could provide protection against heat transfer and flame spreading (Wu et al 2009). The fact that the peak-HRR of 20% EG is lower than that of 25% SY is possibly related to the difference in char formation by the fire retardants. 25% SY broke into pieces during the combustion although it formed a more compact char than 20% EG, which could be the result of the further combustion when a crack developed on the surface.

Surface morphology

The surface morphology of the foams and the char morphology after cone calorimeter test are shown in *Fig 8*. The 25% SY and 20% EG samples showed a larger content of char than the reference. After combustion, 20% EG had a loose char layer due to the expansion of EG. The "worm"- like structure produced by graphite expansion can hinder the spreading of the flame, and the char layer can limit the heat and mass transfer, and hinder further decomposition of the insulation materials (Modesti et al 2002). Al_2O_3 particles from the decomposition of $Al(OH)_3$ were observed in the 25% SY residue.

Thermal Insulation

The thermal conductivities of the 20% EG and 25% SY samples were similar to that of the reference (*Table 4*). The slightly higher thermal conductivity of 25% SY may be due to the higher density as a result of the addition of fire retardant. According to Collet and Prétot (2014), higher density contributes to higher heat transfer through the solid component of the material, and this results in an increased conductivity. Furthermore, EG had no detrimental effect on the thermal conductivity of the cellulosic foams. This result is different from that the thermal conductivity of polyisocyanurate–polyurethane foams increased when EG was added (Modesti et al 2002).

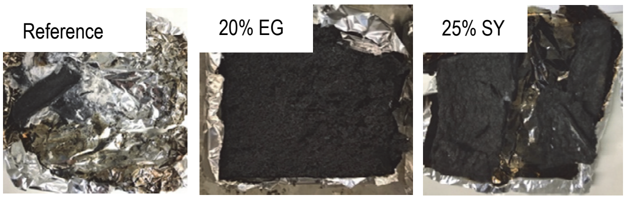

Fig 7 - Residues of the reference without fire retardant, 20% EG, and 25% SY at the end of the cone calorimeter test.

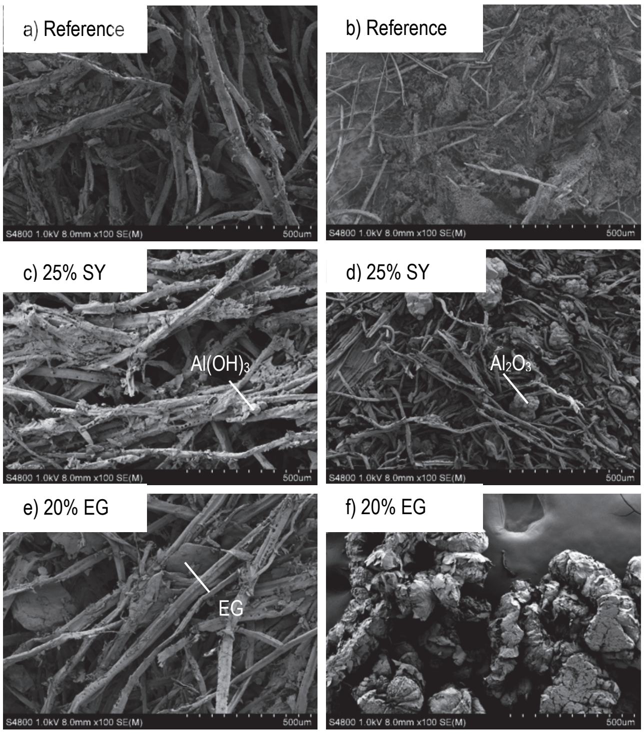

Fig 8 - SEM of the outer surfaces of the reference, 25% SY and 20% EG foams before and after the cone calorimeter test.

Table 4 - Thermal conductivity and density of the reference without fire retardant, 20% EG, and 25% SY.

	Thermal Conductivity (W m^{-1}K^{-1})	**Density (g cm^{-3})**
Reference	0.050±0.001	0.047±0.003
20% EG	0.049±0.001	0.042±0.003
25% SY	0.052±0.001	0.060±0.002

Conclusions

The reaction-to-fire properties of cellulosic thermal insulation foams with low density were substantially improved by adding either 20% expandable graphite or 25% synergetic fire retardant into the formulation. According to the single-flame source test, the performances of these foams could meet the requirements of European fire class E. The results of the cone calorimeter fire test suggested that the peak heat release rate of these foams was significantly lower than that of the reference foam without the fire retardant. The foam with 20% expandable graphite fire retardant in its formulation had better reaction-to-fire properties than the foam with 25% synergetic fire retardant, and there is no negative effect on the thermal conductivity of the cellulosic foams.

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