Flame Retardant Polypropylenes: A Review

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Abstract: Polypropylene (PP) is a commodity plastic known for high rigidity and crystallinity, which is suitable for a wide range of applications. However, high flammability of PP has always been noticed by users as a constraint; therefore, a variety of additives has been examined to make PP flame-retardant. In this work, research papers on the flame retardancy of PP have been comprehensively reviewed, classified in terms of flame retardancy, and evaluated based on the universal dimensionless criterion of Flame Retardancy Index (FRI). The classification of additives of well-known families, i.e., phosphorus-based, nitrogen-based, mineral, carbon-based, bio-based, and hybrid flame retardants composed of two or more additives, was reflected in FRI mirror calculated from cone calorimetry data, whatever heat flux and sample thickness in a given series of samples. PP composites were categorized in terms of flame retardancy performance as Poor, Good, or Excellent cases. It also attempted to correlate other criteria like UL-94 and limiting oxygen index (LOI) with FRI values, giving a broad view of flame retardancy performance of PP composites. The collected data and the conclusions presented in this survey should help researchers working in the field to select the best additives among possibilities for making the PP sufficiently flame-retardant for advanced applications.

Keywords: flame retardancy; polypropylene; Flame Retardancy Index (FRI); cone calorimetry; flame retardants

1. Introduction

Polymers are building blocks of advanced materials and systems, but their flammability has been a serious constraint in their usage in advanced applications [1–3]. Polypropylene (PP) is a commodity plastic widely used in a variety of applications, particularly in the form of composites in load-bearing uses due to its high rigidity and crystallinity [4]. By the end of 2020, the PP market size is expected to reach $112 billion, and it is estimated to reach $155 billion by 2026 [5,6]. Its global production was 56.0 million metric tons in 2018, and it is estimated to reach around 88.0 million metric tons by 2026. This growing demand reflects the importance of PP for applications where low density, hardness, high flexural modulus, and chemical resistance are needed [7,8]. Moreover, PP is a low-
cost plastic capable of being processed with various methods, e.g., extrusion, thermoforming, and injection molding [9,10]. Therefore, a huge number of PP products, including fibers, films, sheets, textiles, pipes, and profiles, have been developed and used in the automotive, electrical and electronic, packaging, and construction industries [11–14]. On the other hand, due to the inherent flammability, the use of flame-retardant additives in PP is necessary to minimize the risk of fire [15]. Different types of flame retardants have been used in PP including minerals, phosphorus-based, nitrogen-based, and intumescent [16–18]. It was recognized that additive selection plays a crucial role in achieving acceptable flame retardancy [19], where the type, the size, and the loading percentage of flame retardants control the fire behavior of PP matrix.

A diversity of additives are used in PP to make it flame retardant. There is a need for a comprehensive survey to classify PP composites in terms of flame retardancy. In the present paper, several families of flame retardants examined in PP have been identified and categorized to evaluate their flame retardancy performance in terms of Flame Retardancy Index (FRI) [19,20]. FRI is a universal dimensionless index that takes into account well-known parameters obtained from cone calorimeter test (peak of heat release rate (pHRR), the total heat release (THR), and the time to ignition (TTI)). FRI can be simply calculated using Equation (1):

\[
FRI = \frac{[THR \times \left( \frac{pHRR}{TTI} \right)_{\text{Neat Polymer}}]}{[THR \times \left( \frac{pHRR}{TTI} \right)_{\text{Composite}}]}
\]  

(1)

Basically, the use of FRI makes it possible to semi-qualitatively classify polymer composites by labeling them as Poor, Good, or Excellent flame retardancy performance and thus enables evaluation of the efficiency of the incorporated flame retardant (FR). There has always been a need for fast-tracking and classifying polymers for their flame retardant performance. The use of FRI made possible classifying polymers and polymer composites in terms of flame retardancy in a simple manner. For FRI values below \(10^0\) obtained by the use of Equation (1), we have the case (namely Poor) where the addition of FR adversely affects flame retardancy of polymer. When FRI takes values in the range of \(10^0-10^1\), we name it Good flame retardancy performance, such that addition of FR enhances the resistance of polymer against fire. For FRI values above \(10^1\), which is rare in practical cases, we have an Excellent case, where FR significantly improves flame retardancy. It is worth mentioning that some important parameters of testing such as irradiance and sample thickness as well as sample weight can be neglected due to the fact that, in the FRI formula, the parameters related to the neat polymer are divided by those of polymer/FR composite. Thus, the dimensionless value obtained can be used as a reliable measure of the efficiency of FR in polymer. In this survey, the data from the literature were extracted first, and five families of flame retardants that served as PP were considered including phosphorus-based, nitrogen-based, mineral, carbon-based, and bio-based flame retardants, and hybrid cases composed of the aforementioned five categories were distinguished. The main aim of the present survey is to give the readers a broad view of FR systems used in PP via FRI classification method. Certainly, this classification is not a precise and unique data set for FR selection for PP, but it can be considered as a database to compare different systems. The focus of this work was particularly placed on the reports in which cone calorimetry test was carried out. However, some other parameters such as smoke quantity or the percentage of FR elements (phosphorus, nitrogen, …) were not systematically given in this research paper due to the lack of data, which could lead to unreliable judgments. For some papers, limiting oxygen index (LOI) and UL-94 data were also available, which were used in finding possible correlations between the FRI variation and other criteria.

2. Phosphorus-Based Flame Retardants

Various types of phosphorus-based flame retardants have been incorporated into PP to make it flame-retardant [21–23]. Table 1 reviews the names and the percentages of these flame retardants incorporated into PP. Moreover, the values obtained from cone calorimetry such as the peak of heat release rate (pHRR), the total heat release (THR), and the time to ignition (TTI) are summarized in
this Table. The FRI value, calculated from cone calorimetry parameters, as well as the LOI and UL-94 values, are also presented in Table 1. In some cases, if LOI and/or UL-94 values were not available, the sign “—” was used.

Table 1. Flame-retardant PP materials containing phosphorus-based (P) flame retardants. Data are extracted from the literature: cone calorimetry parameters (TTI, pHRR, THR), LOI, and UL-94 values. The FRI values were calculated by authors of the present review. The name and the percentage of flame retardants are provided in separate columns. “wt.%” was used for loading level of additives, while “—” stands for the systems free of additive or the neat PP. * FR means flame retardant. Since all comparisons were made in terms of FRI, classification of polymers in terms of their flame retardant properties was not surveyed based on the chemistry of additives, heat flux, sample thickness, etc.

| PP Containing Phosphorus-Based (P) FR * | wt.% | TTI (s) | pHRR (kW·m⁻²) | THR (MJ·m⁻²) | Irradiance (kW·m⁻²) | Sample Thickness (mm) | FRI | LOI | UL-94 | Ref. |
|----------------------------------------|------|---------|---------------|--------------|---------------------|----------------------|-----|-----|-------|------|
| Ammonium polyphosphate (APP)           | 10   | 24      | 925           | 92           | 35                  | 0.4                  | 2.35| —   |       | [24] |
|                                        | 12   | 37      | 510           | 97           | 35                  | 3                   | 2.36| 22.3| V-2   | [25] |
| APP                                    | 15   | 27      | 339           | 89           | 35                  | 3                   | 2.82| 25.4| V-0   | [25] |
| APP                                    | 20   | 21      | 306           | 141.6        | 50                  | 4                   | 2.84| 27   | NR    | [26] |
| APP                                    | 20   | 40      | 787           | 92           | 35                  | 3.2                 | 1.66| 20.5| NR    | [28] |
| APP                                    | 20.3 | 193     | 254.8         | 54.5         | 50                  | 2.4                 | 6.37| V-0  |       | [28] |
| APP                                    | 20   | 31      | 633           | 44.2         | 35                  | 3                   | 1.81| 17   | NR    | [23] |
| APP                                    | 21   | 11      | 397           | 87           | 35                  | 3                   | 1.23| 20.6| NR    | [23] |
| APP                                    | 20   | 21      | 1242          | 111          | 50                  | 3.2                 | 1.64| 23.2| NR    | [30] |
| APP                                    | 25   | 398     | 147.5         | 50           | 3                   | 1.7                 | 2.19| V-2  |       | [29] |
| APP                                    | 25   | 38      | 1455          | 148          | 50                  | 3                   | 1.78| 19.6| NR    | [30] |
| APP                                    | 25   | 18      | 579           | 109          | 50                  | 3.2                 | 1.64| 23.2| NR    | [30] |
| APP                                    | 25   | 43      | 652           | 80           | 35                  | 3.2                 | 1.49| 21   | NR    | [31] |
| APP                                    | 20   | 809     | 96            | 50           | 3                   | 1.8                 | 17.6| NR   | [32] |
| APP                                    | 21   | 397     | 87            | 50           | 3                   | 1.23                | 20.6| NR   | [32] |
| APP                                    | 21   | 1242    | 111           | 50           | 3.2                 | 1.31                | 21.7| NR   | [33] |
| APP                                    | 25   | 398     | 147.5         | 50           | 3                   | 1.7                 | 2.19| V-2  |       | [34] |
| APP                                    | 25   | 33      | 390.8         | 196          | 50                  | 3.2                 | 2.92| 20.9| NR    | [34] |
| APP                                    | 25   | 25      | 841.6         | 89.1         | 50                  | 3                   | 1.81| 18   | NR    | [35] |
| APP                                    | 25   | 13      | 473.3         | 90.2         | 50                  | 3                   | 0.91| 20   | NR    | [35] |
| Piperazine-modified APP (m-APP)        | 25   | 17      | 162.6         | 84.5         | 50                  | 3                   | 3.71| 32.5| V-0   | [35] |
| APP                                    | 25   | 19      | 526           | 180          | 50                  | 6                   | 1.88| 19.6| NR    | [36] |
| Polyisoxane shell-coated APP (mc-APP)  | 25   | 19      | 214           | 137          | 50                  | 6                   | 6.08| 25   | NR    | [36] |
| Melamine and phytic acid-modified APP  | 25   | 33      | 218.1         | 80.6         | 35                  | 3                   | 3.12| 22.5| V-2   | [37] |
| (m-APP)                                | 25   | 37      | 1284          | 121          | 50                  | 3                   | —   | —   | —     | [38] |
| APP                                    | 30   | 22      | 767           | 111          | 50                  | 3                   | 1.08| 21.7| NR    | [38] |
| APP                                    | 48   | 988     | 88.3          | 35           | 3.2                 | 1.6                 | 22.2| NR   | [39] |
| APP                                    | 32   | 459     | 77.6          | 35           | 3                   | 1.63                | 22   | NR   | [39] |
| APP                                    | 50   | 1350    | 91.2          | 35           | 3                   | 1.7                 | 22.2| NR   | [40] |
| APP                                    | 58   | 851     | 74.4          | 35           | 3                   | 1.8                 | 22   | NR   | [40] |
| Melamine-formaldehyde-tris(2-hydroxyethyl) isocyanurate resin microencapsulated APP (mc-APP) | 30   | 24      | 375           | 116.4        | 50                  | 3                   | 2.55| 32   | V-0   | [41] |
| APP                                    | 30   | 30      | 432           | 114          | 35                  | 3                   | 1.81| 22   | NR    | [42] |
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|------------------------|
| 4, 4′-diphenylmethanediisocyanate and melamine microencapsulated APP (mc-APP) | 30 | 27 | 300 | 100 | 35 | 3 | 2.68 | 32.1 | V-0 | [42] |
| APP | — | 29 | 1186 | 215 | 50 | 6 | — | 17 | NR | [43] |
| Epoxy acrylate microencapsulated APP (mc-APP) | 30 | 13 | 332 | 149 | 50 | 6 | 2.31 | 24.8 | NR | [43] |
| APP | — | 40 | 1174.7 | 102.2 | 35 | 3 | — | 17 | NR | [44] |
| 4, 4′-diphenylmethane diisocyanate and melamine and pentaerythritol microencapsulated APP (mc-APP) | 30 | 30 | 301.8 | 65.1 | 35 | 3 | 4.58 | 25 | V-1 | [44] |
| APP | — | 68 | 577.5 | 82.7 | 35 | 3 | — | 18.2 | NR | [45] |
| Thermoplastic polyurethane microencapsulated APP (mc-APP) | 5 | 57 | 395.4 | 67.2 | 35 | 3 | 1.50 | 18.7 | NR | [45] |
| Thermoplastic polyurethane microencapsulated APP (mc-APP) | 10 | 42 | 282.5 | 63.7 | 35 | 3 | 1.63 | 19.6 | NR | [45] |
| Thermoplastic polyurethane microencapsulated APP (mc-APP) | 15 | 40 | 214.9 | 59.9 | 35 | 3 | 2.18 | 20 | NR | [45] |
| Thermoplastic polyurethane microencapsulated APP (mc-APP) | 20 | 32 | 193.6 | 57.3 | 35 | 3 | 2.02 | 20.3 | NR | [45] |
| Thermoplastic polyurethane microencapsulated APP (mc-APP) | 25 | 30 | 145.4 | 64.1 | 35 | 3 | 2.26 | 22.2 | NR | [45] |
| Thermoplastic polyurethane microencapsulated APP (mc-APP) | 30 | 31 | 140.6 | 41.8 | 35 | 3 | 3.70 | 22.9 | NR | [45] |
| APP | — | 25 | 841.6 | 89.1 | 50 | 3 | — | 18 | NR | [46] |
| Ethylenediamine-modified APP (m-APP) | 35 | 11 | 435.9 | 83.9 | 50 | 3 | 0.90 | 20.4 | NR | [46] |
| APP | — | 25 | 841.6 | 89.1 | 50 | 3 | — | 18 | NR | [47] |
| Ethanolamine-modified APP (m-APP) | 35 | 18 | 96.6 | 22.6 | 50 | 3 | 24.73 | 35 | V-0 | [47] |
| APP | — | 33 | 837 | 212 | 50 | 6 | — | 17 | NR | [48] |
| Pentaerythritol triacrylate microencapsulated APP (mc-APP) | 40 | 32 | 214 | 183 | 50 | 6 | 4.39 | 30.6 | V-0 | [48] |
| APP | — | 38 | 1284 | 214 | 50 | 6 | — | 18.2 | NR | [49] |
| APP | 25 | 34 | 537 | 177 | 50 | 6 | 2.58 | 20.9 | NR | [49] |
| Phosphorus-based charring agent: 3,9-Bis-(1-oxo-2,6,7-trioxo-1-phospha-bicyclo[2.2.2]oct-4-ylmethoxy)-2,4,8,10-tetraoxa-3,9-diphosphaspiro[5.5]undecane 3,9-dioxide (P-CA) | 25 | 35 | 480 | 168 | 50 | 6 | 3.13 | 22.6 | NR | [49] |
| APP | — | 42 | 831 | 112 | 35 | 3 | — | 18 | NR | [50] |
| APP | 25 | 36.4 | 578 | 83 | 35 | 3 | 1.68 | 21 | NR | [50] |
| Material                                                                 | phosphorus content | nitrogen content | FSP | MFR | LOI | HT | CI | TGA | FTIR | References |
|----------------------------------------------------------------------------|--------------------|------------------|-----|-----|-----|----|----|-----|------|------------|
| APP-based intumescent flame retardant (APP-IFR)                            |                    |                  |     |     |     |    |    |     |      |            |
| 10                                                                        | 28                 | 266              | 140 | 50  | 3   | 3  | 1.58 | 19.7 |     | [52]       |
| 10                                                                        | 30                 | 315              | 25  | 35  | 5   | 2.14 | 22.8 |     |     | [52]       |
| 25                                                                        | 35                 | 407              | 15.5| 35  | 5   | 4.18 | 29.4 |     |      | [52]       |
| 20                                                                        | 37                 | 363              | 1.6 | 35  | 3   |     |     |     |      |            |
| 28                                                                        | 33                 | 62               | 24  | 35  | 3   | 12.18 |     |      | [54]       |
| 28                                                                        | 37                 | 363              | 1.6 | 35  | 3   |     |     |     |      |            |
| 30                                                                        | 65                 | 1416.6           | 128.5| 35  | 3   |     |     |     |      | [57]       |
| Phosphorus-based FR: Poly(4,4-diaminodiphenyl) methane                   |                    |                  |     |     |     |    |    |     |      |            |
| Obicyclicpentaerythritol phosphate-phosphate (P-IFR)                       |                    |                  |     |     |     |    |    |     |      |            |
| 60                                                                        | 69                 | 620              | 78.5| 35  | 3   | 3.07 |     |      | [58]       |
| 60                                                                        | 61                 | 1026             | 166 | 35  | 4   |     |     |     |      | [58]       |
| 10                                                                        | 60                 | 648              | 141 | 35  | 4   | 1.83 |     |      | [59]       |
| 10                                                                        | 84                 | 1000             | 96  | 35  | 3   |     |     |     |      | [59]       |
| Tetraethyl orthosilicate microencapsulated                                |                    |                  |     |     |     |    |    |     |      |            |
| bisphenol-A bis (diphenyl phosphate) (mc-BDP)                             |                    |                  |     |     |     |    |    |     |      | [59]       |
The information provided in Table 1 clearly reveals that APP is quite frequently used as a major phosphorus flame retardant in PP matrix. The percentage of incorporation of phosphorus flame retardants was variable from 10 to 40 wt.%. Figure 1 displays the FRI as a function of wt.% phosphorus-based FR in PP systems. The name/type of each phosphorus flame retardant is provided in the caption of Figure 1. Three formulations reached the Excellent level of flame retardancy, which is quite rare among such data pool. The loading percentage of FR in these formulations varied from 28 to 35 wt.%. Many additives were modified APP and modified phosphorus-nitrogen flame retardants. It can also be speculated that a high loading percentage cannot necessarily guarantee the Excellent level of flame retardancy; besides, the type of phosphorus FR is also an important parameter. Figure 1 also reveals that the majority of points are located in the Good zone of FRI. Therefore, it can be concluded that phosphorus-based flame retardants have quite satisfactorily reinforced PP against flame.

| Flame Retardancy Index (FRI) values as a function of phosphorus flame retardant (FR) type and content. Symbols are indicative of different types of phosphorus flame retardant used. Here: \( \text{APP-10} \) [24], 
\( \text{APP-12} \), \( \text{APP-15} \) [25], \( \text{APP-20} \) [26], \( \text{APP-20} \) [27], \( \text{APP-20} \) [28], \( \text{APP-20} \) [23], \( \text{APP-20} \) [29], \( \text{APP-25} \) [30], \( \text{APP-25} \) [31], \( \text{APP-25} \) [32], \( \text{APP-25} \) [33], \( \text{APP-25} \) [34], \( \text{APP-25} \) [35], \( \text{APP-25} \) [36], \( \text{m-APP-25} \) [37], \( \text{APP-30} \) [38], \( \text{APP-30} \) [39], \( \text{APP-30} \) [40], \( \text{APP-30} \) [41], \( \text{APP-30} \), \( \text{APP-30} \) [42], \( \text{APP-30} \), \( \text{APP-30} \) [43], \( \text{APP-30} \), \( \text{APP-30} \) [44], \( \text{APP-30} \), \( \text{APP-5} \), \( \text{APP-10} \), \( \text{APP-15} \), \( \text{APP-20} \), \( \text{APP-25} \), \( \text{APP-30} \) [45], \( \text{APP-35} \), \( \text{APP-35} \) [46], \( \text{APP-35} \), \( \text{APP-35} \) [47], \( \text{APP-40} \), \( \text{APP-40} \) [48], \( \text{APP-40} \), \( \text{CA-25} \) [49], \( \text{APP-25} \) [50], \( \text{APP-25} \) [51], \( \text{P-IFR-10} \), \( \text{P-IFR-15} \), \( \text{P-IFR-20} \), \( \text{P-IFR-25} \) [52], \( \text{P-IFR-20} \) [53], \( \text{P-IFR-28} \) [54], \( \text{P-IFR-28} \) [55], \( \text{PN-IFR-30} \) [56], \( \text{P-IFR-30} \) [57], \( \text{P-IFR-20} \) [58], \( \text{DOPO-10} \) [15], \( \text{mc-BDP-10} \), \( \text{mc-BDP-20} \) [59], \( \text{OP-20} \) [60], \( \text{AHPI-30} \) [62], \( \text{PEPA-40} \) [63].|
There has always been interest in exploring possible correlations between the data collected from different analyses made on PP materials. Figure 2 shows the flame retardancy performance of phosphorus FR-containing PP in terms of FRI versus the corresponding UL-94 test outcomes. From these data, it is evident that no specified correlation exists between the qualitative results collected from UL-94 and the quantitative ones obtained in cone calorimeter measurements. However, in the case of LOI results, Figure 3 suggests a meaningful relationship can be drawn among data achieved from the calculated FRI and the LOI test results. The LOI value for pure PP is around 17; however, it is increased by addition of flame retardant up to 36, more than a two-fold rise.

![Figure 2](image1.png)

*Figure 2. FRI values versus UL-94 test results. Symbols are indicative of different types of phosphorus flame retardant (FR) used. The vertical intervals in each category, i.e., V-0, V-1, V-2, and NR, are schematically representative of the amount of additive used. For example, two data distinguished by different symbols having the same or very close FRI values (horizontal quantity) in a given category (e.g., V-1) may have different vertical quantities, e.g., both reveal V-1 behavior in the UL-94 test, but the upper contains more FR in Polypropylene (PP).*

![Figure 3](image2.png)

*Figure 3. FRI values of PP as a function of limiting oxygen index (LOI) test results. Symbols are indicative of different types of phosphorus flame retardant used.*

### 3. Nitrogen-Based Flame Retardants

Nitrogen-based flame retardants have also been used in PP to make it resistant against fire. Table 2 gives the names and the percentages of incorporation of these flame retardants, where the data were obtained in cone calorimetry (pHRR, THR, and TTI), FRI calculated from cone calorimetry parameters, as well as LOI and UL-94 values. Some of the nitrogen-based FRs listed in Table 2 also contain a phosphorus element. However, the percentage of nitrogen is more important, and therefore these FRs are listed in this Table.
Table 2. Flame retardant PP materials containing nitrogen-based (N) flame retardants. Data are extracted from the literature: cone calorimetry parameters (TTI, pHRR, THR), LOI, and UL-94 values. The FRI values were calculated by authors of the present review. The name and the percentage of flame retardants are provided in separate columns. "wt.%" was used for loading level of additives, while "—" stands for the systems free of additive or the neat PP. * FR means flame retardant. Since all comparisons were made in terms of FRI, classification of polymers in terms of their flame-retardant properties was not surveyed based on the chemistry of additives, heat flux, sample thickness, etc.

| PP Containing Nitrogen-Based (N) FR* | wt. % | TTI (s) | pHRR (kW.m⁻²) | THR (MJ.m⁻²) | Irradiance (kW.m⁻²) | Sample Thickness (mm) | FRI | LOI | UL-94 | Ref. |
|-------------------------------------|-------|---------|---------------|--------------|-------------------|----------------------|-----|-----|-------|------|
| Melamine phosphate (MP)             | 40    | 39      | 296           | 78           | 35                | 3                    | —   | —   | —     | [63] |
| Melamine salt of pentaerythritol phosphate kaolinite (MPPK) | 15    | 30      | 208           | 70           | 35                | 4                    | 4.96| —   | NR    | [64] |
| MPPK                               | 20    | 28      | 148           | 42           | 35                | 4                    | 10.86| —   | V-0   | [64] |
| Melamine salt of tripentaerythritol phosphate (MTP) | 15    | 22      | 480           | 101          | 50                | 3                    | 1.88| —   | —     | [65] |
| MTP                                | 20    | 22      | 267           | 91           | 50                | 3                    | 3.75| 28  | V-1   | [65] |
| MTP                                | 25    | 22      | 226           | 73           | 50                | 3                    | 5.83| 32  | V-0   | [65] |
| MTP                                | 30    | 22      | 219           | 72           | 50                | 3                    | 5.78| 35  | V-0   | [65] |
| Methyl hydrogen siloxane modified MTP (or-MTP) | 30    | 21      | 253           | 72           | 50                | 3                    | 4.78| 30  | V-0   | [65] |
| Melamine pyrophosphate (MPyP)       | 30    | 36      | 437           | 103.1        | 35                | 3                    | 2.23| —   | NR    | [57] |
| MPyP                               | 34    | 34      | 1727          | 112          | 35                | 3                    | 17  | NR  | —     | [66] |
| Triazine-based charring foaming agent: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine (TA-CFA) | 30    | 13      | 584           | 96           | 35                | 3                    | 1.31| 24  | NR    | [66] |
| Triazine-based CFA: synthesized by reaction of cyanuric chloride and pipperazine (TA-CFA) | 30    | 34      | 468           | 86.6         | 35                | 3                    | 1.52| 20.5| V-1   | [39] |
| Triazine-based CFA: synthesized by polycondensation of 2-chloro-4,6-di-(2-hydroxyethylamino)-s-triazine (TA-CFA) | 30    | 38      | 518           | 86.7         | 35                | 3                    | 2.08| 23.5| NR    | [40] |
| Triazine-based CFA: synthesized from a macromolecular triazine derivative containing hydroxyethylamino and triazine rings and ethylenediamine groups (TA-CFA) | 25    | 34      | 487.4         | 91.6         | 35                | 3                    | 1.26| 21.9| NR    | [37] |
| Triazine-based CFA: Poly[N4-bis(ethylendiamino)-phenyl phosphonic-N2, N6-bis(ethylendiamino)-1,3,5-triazine-N-phenyl (TA-CFA) | 25    | 12      | 529           | 88           | 50                | 3                    | 1.00| 20.6| NR    | [32] |
| Triazine-based CA—Zinc oxide (TA-CA-ZnO) | 18    | 1457    | 156           | 50           | 3                 | —                    | 19  | NR  | —     | [29] |
| Triazine-based CA: Poly(ethanediamine-1,3,5-triazine-p-4-amino-2,2,6,6-tetramethylpiperidino) (TA-CA) | 25    | 30      | 684           | 106.7        | 35                | 3                    | 0.97| 17.8| NR    | [67] |
| Triazine-based CA: compound containing pentaerythritol and triazine structure (TA-CA) | 48    | 1351    | 107           | 35           | 3.2               | —                    | 18.5| NR  | —     | [27] |
| Triazine-based CA: synthesized by | 20    | 42      | 994           | 98           | 35                | 3.2                  | 1.29| 22  | NR    | [23] |

* FR means flame retardant.
| Reaction Type | Nitrogen Content (%) | Phosphorus Content (%) | V-0 | V-1 |
|---------------|----------------------|------------------------|-----|-----|
| reaction of tris (2-hydroxyethyl) isocyanurate and 2-carboxyethyl (phenyl) phosphonic acid (TA-CA) | — | 31 | 1239 | 123.6 | 50 | 3 | — | 18.5 | NR | [68] |
| Triazin-based IFR: synthesized by reaction of tris(2-hydroxyethyl) isocyanurate and polyphosphoric acid and melamine (TA-IFR) | 20 | 18 | 289.9 | 89 | 50 | 3 | 3.44 | 29.3 | V-0 | [68] |
| Triazin-based IFR: synthesized by reaction of cyanuric chloride and N- amino ethylpiperazine (TA-IFR) | 25 | 38 | 504 | 86.6 | 35 | 3.2 | — | 17 | NR | [31] |
| Piperazine-based FR: synthesized by reaction of diphenylphosphinyl chloride and piperazine (PI-IFR) | 30 | 58 | 487.7 | 87.5 | 50 | 3 | 7.75 | 27 | V-0 | [69] |
| Piperazine-based IFR: Piperazine spirocyclic phosphoramidate (PI-IFR) | — | 45 | 1269 | 146.4 | 50 | 3 | — | 17.5 | NR | [70] |
| Piperazine-based IFR: synthesized by reaction of phosphorus chloride and 2,6,7-trioxo-1-phosphabicyclo[2.2.2]-octane-4-methanol and anhydrous piperazine (PI-IFR) | 20 | 36 | 275 | 78 | 35 | 3 | 2.97 | 24 | NR | [71] |
| Piperazine-based IFR: synthesized by reaction of phosphorus chloride and 2,6,7-trioxo-1-phosphabicyclo[2.2.2]-octane-4-methanol and anhydrous piperazine (PI-IFR) | 30 | 37 | 209 | 74 | 35 | 3 | 4.24 | 27 | NR | [71] |
| Piperazine-based IFR: synthesized by reaction of phosphorus chloride and 2,6,7-trioxo-1-phosphabicyclo[2.2.2]-octane-4-methanol and anhydrous piperazine (PI-IFR) | 40 | 37 | 162 | 60 | 35 | 3 | 6.75 | 29 | V-0 | [71] |
| N-alkoxy hindered amine (NOR116) | 0.5 | 44 | 738.8 | 156.5 | 35 | 4 | 1.44 | 19 | NR | [72] |
| NOR116 | 0.3 | 44 | 738 | 156 | 35 | 4 | — | 17.5 | NR | [73] |
| Polyurethane containing Phosphorus-based CA (PPU-CA) | 25 | 27.3 | 475 | 83 | 35 | 3 | 1.53 | 29 | NR | [50] |
| Nitrogen-based FR: compound containing Nitrogen(27.5 wt.%) and Phosphorus(15.6 wt.%) (N-IFR) | 22 | 22 | 170 | 50.3 | 35 | 4 | — | 8.64 | 32 | V-1 | [74] |
| Nitrogen-based FR: compound containing Nitrogen(27.5 wt.%) and Phosphorus(15.6 wt.%) (N-IFR) | 25 | 21 | 160 | 49.1 | 35 | 4 | — | 8.98 | 34 | V-0 | [74] |
| Nitrogen-based IFR: Poly (diallyldimethylammonium) and polyphosphate polyelectrolyte complex-based IFR (N-IFR) | 5 | 28 | 968.5 | 103.4 | 50 | 3 | — | 1.10 | 20.2 | NR | [75] |
| Nitrogen-based IFR: Poly (diallyldimethylammonium) and polyphosphate polyelectrolyte complex-based IFR (N-IFR) | 10 | 25 | 626.2 | 97.1 | 50 | 3 | — | 1.62 | 22 | NR | [75] |
| Nitrogen-based IFR: Poly (diallyldimethylammonium) and polyphosphate polyelectrolyte complex-based IFR (N-IFR) | 15 | 23 | 543.1 | 94.3 | 50 | 3 | — | 1.76 | 24.4 | NR | [75] |
| Nitrogen-based IFR: Poly (diallyldimethylammonium) and polyphosphate polyelectrolyte complex-based IFR (N-IFR) | 20 | 21 | 443.9 | 90.1 | 50 | 3 | — | 2.06 | 26.3 | NR | [75] |
| Nitrogen-based IFR: Poly (diallyldimethylammonium) and polyphosphate polyelectrolyte complex-based IFR (N-IFR) | 25 | 18 | 335.3 | 83.9 | 50 | 3 | — | 2.52 | 27.5 | V-2 | [75] |
| — | 25 | 874.1 | 89.3 | 50 | 3 | — | 18 | NR | [76] |
Nitrogen-based IFR: compound containing Nitrogen (23%) and Phosphorus (21%) (N-IFR)  

|          | 25 | 12 | 94.9 | 68.2 | 50 | 3 | 5.78 | 33 | V-0 |          |
|----------|----|----|------|------|----|---|------|----|-----|----------|
|          |    |    |      |      |    |   |      |    |     | [76]     |
| Phosphorus and Nitrogen based IFR | 30 | 22 | 229  | 93  | 50 | — | 4.74 | 36.3 |  — | [56]     |

To give a bright view of the variation trend, Figure 4 illustrates the FRI values as a function of wt.% of nitrogen-based flame retardants incorporated into the PP. The percentage of incorporation was changed from 15 to 40 wt.%. Of note, all points are located in the Good zone of FRI, except two points remarked as Excellent. These two points correspond to a kaolinite additive modified with nitrogen and phosphorus agents. A very noticeable point to be considered is that increasing the amount of diallyldimethylammonium (nominated with the symbol in Figure 4) from 5 to 25 has no serious effect on the value of FRI, so that they are aligned vertically around FRI values between 1.0 and 2.5. Overall, like what happened to other polymers [77,78], combinatorial flame retardants may be the solution to flammability reduction of PP materials.

![Figure 4. FRI values as a function of nitrogen FR type and content. Symbols are indicative of different types of nitrogen flame retardant used. Here: ▲ MP-40 [63], ◆ MPPK-15, MPPK-20, MPPK-25 [64], ▲ MTP-15, MTP-20, MTP-25, MTP-30, m-MTP-30 [65], ▽MPyP-30 [57], ◆ MPyP-30, TA-CFA-30 [66], ◆ TA-CFA-30 [39], ▽ TA-CFA-30 [40], ◆ TA-CFA-25 [37], ▼ TA-CFA-25 [32], ◆ TA-CA-ZnO-25 [29], ◆ TA-CA-25 [67], + TA-CA-20 [27], ▹ TA-CA-20 [23], ▼ TA-IFR-20 [68], ▽ TA-IFR-25 [31], ▼ PI-IFR-25 [69], ◆ PI-IFR-30 [70], ▽ PI-IFR-20, PI-IFR-30, PI-IFR-40 [71], ▽ NOR116-0.5 [72], ▼ NOR116-0.3 [73], ◆ PPU-CA-25 [50], ▲ N-IFR-22, N-IFR-25 [74], ▼ N-IFR-5, N-IFR-10, N-IFR-15, N-IFR-20, N-IFR-25 [75], ◆ N-IFR-25 [76], ▼ PN-IFR-30 [56].](image)

Figure 5 patterns UL-94 results as a function of FRI for nitrogen-based flame retardant in PP. It can be observed that even at small quantities of FRI, V0 in UL-94 was achieved. The diversity of data in Figure 5 can be taken as a signature of sensitivity of UL-94 to FRI. Figure 6 shows LOI values as a function of FRI. There is a quite reasonable correlation between the LOI and FRI values, up to FRI value of 6.
Figure 5. FRP values versus UL-94 test results. Symbols are indicative of different types of nitrogen flame retardant (FR) used. The vertical intervals in each category, i.e., V-0, V-1, V-2, and NR, are schematically representative of the amount of additive used. For example, two data distinguished by different symbols having the same or very close FRP values (horizontal quantity) in a given category (e.g., V-1), may have different vertical quantities; e.g., both reveal V-1 behavior in UL-94 test, but the upper contains more FR in PP.

Figure 6. FRP values of PP as a function of LOI test results. Symbols are indicative of different types of nitrogen flame retardant used.

4. Mineral-Based Flame Retardants

Mineral additives have been widely used in polymers for their acceptable cost and properties [79]. Mineral-based flame retardants including clays are widely used in PP due to their low cost and acceptable thermal resistance. In this family, the most used flame retardants in volume were aluminum trihydroxide (ATH) and magnesium dihydroxide (MDH). However, due to their low efficiency, a high percentage of loading was necessary for achieving an acceptable level of flame retardancy of polymers. The name and the percentage of the used mineral-based flame retardants in PP are listed in Table 3. Cone calorimetry data, FRP, LOI, and UL-94 values are also given so as to make possible a detailed view on the status of flame retardant efficiency of PP materials.
Table 3. Flame-retardant PP materials containing mineral-based (M) flame retardants. Data are extracted from the literature: cone calorimetry parameters (TTI, pHRR, THR), LOI, and UL-94 values. The FRI values were calculated by authors of the present review. The name and the percentage of flame retardants are provided in separate columns. “wt. %” was used for loading level of additives, while “—” stands for the systems free of additive or the neat PP. * FR means flame retardant. Since all comparisons were made in terms of FRI, classification of polymers in terms of their flame-retardant properties was not surveyed based on the chemistry of additives, heat flux, sample thickness, etc.

| PP Containing Mineral-Based (M) FR | wt. % | TTI (s) | pHRR (kW·m⁻²) | THR (MJ·m⁻²) | Irradiance (kW·m⁻²) | Sample Thickness (mm) | FRI | LOI | UL-94 | Ref. |
|-----------------------------------|-------|---------|----------------|--------------|----------------------|------------------------|-----|-----|-------|------|
| Aluminum trihydroxide (ATH)      |       |         |                |              |                      |                        |     |     |       | [80] |
|                                   | 50    | 52      | 1425           | 121.4        | 50                   | 3                      | 17.3| NR  |       | [80] |
|                                   | 52    | 539     | 96.6           | 50           | 3                    | 4.66                   | 23.6| NR  |       | [80] |
|                                   | 50    | 63      | 1470           | 175          | 50                   | 4                      | 18  |     |       | [81] |
| ATH                               | 60    | 34      | 280            | 98           | 50                   | 4                      | 9.96|     |       | [81] |
|                                   | 26    | 1967    | 112            | 50           | 3                    | —                      | —   |     |       | [82] |
| ATH                               | 20    | 27      | 817            | 90           | 50                   | 3                      | 3.11|     |       | [82] |
| ATH                               | 40    | 28      | 467            | 70           | 50                   | 7.25                   |     |     |       | [82] |
| Magnesium dihydroxide (MDH)      | 20    | 31      | 1000           | 98           | 50                   | 3                      | 2.68|     |       | [82] |
| MDH                               | 40    | 34      | 433            | 75           | 50                   | 3                      | 8.87|     |       | [82] |
|                                   | 30    | 1684    | 89             | 50           | 3                    | —                      | —   |     |       | [82] |
|                                   | 63.2  | 521.35  | 49.8           | 50           | 3                    | 12.46                  |     |     |       | [82] |
| MDH                               | 62.5  | 81.1    | 115.5          | 75.7         | 50                   | —                      | 3.81|     |       | [83] |
|                                   | 71    | 2283    | 218            | 35           | 1                    | —                      |     |     |       | [82] |
| MDH                               | 50    | 97      | 789            | 238          | 35                   | 1                      | 3.62|     |       | [84] |
|                                   | 38    | 1425    | 121.4          | 50           | 3                    | 17.5                   |     |     |       | [85] |
| MDH                               | 40    | 46      | 548            | 99.1         | 50                   | 3                      | 218 |     |       | [83] |
|                                   | 29    | 1660    | 33.4           | 35           | 1                    | —                      |     |     |       | [86] |
| MDH                               | 30    | 39      | 989            | 28.3         | 35                   | 1                      | 2.66|     |       | [86] |
| Dodecanoic acid-treated MDH (m-MDH) | 30   | 32      | 882            | 28.7         | 35                   | 1                      | 2.41|     |       | [86] |
| Dodecylphosphate treated MDH (m-MDH) | 30   | 29      | 651            | 28.8         | 35                   | 1                      | 2.95|     |       | [86] |
|                                   | 37    | 584     | 75.6           | 50           | 3                    | —                      | —   |     |       | [87] |
| MDH                               | 10    | 33      | 471            | 65.9         | 50                   | 3                      | 1.26|     |       | [87] |
| MDH                               | 15    | 31      | 381            | 61.2         | 50                   | 3                      | 1.58|     |       | [87] |
|                                   | 54    | 930     | 140            | 35           | 4                    | —                      | —   | NR  |       | [64] |
| Kaolinite (Kaol)                  | 25    | 32      | 463            | 116          | 35                   | 4                      | 1.43|     |       | [84] |
| Kaol                              | 0.5   | 29      | 1474           | 142          | 50                   | 3                      | 18  |     |       | [88] |
| Kaol                              | 1.5   | 27      | 1346           | 140          | 50                   | 3                      | 1.03|     |       | [88] |
| Kaol                              | 1.5   | 27      | 1346           | 140          | 50                   | 3                      | 1.08|     |       | [88] |
| Ammonium sulfamate intercalated kaol (m-Kaol) | 0.5  | 27      | 1389           | 141          | 50                   | 3                      | 0.99|     |       | [88] |
| Ammonium sulfamate intercalated kaol (m-Kaol) | 1.5  | 28      | 1169           | 133          | 50                   | 3                      | 1.29|     |       | [88] |
| Ammonium sulfamate intercalated kaol (m-Kaol) | 3    | 27      | 1079           | 126          | 50                   | 3                      | 1.43|     |       | [88] |
| Kaol                              | 1.5   | 27      | 1346           | 140          | 50                   | 3                      | 1.11|     |       | [89] |
| Ammonium sulfamate intercalated Kaol (m-Kaol) | 1.5  | 28      | 1169           | 133          | 50                   | —                      | 1.39|     |       | [89] |
| Kaol                              | 10    | 35      | 634            | 144          | 50                   | —                      | 1.26|     |       | [90] |
| Kaol                              | 20    | 38      | 396            | 136          | 50                   | —                      | 2.33|     |       | [90] |
| Kaol                              | 30    | 41      | 348            | 126          | 50                   | —                      | 3.08|     |       | [90] |
| Trisilanolisooctyl polyhedral oligomeric silsesquioxane modified kaol (m-Kaol) | 10 35 850 140 50 — 0.96 — — [90] |
|---|---|---|---|---|---|---|---|---|
| Trisilanolisooctyl polyhedral oligomeric silsesquioxane modified kaol (m-Kaol) | 20 38 650 141 50 — 1.36 — — [90] |
| Trisilanolisooctyl polyhedral oligomeric silsesquioxane modified kaol (m-Kaol) | 30 50 430 137 50 — 2.79 — — [90] |
| Talc (TC) | 10 49 377 128 50 — 3.34 — — [90] |
| TC | 20 56 341 118 50 — 4.58 — — [90] |
| TC | 30 50 295 112 50 — 4.98 — — [90] |
| Ni-Al layered double hydroxide (LDH) | 0.5 53 1635.53 106.8 50 — 1.36 — — [91] |
| Ni-Al LDH (LDH) | 1 92 1430.59 117.8 50 — 2.46 — — [91] |
| Ni-Al LDH (LDH) | 1.5 41 1266.66 129.1 50 — 1.13 — — [91] |
| Organically modified Ni-Al LDH (m-LDH) | 0.5 59 1116.37 70.2 50 — 3.39 — — [91] |
| Organically modified Ni-Al LDH (m-LDH) | 1 45 1026.86 81.24 50 — 2.43 — — [91] |
| Organically modified Ni-Al LDH (m-LDH) | 1.5 49 1254.95 111.1 50 — 1.58 — — [91] |
| Cu-Al LDH (LDH) | 0.5 45 1026.86 81.2 50 — 2.43 — — [91] |
| Cu-Al LDH (LDH) | 1 57 1276.46 123 50 — 1.63 — — [91] |
| Cu-Al LDH (LDH) | 1.5 50 1448.98 121.8 50 — 1.27 — — [91] |
| Organically modified Cu-Al LDH (m-LDH) | 0.5 69 985.91 120 50 — 2.63 — — [91] |
| Organically modified Cu-Al LDH (m-LDH) | 1 54 1175.99 121.6 50 — 1.70 — — [91] |
| Organically modified Cu-Al LDH (m-LDH) | 1.5 54 1345.14 114.3 50 — 1.58 — — [91] |
| Mg-Al LDH with mole ratio: Zn:MgAl0.5:1 (A-LDH) | 1 15 1981 141 50 3 0.60 — — [92] |
| A-LDH | 2 16 1764 139 50 3 0.73 — — [92] |
| Zn-Mg-Al LDH with mole ratio: Zn:MgAl0.5:1.5:1 (B-LDH) | 1 14 1997 136 50 3 0.57 — — [92] |
| B-LDH | 2 14 1512 133 50 3 0.77 — — [92] |
| B-LDH | 4 13 1153 128 50 3 0.98 — — [92] |
| Zn-Mg-Al LDH with mole ratio: Zn:MgAl1:1:1 (C-LDH) | 1 18 2004 135 50 3 0.74 — — [92] |
| C-LDH | 2 14 1546 132 50 3 0.76 — — [92] |
| C-LDH | 4 12 1225 125 50 3 0.87 — — [92] |
| Zn-Mg-Al LDH with mole ratio: Zn:MgAl1:5:0.5:1 (D-LDH) | 1 18 1938 135 50 3 0.76 — — [92] |
| D-LDH | 2 15 1656 130 50 3 0.77 — — [92] |
| D-LDH | 4 13 1294 123 50 3 0.91 — — [92] |
| Zn-Al LDH with mole ratio: Zn:Al2:0:1 (E-LDH) | 1 16 1977 136 50 3 0.66 — — [92] |
| E-LDH | 2 17 1543 113 50 3 1.09 — — [92] |
| E-LDH | 4 14 1382 126 50 3 0.89 — — [92] |
| A-LDH | 1 20 1906 135 50 3 1.52 — — [92] |
| Material Description                          | A-LDH | B-LDH | C-LDH | D-LDH | E-LDH |
|----------------------------------------------|-------|-------|-------|-------|-------|
| MMT                                          | 4     | 1     | 1     | 4     | 1     |
| Modified MMT (MMT)                          | 16    | 17    | 16    | 14    | 17    |
| Dioctadecyldimethyl ammonium chloride        | 1137  | 1715  | 1875  | 992   | 2008  |
| Octadecyltrimethyl ammonium chloride (alkyl-NHCl)) | 129  | 134   | 130   | 125   | 135   |
| Dihydrogen phosphate                         | 50    | 50    | 50    | 50    | 50    |
| Intercalated Mg-Al LDH (m-LDH)               | 3     | 3     | 3     | 3     | 3     |
| Undecenoate modified Mg-Al LDH (m-LDH)       |       |       |       |       |       |
| Carbonate intercalated Mg-Al LDH (LDH)       | 10.7  | 17    | 19.7  | 52    | 31    |
| Dihydrogen phosphate intercalated Mg-Al LDH (LDH) | 50   | 50    | 50    | 50    | 50    |
| Octadecyltrimethyl ammonium chloride (alkyl-NHCl)) | 3     | 3     | 3     | 3     | 3     |
| Montmorillonite (MMT)                        | 1.2   | 3.3   | 2.7   | 1.3   | 1.5   |
| Protoned MMT (H-MMT)                         | 5.4   | 4.2   | 4.1   | 4.1   | 4.1   |
| Dioctadecyldimethyl ammonium chloride modified MMT (m-MMT) | 1.2   | 3.3   | 2.7   | 1.3   | 1.5   |
| Dioctadecyldimethyl ammonium chloride modified MMT (m-MMT) | 5.4   | 4.2   | 4.1   | 4.1   | 4.1   |

**Table Notes:**
- **A-LDH, B-LDH, C-LDH, D-LDH, E-LDH:** Various types of layered double hydroxides (LDHs) and modified MMTs.
- **Sodium dodecyl sulphate modified Ni-Al LDH (m-LDH):** Various forms of modified LDHs.
- **Undecenoate modified Mg-Al LDH (m-LDH):** Various forms of modified LDHs.
- **Carbonate intercalated Mg-Al LDH (LDH):** Various forms of intercalated LDHs.
- **Dihydrogen phosphate intercalated Mg-Al LDH (LDH):** Various forms of intercalated LDHs.
- **Octadecyltrimethyl ammonium chloride (alkyl-NHCl):** Various forms of modified compounds.
- **Montmorillonite (MMT):** Various forms of montmorillonite.
- **Protoned MMT (H-MMT):** Various forms of protonated montmorillonite.
- **Dioctadecyldimethyl ammonium chloride modified MMT (m-MMT):** Various forms of modified compounds.
- **Dioctadecyldimethyl ammonium chloride modified MMT (m-MMT):** Various forms of modified compounds.
|                          | MMT (m-MMT) | Modified MMT (m-MMT) | Alkylamide surfactant modified MMT (m-MMT) | Alkylamide surfactant modified MMT (m-MMT) | Alkylamide surfactant modified MMT (m-MMT) | Ammonium salt of an oligomer modified MMT (m-MMT) | Ammonium salt of an oligomer modified MMT (m-MMT) | Ammonium salt of an oligomer modified MMT (m-MMT) | Styrene-vinylbenzyl chloride copolymer modified MMT (m-MMT) | Styrene-vinylbenzyl chloride copolymer modified MMT (m-MMT) | Styrene-vinylbenzyl chloride copolymer modified MMT (m-MMT) | Styrene-vinylbenzyl chloride copolymer modified MMT (m-MMT) |
|--------------------------|-------------|----------------------|------------------------------------------|------------------------------------------|------------------------------------------|---------------------------------------------|---------------------------------------------|---------------------------------------------|-------------------------------------------------|-------------------------------------------------|-------------------------------------------------|-------------------------------------------------|
| Methyl tallow bis(2-     | 4.75        | 10                   | 3                                        | 10                                       | 5                                        | 2                                      | 2                                           | 12                                          | 25                                          | 5                                              | 15                                              | 25                                              |
| hydroxyethyl) amonium     | 27          | 1005                 | 35                                       | 35                                       | 35                                       | 1897                                      | 1897                                      | 1897                                      | 1897                                          | 1897                                          | 1897                                          | 1897                                          |
| modified MMT (m-MMT)      | 1365        | 99                   | 35                                       | 0.4                                      | 0.4                                      | 1.53                                      | 1.53                                      | 1.53                                      | 1.53                                          | 1.53                                          | 1.53                                          | 1.53                                          |
| Silica pillared methyl    | 4.75        | 19                   | 94                                       | 3                                        | 3                                        | 1.61                                      | 1.61                                      | 1.61                                      | 1.61                                          | 1.61                                          | 1.61                                          | 1.61                                          |
| tallow bis(2-            | 22          | 925                  | 35                                       | 0.4                                      | 0.4                                      | 1.75                                      | 1.75                                      | 1.75                                      | 1.75                                          | 1.75                                          | 1.75                                          | 1.75                                          |
| hydroxyethyl) amonium     | 2315        | 98                   | 35                                       | 0.4                                      | 0.4                                      | 1.75                                      | 1.75                                      | 1.75                                      | 1.75                                          | 1.75                                          | 1.75                                          | 1.75                                          |
| modified MMT powder       | 132         | 35                   | 3                                        | 0.73                                     | 0.73                                     | 2.27                                      | 2.27                                      | 2.27                                      | 2.27                                          | 2.27                                          | 2.27                                          | 2.27                                          |
| supported with CuO        | 35          | 0.60                 | 0.60                                     | 0.60                                     | 0.60                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 0.73                 | 0.73                                     | 0.73                                     | 0.73                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 1.61                 | 1.61                                     | 1.61                                     | 1.61                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 1.75                 | 1.75                                     | 1.75                                     | 1.75                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 2.27                 | 2.27                                     | 2.27                                     | 2.27                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 3.00                 | 3.00                                     | 3.00                                     | 3.00                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 3.50                 | 3.50                                     | 3.50                                     | 3.50                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 4.00                 | 4.00                                     | 4.00                                     | 4.00                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 4.50                 | 4.50                                     | 4.50                                     | 4.50                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 5.00                 | 5.00                                     | 5.00                                     | 5.00                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 5.50                 | 5.50                                     | 5.50                                     | 5.50                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 6.00                 | 6.00                                     | 6.00                                     | 6.00                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 6.50                 | 6.50                                     | 6.50                                     | 6.50                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 7.00                 | 7.00                                     | 7.00                                     | 7.00                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 7.50                 | 7.50                                     | 7.50                                     | 7.50                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 8.00                 | 8.00                                     | 8.00                                     | 8.00                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 8.50                 | 8.50                                     | 8.50                                     | 8.50                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 9.00                 | 9.00                                     | 9.00                                     | 9.00                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 9.50                 | 9.50                                     | 9.50                                     | 9.50                                     | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35          | 10.00                | 10.00                                    | 10.00                                    | 10.00                                    | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35         | 10 OR 35             | 10 OR 35                                 | 10 OR 35                                 | 10 OR 35                                 | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
|                          | 35         | 50 OR 35             | 50 OR 35                                 | 50 OR 35                                 | 50 OR 35                                 | 3                                         | 3                                         | 3                                         | 3                                              | 3                                              | 3                                              | 3                                              |
| Modified MMT (m-MMT) | Methyl methacrylate-vinylbenzyl chloride copolymer modified MMT (m-MMT) | 2.5 | 44 | 2025 | 123 | 50 OR 35 | — | 0.89 | — | — | [101] |
| Methyl methacrylate-vinylbenzyl chloride copolymer modified MMT (m-MMT) | 5 | 42 | 1738 | 120 | 50 OR 35 | — | 1.01 | — | — | [101] |
| Methyl methacrylate-vinylbenzyl chloride copolymer modified MMT (m-MMT) | 15 | 39 | 1651 | 115 | 50 OR 35 | — | 1.04 | — | — | [101] |
| Methyl methacrylate-vinylbenzyl chloride copolymer modified MMT (m-MMT) | 25 | 41 | 1139 | 105 | 50 OR 35 | — | 1.73 | — | — | [101] |
| Methyl methacrylate modified MMT (m-MMT) | — | 55 | 1586 | 113 | 35 | — | — | — | — | [102] |
| Methyl methacrylate modified MMT (m-MMT) | 3 | 44 | 839 | 87 | 35 | — | 1.96 | — | — | [102] |
| Methyl methacrylate modified MMT (m-MMT) | 5 | 35 | 557 | 77 | 35 | — | 2.65 | — | — | [102] |
| Nanofil (NF) | 5 | 48 | 739 | 173.4 | 35 | — | 0.92 | 22 | — | [103] |
| Organically modified bentonite (m-BT) | 5 | 45.6 | 774 | 166.6 | 35 | — | 0.87 | 22 | — | [103] |
| NF | 5 | 37 | 1047 | 174 | 50 | — | 0.83 | 22 | — | [103] |
| m-BT | 5 | 36 | 1093 | 164 | 50 | — | 0.82 | 22 | — | [103] |
| Cloisite 20A: Dimethyl, dihydrogenated tallow ammonium modified MMT (C20A) | 1 | 33 | 1751 | 105 | 35 | 3 | 0.85 | — | — | [104] |
| C20A | 3 | 34 | 1874 | 107 | 35 | 3 | 0.80 | — | — | [104] |
| C20A | 5 | 39 | 1487 | 105 | 35 | 3 | 1.19 | — | — | [104] |
| C20A | 44 | 1172 | 87.1 | 35 | 2.5 | — | 18.1 | NR | [60] |
| Cloisite 15A: Dimethyl dihydrogenated tallow ammonium cation modified sodium MMT (C15A) | 5 | 41 | 1050 | 88.2 | 35 | 2.5 | 1.02 | 18.1 | NR | [60] |
| — | 88 | 565.9 | 71.9 | 35 | 3 | — | — | — | [105] |
| C20A | 5 | 76 | 518.2 | 75.9 | 35 | 3 | 0.89 | 20 | — | [105] |
| C20A | 5 | 89 | 415.6 | 73 | 35 | 3 | 1.35 | 20 | — | [105] |
| Titanium dioxide (TiO2) | 0.5 | 99 | 488.1 | 75 | 35 | 3 | 1.25 | 20 | — | [105] |
| — | 49 | 1247 | 114.2 | 35 | — | — | — | — | [106] |
| Activated alumina (ALOx) | 2 | 35 | 943 | 108.2 | 35 | — | 0.99 | — | — | [106] |
| — | 28 | 1633 | 132 | 50 | 4 | — | 18 | — | — | [107] |
| NiFeO | 2 | 27 | 1372 | 129 | 50 | 4 | 1.17 | 18 | — | [107] |
| CoFeO | 2 | 24 | 1335 | 127 | 50 | 4 | 1.08 | 18 | — | [107] |
| — | 38 | 1284 | 241 | 50 | 6 | — | — | — | [108] |
| NiO | 7.5 | 53 | 655 | 161 | 50 | 6 | 4.09 | — | — | [108] |
| — | 64 | 1909 | 254 | 50 | 3 | — | — | — | [109] |
| Mo/Mg/Co/O catalysts (Nmm-cat) | 1 | 62 | 490 | 205 | 50 | 3 | 4.67 | — | — | [109] |
| Nmm-cat | 2 | 63 | 292 | 168 | 50 | 3 | 9.72 | — | — | [109] |
| Nmm-cat | 3 | 60 | 275 | 149 | 50 | 3 | 11.09 | — | — | [109] |
| — | 65 | 915.7 | 112.5 | 50 | 3 | — | 18 | — | — | [110] |
| Magnesium oxyxulfate whisker (MOSw) | 30 | 62 | 259.1 | 90.4 | 50 | 3 | 5.24 | 24.7 | — | [110] |
|                      | 30 | 64 | 243.3 | 72.8 | 50 | 3 | 7.15 | 26.1 | — | [110] |
|----------------------|----|----|--------|------|----|---|------|------|---|------|
| Manganese oxide (Mno) | 10 | 54 | 233.7 | 31.8 | 35 | 2 | 0.84 | —    | — | [111] |
| Manganese oxide (MnO) | 10 | 48 | 271.3 | 31.5 | 35 | 2 | 0.65 | —    | — | [111] |
| Manganese oxalate (MnCaO) | 10 | 50 | 281.6 | 29.3 | 35 | 2 | 0.70 | —    | — | [111] |
| Zinc acetyl acetonate (Znacac) | 1  | 31 | 366.28 | 35 | 1.6 | 1.73 | 19.5 | — | [53] |
| Chromium acetyl acetonate (Cracac) | 1  | 31 | 307.28 | 35 | 1.6 | 2.06 | 20.7 | — | [53] |
| Zirconium phenylphosphonate (ZrPP) | 2  | 34 | 754.99 | 35 | 3 | 1.10 | —    | — | [112] |
| Siloxane silsesquioxane resin (S4SQH) | 0  | 1  | 34  | 47 | 35 | 2 | 0.89 | —    | — | [113] |
| S4SQH | 5  | 21 | 500.44 | 44 | 35 | 2 | 0.34 | —    | — | [113] |
| S4SQH | 10 | 19 | 445.44 | 44 | 35 | 2 | 0.35 | —    | — | [113] |
| n-octyl functionalized S4SQH (m-S4SQH) | 1  | 43 | 227.29 | 35 | 2 | 2.37 | —    | — | [113] |
| n-octyl functionalized S4SQH (m-S4SQH) | 5  | 40 | 481.48 | 35 | 2 | 0.63 | —    | — | [113] |
| n-octadecyl functionalized S4SQH (m-S4SQH) | 1  | 40 | 168.22 | 35 | 2 | 3.93 | —    | — | [113] |
| n-octadecyl functionalized S4SQH (m-S4SQH) | 5  | 43 | 328.42 | 35 | 2 | 1.13 | —    | — | [113] |
| n-octadecyl functionalized S4SQH (m-S4SQH) | 10 | 47 | 391.47 | 35 | 2 | 0.93 | —    | — | [113] |
| Polysiloxane based FR (Si-FR) | 25 | 18 | 624.110 | 50 | 50 | — | 1.51 | 24.1 | NR | [30] |
| Sepiolite (SEP) | 5  | 48 | 1701.108 | 35 | 3 | 0.82 | 20 | NR | [25] |
| Organically modified SEP (m-SEP) | 0.5 | 46 | 1665.106 | 35 | 3 | 0.82 | 19.2 | NR | [25] |
| SEP | 5  | 24 | 533.68.1 | 50 | 3 | 0.78 | —    | — | [87] |
| Organically treated SEP (m-SEP) | 5  | 23 | 515.66.1 | 50 | 3 | 0.80 | —    | — | [87] |
| Methylo polyhedral oligomeric silsesquioxane (me-POSS) | 1.95 | 54 | 1023.100 | 35 | 3 | 0.85 | —    | — | [114] |
| Phenyl POSS (ph-POSS) | 3.75 | 61 | 858.98 | 35 | 3 | 1.17 | —    | — | [114] |
| Ph POSS | 12.5 | 53 | 872.96 | 35 | 3 | 1.02 | 19.5 | — | [114] |
| Octaisobutyl POSS (8-POSS) | 10 | 50 | 1325.112 | 35 | 3 | 0.73 | —    | — | [115] |
| Al POSS | 10 | 37 | 624.98 | 35 | 3 | 1.32 | —    | — | [115] |
| Zn POSS | 10 | 54 | 1069.108 | 35 | 3 | 1.02 | —    | — | [115] |
| Silica aerogel (SA) | 10 | 57 | 892.203 | 35 | 6 | 1.60 | 25.1 | — | [5] |
| Halloysite nanotube (HNT) | 8  | 44 | 495.68.5 | 50 | 3 | 1.21 | —    | — | [116] |
| HNT-Water injection (HNT-W) | 8  | 45.5 | 451.66.5 | 50 | 3 | 1.42 | —    | — | [116] |
| —  | 52.5 | 620.70.5 | 50 | 3 | — | — | — | [116] |
Figure 7 visualizes the variation of FRI value as a function of flame retardant loading in PP systems (for the convenience of readers, two figures are added for giving a better zoom on data points). This figure clearly shows that even at low loading percentages, it is possible to achieve a relatively high FRI value depending on the type of mineral. There is no denying that some parameters such as the state of dispersion and size of particles are important factors affecting the flame retardant properties.
Regarding the relationship between LOI and correlation between quantitative and qualitative parameters based on such a tiny set of data. In studied was indeed

Figure 7. FRI values as a function of the mineral FR type and content from close-up and long-shot views. Symbols are indicative of different types of mineral flame retardant used. The diversity and abundance of data were reasons why such different scales were provided for detection of behavior of PP against flame. Here: ▲ ATH-50 [80], ▲ ATH-60 [81], ▲ ATH-20, ATH-40, MDH-20, MDH-40 [82], ▲ MDH-40, MDH-60 [82], ▲ MDH-62.5 [83], ▲ MDH-50 [84], ▲ MDH-40 [85], ▲ MDH-30, m-MDH-30, m-MDH-30 [86], ▲ MDH-10, MDH-15 [87], ▲ Kaol-25 [64], ▲ Kaol-0.5, Kaol-1.5, Kaol-3, m-Kaol-0.5, m-Kaol-1.5, m-Kaol-3 [88], ▲ Kaol-1.5, m-Kaol-1.5 [89], ▲ Kaol-10, Kaol-20, Kaol-30, m-Kaol-10, m-Kaol-20, m-Kaol-30, TC-10, TC-20, TC-30 [90], ▲ LDH-0.5, LDH-1.5, m-LDH-0.5, m-LDH-1, m-LDH-1.5 [91]. ▲ A-LDH-1, A-LDH-2, B-LDH-1, B-LDH-2, B-LDH-4, C-LDH-1, C-LDH-2, C-LDH-4, D-LDH-1, D-LDH-2, D-LDH-4, E-LDH-1, E-LDH-2, E-LDH-4 [92]. ▲ A-LDH-1, A-LDH-4, B-LDH-1, B-LDH-4, C-LDH-1, C-LDH-4, D-LDH-1, D-LDH-4, E-LDH-1, E-LDH-4 [92]. ▲ A-LDH-1, A-LDH-4, B-LDH-1, B-LDH-4, C-LDH-1, C-LDH-4, D-LDH-1, D-LDH-4, E-LDH-1, E-LDH-4 [92]. ▲ A-LDH-1, A-LDH-4, C-LDH-1, C-LDH-4, E-LDH-1, E-LDH-4 [92]. ▲ m-LDH-1, m-LDH-3, m-LDH-5 [93], ▲ m-LDH-3, m-LDH-5, m-LDH-10 [94], ▲ LDH-10, m-LDH-10.7 [95], ▲ alkyl-NH3Cl-1,2, MMT-5, H-MMT-5, m-MMT-5 [96], ▲ m-MMT-4.75, m-MMT-4.75 [97], ▲ MMT-10, m-MMT-10 [24], ▲ m-MMT-3, m-MMT-10, m-MMT-16 [98], ▲ MMT-2, m-MMT-2, m-MMT-5, m-MMT-10 [99], ▲ m-MMT-3, m-MMT-8, m-MMT-12 [100], ▲ m-MMT-2.5, m-MMT-5, m-MMT-15, m-MMT-25, m-MMT-2.5, m-MMT-5, m-MMT-15, m-MMT-25 [101], ▲ m-MMT-1, ▲ m-MMT-5, ▲ Ni-5, ▲ m-BT-5 [103], ▲ Ni-5, ▲ m-BT-5 [103], ▲ C20A-1, C20A-3, C20A-5 [104], ▲ C15A-5 [60], ▲ C20A-5, C20A-5, TiO2-0.5 [105], ▲ Al2O3-2 [106], ▲ NiFeO2, CoFeO2 [107], ▲ NiO2-0.5 [108], ▲ Nmm-cat-1, Nmm-cat-2, Nmm-cat-3, Nmm-sq-1 [99], ▲ MOSW-30, ▲ m-MOSW-30 [110], ▲ MnO-10, MnO-10, MnCO3-10 [111], ▲ Znacac-1, Cracac-1 [53], ▲ ZrPP-2 [112], ▲ 54SQH-1, 54SQH-5, 54SQH-10, ▲ m-S4SQH-1, ▲ m-S4SQH-5, ▲ m-S4SQH-10 [113], ▲ SiO2-25 [30], ▲ SEP-0.5, ▲ m-SEP-0.5 [25], ▲ SE-5, ▲ m-SEP-5 [87], ▲ m-POSS-1.95, ▲ m-POSS-6.5, ▲ m-POSS-3.75, ▲ m-POSS-12.5 [114], ▲ T8-POSS-10, Al-POSS-10, Zn-POSS-10 [115], ▲ SA-10 [5], ▲ HNT-8, ▲ HNT-W-8 [116], ▲ HNT-8, ▲ HNT-W-4, ▲ HNT-W-8, ▲ HNT-W-16 [116], ▲ HNT-5, ▲ HNT-10, ▲ HNT-15, ▲ m-HNT-5, ▲ m-HNT-10, ▲ m-HNT-15 [117].

Unfortunately, the number of papers in which cone calorimetry, UL-94, and LOI values were studied was indeed limited, but the ones available are used plotting Figure 8. It should be noted that no formulation among studied ones is rated at VO. In conclusion, it is quite difficult to find a correlation between quantitative and qualitative parameters based on such a tiny set of data. In regard to the relationship between LOI and FRI, a meaningful trend can still be seen in Figure 9.
Figure 8. FRI values versus UL-94 test results. Symbols are indicative of different types of mineral flame retardant (FR) used. The vertical intervals in each category, i.e., V-0, V-1, V-2, and NR, are schematically representative of the amount of additive used. For example, two data distinguished by different symbols having the same or very close FRI values (horizontal quantity) in a given category (e.g., V-1), may have different vertical quantities, e.g., both reveal V-1 behavior in UL-94 test, but the upper contains more FR in PP.

Figure 9. FRI values of PP as a function of LOI test results. Symbols are indicative of different types of mineral flame retardant used.

5. Carbon-Based Flame Retardants

Carbon-based additives have been widely used in developing polymer composites and nanocomposites [118–121]. However, due to expense and limited interaction with PP, a few works based on carbon-based flame retardants have been reported on flame-retardant PP materials. Table 4 summarizes all information available on the flame-retardant PP materials containing carbon-based additives.
Table 4. Flame-retardant PP materials containing carbon-based (C) flame retardants. Data are extracted from the literature: cone calorimetry parameters (TTI, pHRR, THR), LOI, and UL-94 values. The FRI values were calculated by authors of the present review. The name and the percentage of flame retardants are provided in separate columns. "wt.%" was used for loading level of additives, while "—" stands for the systems free of additive or the neat PP. * FR means flame retardant. Since all comparisons were made in terms of FRI, classification of polymers in terms of their flame-retardant properties was not surveyed based on the chemistry of additives, heat flux, sample thickness, etc.

| PP Containing Carbon-Based (C) FR* | wt.% | TTI (s) | pHRR (kW.m\(^{-2}\)) | THR (MJ.m\(^{-2}\)) | Irradiance (kW.m\(^{-2}\)) | Sample Thickness (mm) | FRI | LOI | UL-94 | Ref. |
|----------------------------------|------|---------|-------------------|------------------|---------------------|----------------------|-----|-----|-------|------|
| Graphene (GN)                    | 25   | 2.4     | 105.9             | 35               | 0.95                | 3                    | —   | —   | —     | [106]|
| Activated alumina decorated GN (m-GN) | 2    | 34     | 103.4             | 35               | 1.24                | 3                    | —   | —   | —     | [106]|
| P-phenylenediamine modified reduced graphene oxide (m-rGNO) | 2    | 33     | 98.4              | 35               | 0.87                | 3                    | —   | —   | —     | [122]|
| Polyaniline nanofiber modified rGNO (m-rGNO) | 2    | 27     | 967              | 35               | 0.76                | 3                    | —   | —   | —     | [123]|
| Hexachlorocyclopiphosph azene modified rGNO (m-rGNO) | 2    | 35     | 829              | 92               | 1.42                | 3                    | —   | —   | —     | [123]|
| Zirconium phenylphosphonate decorated rGNO (m-rGNO) | 2    | 39     | 676             | 89.8             | 1.55                | 3                    | —   | —   | —     | [112]|
| Graphene oxide (GNO)             | 2    | 33     | 979             | 108.2             | 35               | 0.65                | —   | —   | —     | [124]|
| Melamine modified GNO (m-GNO)    | 0.5  | 40     | 892             | 104.1             | 35               | 0.91                | —   | —   | —     | [124]|
| Melamine modified GNO (m-GNO)    | 1    | 37     | 834             | 100.6             | 35               | 0.93                | —   | —   | —     | [124]|
| Melamine modified GNO (m-GNO)    | 2    | 33     | 739             | 98.7             | 35               | 0.95                | —   | —   | —     | [124]|
| GN                               | 2.5  | 39     | 1279              | 58.8             | 35               | 0.98                | —   | —   | —     | [125]|
| GN-Nickel oxide (GN-NiO)         | 2.5  | 35     | 1110              | 45.4             | 35               | 1.32                | —   | —   | —     | [125]|
| GN and Ni-Ce mixed oxide (GN-NiCeO\(\)) | 2.5  | 32     | 956             | 39.2             | 35               | 1.62                | —   | —   | —     | [125]|
| rGNO                             | 2    | 32     | 1199              | 97.8             | 35               | 1.14                | —   | —   | —     | [126]|
| Phosphomolybdic acid modified rGNO (m-rGNO) | 1    | 27     | 773              | 39.6             | 35               | 1.14                | —   | —   | —     | [126]|
| Phosphomolybdic acid modified rGNO (m-rGNO) | 2    | 23     | 737             | 38.4             | 35               | 1.05                | —   | —   | —     | [126]|
| Phosphomolybdic acid modified rGNO (m-rGNO) | 3    | 25     | 700             | 38.4             | 35               | 1.21                | —   | —   | —     | [126]|
| Poly(4,4-diaminodiphenyl methane spirocyclicpentaerythiol bisphosphonate)-4,4-diaminodiphenyl methane modified rGNO (m-rGNO) | 20   | 66     | 397              | 73.9             | 35               | 4.88                | —   | —   | —     | [58]|

* FR means flame retardant.
Figure 10 shows that with low loading percentage (1 wt.%) of carbon nanotubes, it is possible to achieve the Good FRI. No data were available for UL-94 tests. Comparison between Figures 7 and 10 also suggests that low-cost minerals were used at higher loadings, while carbon-based additives were used almost at loadings below 10 wt.%. A limited number of data have also been reported on LOI values. These points are plotted as a function of FRI in Figure 11, where a good correlation can be established between FRI and LOI values. Deeper understanding of the mechanism behind such correlation requires a detailed view of the origin of tests as well as the chemical structure of additives and possible interaction between the PP and additives.
7. Bio-Based Flame Retardants

In recent years, due to sustainability issues, the use of bio-based additives has also been investigated in PP. However, the number of research papers is limited on this subject. Table 5 gives the name and loading percentage of these bio-based FR. The obtained results from cone calorimetry, LOI, and UL-94 tests are also listed in Table 5. Figures 12 and 13 display UL-94 and LOI results as a function of FRI for bio-based flame retardant in PP, respectively.

Table 5. Flame-retardant PP materials containing bio-based (Bio) flame retardants. Data are extracted from the literature: cone calorimetry parameters (TTI, pHRR, THR), LOI, and UL-94 values. The FRI values were calculated by authors of the present review. The name and the percentage of flame retardants are provided in separate columns. “wt. %” was used for loading level of additives, while “—” stands for the systems free of additive or the neat PP. * FR means flame retardant. Since all
comparisons were made in terms of FRI, classification of polymers in terms of their flame-retardant properties was not surveyed based on the chemistry of additives, heat flux, sample thickness, etc.

| PP Containing Bio-Based (Bio) FR * | wt.% | TTI (s) | pHRR (kW.m⁻²) | THR (MJ.m⁻²) | Irradiance (kW.m⁻²) | Sample Thickness (mm) | FRI | LOI | UL-94 | Ref. |
|-----------------------------------|------|--------|----------------|-------------|-------------------|----------------------|-----|-----|-------|------|
| Cycloextrin nanospponge (CD)      | 10   | 34     | 1462           | 80          | 35                | 3                    | --  | --  | --    | [135]|
| Hydroxyapatite and Cycloextrin-based FR (HAandCD-FR) | 10   | 32     | 708            | 156         | 35                | 4                    | 0.87| --  | --    | [15] |
| Propylene-block-ethylene copolymer | 20   | 38     | 380            | 74.2        | 35                | 3                    | 3.24| 22.5| --    | [136]|
| Phosphorus and nitrogen elements modified lignin (m-lig) | 20   | 38     | 380            | 74.2        | 35                | 3                    | 3.24| 22.5| --    | [136]|
| Phytic acid and Piperazine-based FR (PHPI-FR) | 15   | 17     | 388.5          | 108.5       | 50                | 3.2                  | 2.22| 25  | V-2   | [137]|
| PHPI-FR                           | 15   | 17     | 386.2          | 108.4       | 50                | 3.2                  | 2.22| 25  | V-2   | [137]|
| PHPI-FR                           | 25   | 16.5   | 303.4          | 105.4       | 50                | 3.2                  | 2.82| 27  | V-0   | [137]|
| Biochar (BC)                      | 15   | 12     | 753.01         | 112.68      | 50                | 3.2                  | --  | 0.49| --    | [138]|
| BC                                | 25   | 13.3   | 616.31         | 111.26      | 51                | 3.2                  | --  | 0.68| --    | [138]|
| BC                                | 30   | 15     | 539.34         | 101.2       | 52                | 3.2                  | --  | 0.96| --    | [138]|
| BC                                | 35   | 16.3   | 477.22         | 98.31       | 53                | 3.2                  | --  | 1.22| --    | [138]|
| Wool                              | 40   | 12.3   | 858.7          | 1388.3      | 12.4              | 3.2                  | 0.85| --  | NR    | [28] |
| Phosphoric acid-treated wool fiber (m-wool) | 40   | 14.3   | 426.7          | 72          | 50                | 2.4                  | 2.13| --  | NR    | [28] |
| Phosphoric acid-treated wool fiber (m-wool) | 40   | 15     | 436.3          | 65.3        | 50                | 2.4                  | 2.41| --  | V-0   | [28] |
| Phosphoric acid-treated chicken feather (m-CF) | 40   | 14.7   | 336.7          | 57          | 50                | 2.4                  | 3.51| --  | V-0   | [28] |
| Chicken feather (CF)              | 40   | 17     | 1234.1         | 76.1        | 50                | 2.4                  | 0.69| --  | NR    | [139]|
| Phosphoric acid and ethylenediamine treated chicken feather (m-CF) | 40   | 19.3   | 280.5          | 58.7        | 50                | 2.4                  | 4.47| --  | V-0   | [139]|
| Phosphoric acid and ethylenediamine treated chicken feather (m-CF) | 40   | 17.7   | 216.1          | 52.4        | 50                | 2.4                  | 5.96| --  | V-0   | [139]|

**Figure 12.** FRI values versus UL-94 test results. Symbols are indicative of different types of bio-based flame retardant (FR) used. The vertical intervals in each category, i.e., V-0, V-1, V-2, and NR, are schematically representative of the amount of additive used. For example, two data distinguished by
different symbols having the same or very close FRI values (horizontal quantity) in a given category (e.g., V-1) may have different vertical quantities, e.g., both reveal V-1 behavior in UL-94 test, but the upper contains more FR in PP.

Figure 13. FRI values of PP as a function of LOI test results. Symbols are indicative of different types of bio-based flame retardant used. The green triangles are related to a mixture of phytic acid and piperazine-based FR. The increase of LOI is directly related to the percentage of FR loading, 15, 18, 20, and 25 wt.%.

FRI values are plotted as a function of loading percentage of bio-based FR in Figure 14. It can be observed that a high quantity of bio-based FR, 40 wt.% is needed to achieve FRI equal to 6.

Figure 14. FRI values as a function of bio-based FR type and content. Symbols are indicative of different types of bio-based flame retardant used. Here: ▲ CD-10 [135], ● HAandCD-FR-10 [15], ▲ m-lig-20 [136], ▼ PHPI-FR-15, PHPI-FR-18, PHPI-FR-20, PHPI-FR-25 [137], ▲ BC-15, BC-25, BC-30, BC-35 [138], □ Wool-40, m-wool-40, m-wool-40, m-CF-40 [28], ▼ CF-40, m-CF-40, m-CF-40 [139].

6. Combination of Flame Retardants

As observed in previous sections, using an additive alone can to a limited extent improve flame-retardant properties of PP. Combination of flame retardants is a strategy to improve further the flame retardancy via synergism between various flame retardants [140–142]. Moreover, the quantity of the used flame retardant can be reduced in polymer so as to prevent mechanical properties deterioration. Different combinative additive systems were considered in PP. The corresponded data are collected and summarized in Table 6. The third column gives the ratio between flame retardants.
| Name                        | wt. | Type of FR | TTI (s) | pHRR (kW.m⁻²) | THR (MJ.m⁻²) | Irradiance (kW.m⁻²) | Sample Thickness (mm) | FRI  | LOI  | UL-94 | Ref. |
|-----------------------------|-----|------------|---------|---------------|--------------|---------------------|----------------------|------|------|-------|------|
| APP/Pentaerythritol (APP/PER) | 20  | PnP 2:1    | 18      | 514.7         | 92.6         | 50                  | 3                    | 1.86 | 27.6 | V-2   | [68] |
| APP/PER                      | 25  | PnP 2:1    | 17      | 442.3         | 98.8         | 50                  | 3                    | 2.38 | 30.1 | V-0   | [143]|
| APP/PER                      | 25  | PnP 3:1    | 32      | 363.7         | 82           | 35                  | 3                    | 2.60 | 24.7 | V-1   | [67] |
| APP/PER                      | 30  | PnP 3:1    | 13      | 392           | 80           | 35                  | 3                    | 2.35 | 31   | V-0   | [144]|
| Hydroxy silicone oil co-     | 30  | PnP 3:1    | 10      | 325           | 78           | 35                  | 3                    | 2.24 | 32.5 | V-0   | [144]|
| microencapsulated APP and PER (APPandPER)) |      |            |         |               |              |                     |                      |      |      |       |      |
| APP/PER/Melamine (APP/PER/MEL) | 29  | PnP 1:0.7:1:0.92 | 175 | 76.4 | 55 | 25 | 10.85 | — | — | — | [145] |
| APP/PER/MEL                  | 33  | PnP 1:64:1:0.94 | 188 | 65 | 46.3 | 25 | 3 | 16.27 | — | — | — | [145] |
| APP/PER/MEL                  | 36  | PnP 1:2:1:0.92 | 180 | 68 | 49.4 | 25 | 3 | 13.96 | — | — | — | [145] |
| APP/PER/MEL                  | 29  | PnP 1:0.7:1:0.92 | 158 | 72.5 | 50 | 3 | 9.81 | — | — | — | [145] |
| APP/PER/MEL                  | 33  | PnP 1:64:1:0.94 | 115 | 67.8 | 50 | 3 | 15.72 | — | — | — | [145] |
| APP/PER/MEL                  | 36  | PnP 1:2:1:0.92 | 133 | 73.2 | 50 | 3 | 10.49 | — | — | — | [145] |
| APP/PER/MEL                  | 29  | PnP 1:0.7:1:0.92 | 115 | 67.8 | 50 | 3 | 15.72 | — | — | — | [145] |
| APP/PER/MEL/MDH              | 37  | PnP 1:64:1:0.90.7 | 156 | 63.9 | 25 | 3 | 12.30 | 25.2 | — | — | — | [146] |
| APP/PER/MDH                  | 37  | PnP 1:64:1:0.90.7 | 196 | 79.5 | 45.1 | 50 | 3 | 14.24 | 25.2 | — | — | — | [146] |
| APP/PER/MEL/20A              | 21  | PnP 3:1:10:2:5 | 25 | 463 | 89 | 35 | 3 | 2.89 | — | — | — | [104] |

Table 6. The flame retardancy performance of PP containing various combinations of flame retardants in terms of FRI (*the name and percentage of incorporated flame retardants are given after PP). P = phosphorus FR, Np = non-phosphorus FR, N = nitrogen FR, nN = non-nitrogen-based FR, M = mineral FR, Bio = bio-based FR, nBio = non-bio-based FR (one can also consider some nitrogen-based FRs containing phosphorus as the combination of phosphorus and nitrogen resulting in synergism, Table 2). Since all comparisons were made in terms of FRI, classification of polymers in terms of their flame-retardant properties was not surveyed based on the chemistry of additives, heat flux, sample thickness, etc.
| System                                      | C20A | APP/Pn| P:nP:nP | Tg (°C) | Td (°C) | Shore D (A) | V0 (%) | V1 (%) | NR (%) |
|---------------------------------------------|------|-------|---------|---------|---------|-------------|--------|--------|--------|
| APP/PER/MEL/C20A                            | 23   | PnP^nP| 3:1:1:0.7:5 | 26      | 430     | 91          | 35     | 3      | 3.17   |
| APP and MMT/PER/MEL/C20A                    | 21   | PnP^nP| 3:1:1:0.2:5 | 24      | 306     | 81          | 35     | 3      | 4.62   |
| APP and MMT/PER/MEL/C20A                    | 23   | PnP^nP| 3:1:1:0.7:5 | 21      | 344     | 80          | 35     | 3      | 3.64   |
| γ-aminopropyl triethoxysilane modified APP/Dipentaerythritol/MEL (m-APP/DPER/MEL) | 25   | PnP^nP| 4:1:1     | 30      | 71      | 32.5        | 35     | 4      | 14.93  |
| γ-aminopropyl triethoxysilane modified APP/DPER/MEL/SEP (m-APP/DPER/MEL/SEP) | 25   | PnP^nP| 4:1:1:0.2:5 | 29      | 51      | 30.8        | 35     | 4      | 21.20  |
| APP/PER/Melamine formaldehyde (APP/PER/MF)   | 18   | PnP^nP| 3:1:0.2   | 48      | 352.3   | 110.4       | 35     | 3      | 3.27   |
| APP/PER/Adenosine monophosphate embedded Melamine formaldehyde (APP/PER/MFA) | 18   | PnP^nP| 3:1:0.2   | 49      | 355.1   | 108.1       | 35     | 3      | 3.38   |
| APP/PER/Trizine-based FR: N,N',N''-1,3,5-triazine-2,4,6-triytrimethylammonium bis(triethylene glycol) bisamide (APP/PER/TAF) | 18   | PnP^nP| 2:4:1:0.2 | 42      | 323.3   | 126         | 35     | 3      | 3.06   |
| APP/PER/Uracil: nitrogenous bases (APP/PER/G bases) | 18   | PnP^nP| 3:1:0.3   | 46      | 293     | 105         | 35     | 3      | 3.25   |

[References: 104, 107, 147, 148, 149, 150, 72, 71]
| Material Type                          | P:nP   | 2886 | 144  | 50  | 3     | 17.6 | NR   | [152] |
|---------------------------------------|--------|------|------|-----|-------|------|------|-------|
| APP/PER                               | 25     | PnP  | 2.2:1| 30  | 386   | 117  | 50   | 3     | 2.23  | 26.8  | V-1   | [152] |
| Aluminum chloride modified APP/DPER   | 25     | PnP  | 2.2:1| 25  | 226   | 104  | 50   | 3     | 3.57  | 32.1  | V-0   | [152] |
| APP/DPER/AT                           | 25     | PnP  | 2.1:1| 28  | 381   | 108  | 50   | 3     | 2.28  | 28.7  | V-0   | [152] |
| APP/PER                               | 28.5   | PnP  | 3:1  | 24  | 318   | 122  | 50   | 4     | 2.27  | 30    | V-0   | [70]  |
| APP/PER/KAOL                          | 28.5   | PnP  | 3:1:2| 22  | 222   | 131  | 50   | 4     | 2.78  | 33    | V-0   | [70]  |
| APP/PER                               | 29     | PnP  | 3:1  | 21  | 160.7 | 94.2 | 35   | 3     | 0.82  | 29.5  | V-0   | [153] |
| APP/PER/MT                            | 29     | PnP  | 3:1:0.46 | 42 | 149.8 | 69.5 | 35   | 3     | 2.38  | 34.5  | V-0   | [153] |
| APP/PER/Melamine modified MMT         | 29     | PnP  | 3:1:0.46 | 37 | 157.9 | 55.1 | 35   | 3     | 2.51  | 36.5  | V-0   | [153] |
| APP/PER/Triphenylphosphonium modified MMT | 29 | PnP  | 3:1:0.46 | 38 | 168.2 | 84.7 | 35   | 3     | 1.57  | 34.8  | V-0   | [154] |
| APP/PER                               | 25     | PnP  | 3:1  | 39  | 267   | 111  | 35   | 4     | 2.57  | 26.3  | V-1   | [155] |
| APP/PER/acid-treated waste silicon rubber composite insulator (APP/PER/m-SiR) | 25 | PnP  | 3:1:0.16 | 35 | 296   | 109  | 35   | 4     | 2.12  | 27    | V-1   | [155] |
| APP/PER/acid and N₂ plasma-treated SiR (APP/PER/m-SiR) | 25 | PnP  | 3:1:0.16 | 39 | 271   | 102  | 35   | 4     | 2.76  | 29.3  | V-0   | [155] |
| APP/PER/acid and N₂ plasma-treated SiR | 25 | PnP  | 3:1:0.16 | 53 | 655   | 108.1| 35   | 3     | 1.75  | 28.9  | V-2   | [156] |
| System                        | P:nP Ratio | P1 | P2 | P3 | N1 | N2 | N3 | V0 | V1 | NR | Ref. |
|-------------------------------|------------|----|----|----|----|----|----|----|----|----|------|
| Methyl hydrogen siloxane-treated APP/DPER (m-APP/DPER) | 25         | PnP 2:1 | 19 | 347 | 113 | 50 | 3  | 2.02 | 32.5 | V-0 | [157] |
| Methyl hydrogen siloxane-treated APP/DPER/Zeolite (m-APP/DPER/Z) | 26         | PnP:nP 2:1:0.1 | 21 | 209 | 50 | 50 | 3  | 8.41 | 35.6 | V-0 | [157] |
| Methyl hydrogen siloxane-treated APP/DPER/Z/MWCNT (m-APP/DPER/Z/MWCNT) | 26.1       | PnP 2:1:0.1 | 21 | 226 | 60 | 50 | 3  | 6.48 | 34.3 | V-0 | [157] |
| Methyl hydrogen siloxane-treated APP/DPER/Allop (APP/DPER/ALL) | 27         | PnP 3:1:0.3 | 53 | 149 | 68 | 35 | 3  | 3.77 | 35  | V-0 | [158] |
| APP/DPER/Zeolite (APP/DPER/Z) | 25         | PnP 2:1 | 18 | 436 | 123 | 50 | 3  | 2.28 | 27.6 | V-1 | [159] |
| APP/DPER/Meso porous aluminosilicate oxide (APP/DPER/MAO) | 25         | PnP 2:1:0.33 | 31 | 188 | 55 | 50 | 3  | 20.37 | 33.9 | V-0 | [159] |
| APP/DPER/Zn-MAO | 25         | PnP 2:1:0.12 | 27 | 223 | 105.6 | 50 | 4  | 4.83 | 30  | V-0 | [160] |
| APP/DPER/Organ (APP/DPER/m-SEP) | 25         | PnP 2:1:0.41 | 31 | 237 | 104.3 | 50 | 4  | 5.28 | 35  | V-0 | [160] |
| APP/DPER/Organ (APP/DPER/m-SEP) | 25         | PnP 2:1:0.57 | 29 | 263 | 135.8 | 50 | 4  | 3.42 | 24.5 | NR | [160] |
| APP/DPER/Organ (APP/DPER/m-SEP) | 25         | PnP 2:1:0.74 | 29 | 362 | 158.4 | 2.13 | 21.5 | NR | [160] |
| APP/DPER/Octaphenyl POSS (APP/DPER/OP-PSS) | 20         | PnP 2:1:0.12 | 30 | 247 | 104.1 | 2.17 | 21.5 | NR | [160] |
| APP/DPER/Aminopropyl isobutyl-octaphenyl | 20         | PnP 3:1 | 27.2 | 229 | 44 | 35 | 3  | 1.67 | 24.5 | NR | [161] |
| APP/DPER/Organo POSS (APP/DPER/OP-PSS) | 20         | PnP 3:1:0.2 | 32.3 | 178 | 27 | 35 | 3  | 4.17 | 28.1 | V-1 | [161] |
| System | Molar Ratio | P₀ | P₁ | P₃ | T | V | Reference |
|--------|-------------|----|----|----|---|---|-----------|
| APP/PER/Octaammonium POSS (APP/PER/OA-POSS) | 20 | P₀P₁P₃ 3:1:0.2 | 37.7 | 164 | 26 | 35 | 3 | 5.48 | 29.7 | V-1 | [161] |
| APP/PER/Trisulfonylpropyl POSS (APP/PER/TS-POSS) | 20 | P₀P₁P₃ 3:1:0.2 | 35.4 | 153 | 29 | 35 | 3 | 4.95 | 32.4 | V-1 | [161] |
| APP/PER | 30 | P₀ | 52.5 | 70.43 | 49.96 | 35 | 3 | 7.10 | 29 | — | [162] |
| APP/PER/Thermally-treated solid waste (APP/PER/T-RS) | 33.5 | P₀P₁P₃ 2:1:0.5 | 48 | 65.71 | 40.21 | 35 | 3 | 8.65 | 41 | — | [162] |
| APP/PER/Volcanic ash (APP/PER/VC) | 33.5 | P₀P₁P₃ 2:1:0.5 | 101 | 19.73 | 29.48 | 35 | 3 | 82.72 | 37 | — | [162] |
| APP/PER/Rice husk ash (APP/PER/CHR) | 33.5 | P₀P₁P₃ 2:1:0.5 | 62 | 48.16 | 31.36 | 35 | 3 | 19.55 | 40 | — | [162] |
| APP/PER | — — — | 65 | 920 | 145 | 35 | 4 | — | 17.5 | NR | — | [163] |
| APP/PER/Zinc borate (APP/PER/ZnB) | 20 | P₀P₁P₃ 3:1:0.2 | 37 | 330 | 125 | 35 | 4 | 1.84 | 29.5 | V-0 | [163] |
| APP/PER/Borophosphate (APP/PER/BPO₄) | 20 | P₀P₁P₃ 3:1:0.2 | 33 | 226 | 53 | 35 | 4 | 5.65 | 30 | V-0 | [163] |
| APP/PER/Boron silicon containing preceramic oligomer (APP/PER/Bsi) | 20 | P₀P₁P₃ 3:1:0.2 | 34 | 255 | 70 | 35 | 4 | 3.90 | 25.5 | V-0 | [163] |
| APP/PER/Lanthanum borate (APP/PER/LaB) | 20 | P₀P₁P₃ 3:1:0.2 | 43 | 260 | 97 | 35 | 4 | 3.49 | 27 | V-0 | [163] |
| APP/PER | — — — | 28 | 1633 | 132 | 50 | 4 | — | 18 | — | [107] |
| APP/PER/NiFeO | 25 | P₀P₁P₃ 2:1:0.35 | 20 | 425 | 107 | 50 | 4 | 3.38 | 34.6 | — | [107] |
| APP/PER/CoFeO | 25 | P₀P₁P₃ 2:1:0.35 | 19 | 323 | 124 | 50 | 4 | 3.65 | 35 | — | [107] |
| APP/PER | — — — | 28 | 1337 | 95.1 | 35 | 3 | — | 18 | NR | — | [164] |
| APP/PER/Zinc hydroxystannate | 25 | P₀P₁P₃ 2:1:0.26 | 54 | 363.2 | 88.2 | 35 | 3 | 7.65 | 36 | V-0 | [164] |
| APP/PER/Nickel phosphide nanocrystalline (APP/PER/Ni₃P) | 25 | P₀P₁P₂ 3:2:1 | 79 | 306.6 | 89.1 | 35 | 3 | 13.12 | 34 | V-0 | [164] |
| APP/PER/Cobalt phosphide nanocrystalline (APP/PER/Co₃P) | 25 | P₀P₁P₂ 3:2:1 | 42 | 562.4 | 93.2 | 35 | 3 | 3.63 | 31.5 | V-1 | [164] |
| APP/PER/Copper phosphide nanocrystalline (APP/PER/Cu₃P) | — — — | 75 | 471 | 102 | 50 | 3 | — | 18 | NR | — | [165] |
| APP/PER | 25 | P₀P₁P₂ 3:2:1 | 45 | 265 | 83 | 50 | 3 | 1.31 | 27 | V-1 | [165] |
| APP/PER/Zinc hydrosulfostannate | 25 | P₀P₁P₂ 3:2:1:0.16 | 40 | 193 | 75 | 50 | 3 | 1.77 | 32 | V-0 | [165] |
| e         | (APP/PER/ZHS) |         | 24 | 1361 | 107.5 | 50 | 3 | — | — | — | 166 |
|-----------|---------------|---------|----|------|--------|----|----|---|---|---|-----|
| APP/PER   | 25 PnP 3:1    | 21      | 455| 85.4 | 50     | 3  | 3.29| — | V-2| — | 166 |
| APP/PER/Mang anese acetate | (APP/PER/MnAc) | 26 PnPnP 3:1:0.16 | 23 | 372 | 75.2 | 50 | 3 | 5.01| — | V-0| 166 |
| APP/PER/MnAc | 27 PnPnP 3:1:0.32 | 19 | 366 | 74.1 | 50 | 3 | 4.27| — | V-0| 166 |
| APP/PER/MnAc | 28 PnPnP 3:1:0.48 | 19 | 383 | 83.6 | 50 | 3 | 3.61| — | V-0| 166 |
| APP/PER/MnAc | 29 PnPnP 3:1:0.64 | 18 | 369 | 96.1 | 50 | 3 | 3.09| — | V-0| 166 |
| APP/DPER/phosphorylated sodium alginate | (APP/DPER/m-SA) | 35 PnPnP 3:1:1 | 27 | 335 | 128 | 35 | 4 | 7.55| — | — | 167 |
| APP/PEPA | 23 PnP 2:1 | 38 | 297.9 | 113.8 | 35 | 4 | 2.08 | 30.5 | NR | 168 |
| APP/PEPA/NO R16 | 25 PnPnP 2:1:0.26 | 36 | 260.3 | 112.5 | 35 | 4 | 2.28 | 34 | V-2 | 168 |
| APP/PEPA/Zirconium phosphate | (APP/PEPA/ZrP) | 25 PnPnP 2:1:0.26 | 41 | 221.8 | 112.5 | 35 | 4 | 3.05 | 31.5 | V-2 | 168 |
| APP/PEPA/Macromolecular N-alkoxy hindered amine functionalized ZrP | (APP/PEPA/m-ZrP) | 25 PnPnP 2:1:0.26 | 40 | 157 | 112.2 | 35 | 4 | 4.22 | 33 | V-0 | 168 |
| APP/PEPA/PER/Kaol | 27 | 1474 | 142 | 50 | 3 | — | 18 NR | 169 |
| MCAPP/PER/ | 25 PnP/PnP 2:1:0.2 | 17 | 373 | 123 | 50 | 3 | 2.87 | 32.5 | V-0 | 169 |
| MCAPP/PER/ Acidically modified kaol (MCAPP/PEPA/m-Kaol) | 25 PnPnP 2:1:0.2 | 20 | 233 | 105 | 50 | 3 | 6.33 | 34.9 | V-0 | 169 |
| MCAPP/PER/ | 27 | 1474 | 142 | 50 | 3 | — | 18.1 NR | 170 |
| MCAPP/PER/ | 25 PnP 2:1 | 18 | 438 | 123 | 50 | 3 | 2.59 | 31.1 | V-2 | 170 |
| MCAPP/PER/ | 25 PnPnP 2:1:0.2 | 17 | 372 | 123 | 50 | 3 | 2.88 | 32.5 | V-0 | 170 |
| MCAPP/PER/ Thiourea modified kaol (MCAPP/PEPA/m-Kaol) | 25 PnPnP 2:1:0.2 | 21 | 291 | 103 | 50 | 3 | 5.43 | 35.4 | V-0 | 170 |
| MCAPP/PER/ | 27 | 1474 | 142 | 50 | 3 | — | 18 NR | 171 |
| MCAPP/PER/ | 25 PnP 2:1 | 18 | 438 | 123 | 50 | 3 | 2.59 | 31.1 | V-2 | 171 |
| MCAPP/PER/ | 25 PnPnP 2:1:0.2 | 17 | 373 | 123 | 50 | 3 | 2.87 | 32.5 | V-0 | 171 |
| MCAPP/PER/ Kaol nanoroll (MCAPP/PEPA/Kaol nanoroll) | 25 PnPnP 2:1:0.2 | 19 | 269 | 120 | 50 | 3 | 4.56 | 34.5 | V-0 | 171 |
| MCAPP/PER/ | 27 | 1474 | 142 | 50 | 3 | — | 18 NR | 89 |
| MCAPP/PER/ | 25 PnP 2:1 | 18 | 438 | 123 | 50 | — | 2.59 | 31.1 | V-2 | 89 |
| MCAPP/PER/ | 25 PnPnP 2:1:0.2 | 17 | 373 | 123 | 50 | — | 2.87 | 32.5 | V-0 | 89 |
| MCAPP/PEPA/  | P:P:nP  | 18  | 309 | 125 | 50  | 3   | 2.61 | 35.3 | V-0  | [89] |
| Ammonium   | 2:1:0.2 |     |     |     |     |     |     |      |      |      |
| sulfamate  |          |     |     |     |     |     |     |      |      |      |
| intercalated|          |     |     |     |     |     |     |      |      |      |
| kaol       |          |     |     |     |     |     |     |      |      |      |
| (MCAPP/PEPA/  |          |     |     |     |     |     |     |      |      |      |
| m-Kaol)    |          |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P  | 18  | 438 | 123 | 50  | 3   | 2.59 | 31   | V-2  | [172]|
| d APP/PEPA  | 2:1     |     |     |     |     |     |     |      |      |      |
| (mc-APP/PEPA) |          |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P:nP  | 17  | 373 | 123 | 50  | 3   | 2.87 | 32.5 | V-0  | [172]|
| d APP/PEPA/Kaol | 2:1:0.2 |     |     |     |     |     |     |      |      |      |
| (mc-APP/PEPA/Kaol) |          |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P:nP  | 18  | 341 | 109 | 50  | 3   | 3.75 | 35.2 | V-0  | [172]|
| d APP/PEPA/HNT | 2:1:0.2 |     |     |     |     |     |     |      |      |      |
| (mc-APP/PEPA/HNT) |          |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P:nP: | 19  | 263 | 97  | 50  | 3   | 5.77 | 36.9 | V-0  | [172]|
| d APP/PEPA/Kaol/ | nP:  |     |     |     |     |     |     |      |      |      |
| HNT (mc-APP/PEPA/Kaol/HNT) | 2:1:0.18:0.2 |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P  | 18  | 436 | 123 | 50  | 3   | 2.60 | 31.2 | V-2  | [173]|
| d APP/PEPA  | 2:1     |     |     |     |     |     |     |      |      |      |
| (mc-APP/PEPA) |          |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P:nP  | 17  | 374 | 122 | 50  | 3   | 2.88 | 32.5 | V-2  | [173]|
| d APP/PEPA/Kaol | 2:1:0.2 |     |     |     |     |     |     |      |      |      |
| (mc-APP/PEPA/Kaol) |          |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P:nP  | 23  | 299 | 106 | 50  | 3   | 5.62 | 34.1 | V-0  | [173]|
| d APP/PEPA/HSA -A | 2:1:0.2 |     |     |     |     |     |     |      |      |      |
| (mc-APP/PEPA/HSA -A) |          |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P:nP  | 20  | 257 | 84  | 50  | 3   | 7.18 | 35.1 | V-0  | [173]|
| d APP/PEPA/HSA -P | 2:1:0.2 |     |     |     |     |     |     |      |      |      |
| (mc-APP/PEPA/HSA -P) |          |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P:nP  | 16  | 248 | 103 | 50  | 3   | 4.85 | 35.5 | V-0  | [173]|
| d APP/PEPA/ | 2:1:0.2 |     |     |     |     |     |     |      |      |      |
| HSA-A-La (mc-APP/PEPA/HSA-A-La) |          |     |     |     |     |     |     |      |      |      |
| Microcapsulate | P:P:nP  | 17  | 212 | 82  | 50  | 3   | 7.58 | 37.5 | V-0  | [173]|
| d APP/PEPA/ | 2:1:0.2 |     |     |     |     |     |     |      |      |      |
| HSA-P-La (mc-APP/PEPA/HSA-P-La) |          |     |     |     |     |     |     |      |      |      |
| — — —      | 38  | 1284| 214 | 50  | 6   | 18  | 18.2 | NR   |      | [49]|
| APP/Phosphorus based CA: 3,9- Bis-(1-oxo-2,6,7-trioxo-1-phospha- bicyclo[2.2.2]oct-4-ylmethoxy)-2,4,8,10-tetraoxa- | 25  | P:P  | 1:1  |     |     |     |     |      |      |      |
| System                        | P:nP       | ρ      | Δt      | ΔL   | Mass Loss | V   | References |
|-------------------------------|------------|--------|---------|------|-----------|-----|------------|
| 3,9-diphospha-spiro[5.5]undecane 3,9-dioxide (APP/P-CA) | —          | —      | 37      | 1284 | 121    | 50  | 3          | —    | —    | [38] |

| APP/Phosphorus-based FR: Cyclotriphosph azene containing six (aminopropyl)triethoxysilicon e groups (APP/P-FR) | P:nP       | ρ      | Δt      | ΔL   | Mass Loss | V   | References |
|------------------------------------------------------------------------------------------------------------------|
| 30:14:1                                                            | 18         | 596    | 114     | 50   | 3        | 1.11| 22.2      | NR   | [38] |
| 6:5:1                                                            | 17         | 420    | 109     | 50   | 3        | 1.55| 22.4      | NR   | [38] |
| 30:4:1                                                            | 18         | 382    | 95      | 50   | 3        | 2.08| 23.5      | V-2  | [38] |
| 30:2.75:1                                                         | 17         | 282    | 95      | 50   | 3        | 2.66| 26.5      | V-2  | [38] |

| Melamine-formaldehyde-tris(2-hydroxyethyl) isocyanurate resin microencapsulated APP/Tris(2-hydroxyethyl) isocyanurate (mc-APP/THIEC) | P:nP       | ρ      | Δt      | ΔL   | Mass Loss | V   | References |
|------------------------------------------------------------------------------------------------------------------|
| 30:3:1                                                            | 28         | 232    | 100.7   | 50   | 3        | 5.56| 36        | V-0  | [41] |

| APP/Polyurethane containing phosphorus-based CA (APP/PPU-CA) | P:nP       | ρ      | Δt      | ΔL   | Mass Loss | V   | References |
|------------------------------------------------------------------------------------------------------------------|
| 25:2:1                                                            | 19.8       | 232    | 69      | 35   | 3        | 2.74| 24.5      | V-2  | [50] |

| APP/PPU-CA | P:nP       | ρ      | Δt      | ΔL   | Mass Loss | V   | References |
|-------------------------------------------------------------|------------|--------|---------|------|-----------|-----|------------|
| 25:1:1                                                            | 17.1       | 288    | 70      | 35   | 3        | 1.87| 25.5      | V-1  | [50] |

| APP/Triazine-based CFA (APP/TA-CFA) | P:nP       | ρ      | Δt      | ΔL   | Mass Loss | V   | References |
|------------------------------------|------------|--------|---------|------|-----------|-----|------------|
| 22:4:1                                                            | 21         | 397.3  | 161.1   | 50   | 4        | 2.74| 30.4      | V-0  | [95] |

| 3-(Aminopropyl)triethoxysilane modified APP microcapsulated with methylpolysilox | P:nP       | ρ      | Δt      | ΔL   | Mass Loss | V   | References |
|--------------------------------------------------------------------------------|------------|--------|---------|------|-----------|-----|------------|
| 22:4:1                                                            | 16         | 271.7  | 140.8   | 50   | 4        | 3.49| 31.7      | V-0  | [95] |
| APP/Triazine-based CFA (m-APP/TA-CFA) | — | 20 | 809 | 96 | 50 | 3 | — | 17.6 | NR | [32] |
|--------------------------------------|---|----|-----|----|----|---|---|-----|----|-----|
| APP/Triazine-based CFA: Poly[N4-bis(ethylenediamino)-phenyl phosphonic-N2, N6-bis(ethylenediamino)-1,3,5-triazine-N-phenyl (APP/TA-CFA)] | 25 | P:nP | 2:1 | 11 | 121 | 81 | 50 | 3 | 4.35 | 34 | V-0 | [32] |
| APP/Triazine-based CFA: synthesized from a macromolecular triazine derivative containing hydroxyethylamino and triazine rings and ethylenediamino groups (APP/TA-CFA) | 25 | P:nP | 4:1 | 40 | 167.6 | 82.5 | 35 | 3 | 4.82 | 34 | V-0 | [37] |
| Melamine and phytic acid modified APP/Triazine-based CFA: synthesized from a macromolecular triazine derivative containing hydroxyethylamino and triazine rings and ethylenediamino groups (m-APP/TA-CFA) | 25 | P:nP | 4:1 | 43 | 115.6 | 82.3 | 35 | 3 | 7.53 | 35 | V-0 | [37] |
| APP/Triazine-based CFA: synthesized by polycondensation of 2-chloro-4,6-di(2-hydroxyethylamino)-S-triazine (APP/TA-CFA) | 30 | P:nP | 2:1 | 56 | 422 | 70.7 | 35 | 3 | 4.62 | 32.5 | V-0 | [40] |
| APP/Triazine-based CFA: synthesized by polycondensation of 2-chloro-4,6-di(2-hydroxyethylamino)-S-triazine (APP/TA-CFA) | 30 | P:nP | 3:1 | 48 | 316 | 68.8 | 35 | 3 | 5.43 | 33 | V-0 | [40] |
| APP/Triazine-based CFA: synthesized by polycondensation of 2-chloro- | 30 | P:nP | 4:1 | 52 | 414 | 71.1 | 35 | 3 | 4.35 | 31.5 | V-0 | [40] |
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| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and piperazine (APP/TA-CFA) | — | — | 48 | 988 | 88.3 | 35 | 3 | — | 17 | NR | 39 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and piperazine (APP/TA-CFA) | 30 | P:N 1:1 | 32 | 82.4 | 77.9 | 35 | 3 | 9.06 | 29 | V-0 | 39 |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and piperazine (APP/TA-CFA) | 30 | P:nP 2:1 | 52 | 94.2 | 78.4 | 35 | 3 | 12.79 | 32 | V-0 | 39 |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and piperazine (APP/TA-CFA) | 30 | P:nP 3:1 | 34 | 167 | 83.4 | 35 | 3 | 4.43 | 34 | V-0 | 39 |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and piperazine (APP/TA-CFA) | 30 | P:nP 4:1 | 36 | 163.6 | 67.9 | 35 | 3 | 9.16 | 29.5 | V-0 | 39 |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and piperazine (APP/TA-CFA) | 20 | P:nP 3:1 | 36 | 132 | 58.7 | 35 | 3 | 7.91 | 31 | V-0 | 174 |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and piperazine /hexadecyl trimethyl ammonium bromide modified MMT (APP/TA-CFA/m-MMT) | 20 | P:nP:nP 3:1:0.1 | 36 | 132 | 58.7 | 35 | 3 | 7.91 | 31 | V-0 | 174 |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and piperazine /hexadecyl trimethyl ammonium bromide modified MMT (APP/TA-CFA/m-MMT) | 20 | P:nP:nP 3:1:0.2 | 36 | 90 | 58.6 | 35 | 3 | 11.63 | 30.5 | V-0 | 174 |
| APP/Triazine-based CFA: |  |  |  |  |  |  |  |  |
|------------------------|---|---|---|---|---|---|---|
| reaction of cyanuric   | 20| P:nP:nP| 32| 52.6| 35.5| 35| 3.2|
| chloride and piperazine |  |  |  |  |  |  |  | [174]
| /hexadecyl trimethyl   |  |  |  |  |  |  |  |
| ammonium bromide       |  |  |  |  |  |  |  |
| modified MMT           |  |  |  |  |  |  |  |
| (APP/TA-CFA/m-MMT)     |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |
| APP/Triazine-based CFA: |  |  |  |  |  |  |  | [174]
| reaction of cyanuric   | 20| P:nP:nP| 38| 55.2| 38.5| 35| 3.2|
| chloride and piperazine |  |  |  |  |  |  |  | [174]
| /hexadecyl trimethyl   |  |  |  |  |  |  |  |
| ammonium bromide       |  |  |  |  |  |  |  |
| modified MMT           |  |  |  |  |  |  |  |
| (APP/TA-CFA/m-MMT)     |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |
| APP/Triazine-based CFA: |  |  |  |  |  |  |  | [174]
| reaction of cyanuric   | 20| P:nP:nP| 36| 112.7| 41.4| 35| 3.2|
| chloride and            |  |  |  |  |  |  |  | [174]
| ethanolamine            |  |  |  |  |  |  |  |
| and ethylenediamine    |  |  |  |  |  |  |  | [61]
| and Silicon            |  |  |  |  |  |  |  |
| dioxide (APP/TA-CFA/SiO) |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |
| APP/Triazine-based CFA: |  |  |  |  |  |  |  | [61]
| reaction of cyanuric   | 24| P:nP:nP| 30| 236.6| 84.7| 50| 3|
| chloride and            |  |  |  |  |  |  |  | [61]
| ethanolamine            |  |  |  |  |  |  |  |
| and ethylenediamine/e   |  |  |  |  |  |  |  |
| Silicon dioxide (APP/TA-CFA/SiO) |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |
| APP/Triazine-based CFA: |  |  |  |  |  |  |  |  |
| reaction of cyanuric    | 24| P:nP:nP| 33| 221| 83.2| 50| 3|
| chloride and            |  |  |  |  |  |  |  | [61]
| ethanolamine            |  |  |  |  |  |  |  |
| and ethylenediamine/e   |  |  |  |  |  |  |  |
| AHP/SiO (APP/TA-CFA/AHP/SiO) |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |
| APP/Triazine-based CFA: |  |  |  |  |  |  |  |  |
| reaction of cyanuric    | 24| P:nP:nP| 26| 88| 20| 50| —|
| chloride and            |  |  |  |  |  |  |  |  |
| ethanolamine            |  |  |  |  |  |  |  |
| and ethylenediamine/e   |  |  |  |  |  |  |  |
| N-ethyl triazinepiperazine copolymer/SiO |  |  |  |  |  |  |  |
| (APP/TA-CFA/N-ethyl triazinepiperazine copolymer/SiO) |  |  |  |  |  |  |  | [175]

[174]"[175]"
| (APP/TA-CFA/SiO₂)                        | P:nP:nP 4:1:0.26 | 27 | 95 | 24 | 50 | — | 49.30 | 33.5 | V-0 | [175] |
|----------------------------------------|------------------|----|----|----|----|---|-------|------|-----|-------|
| APP/Triazine-based CFA: N-ethyl triazineepiperazine copolymer/SiO₂ (APP/TA-CFA/SiO₂) |                  |    |    |    |    |   |       |      |     |       |
| — — 50 1025 110.8 35 3 — 17 NR [176] |                  |    |    |    |    |   |       |      |     |       |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine (APP/TA-CFA) |                  |    |    |    |    |   |       |      |     |       |
| — — 50 1025 110.8 35 3 — 17 NR [176] |                  |    |    |    |    |   |       |      |     |       |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine/rGNO (APP/TA-CFA/rGNO) |                  |    |    |    |    |   |       |      |     |       |
| 25 P:nP:nP 4:1:0.1 | 35 | 140 | 90.4 | 35 | 3 | 6.28 | 32 | V-0 | [176] |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine/rGNO (APP/TA-CFA/rGNO) |                  |    |    |    |    |   |       |      |     |       |
| 25 P:nP:nP 4:1:0.2 | 34 | 156 | 86 | 35 | 3 | 5.75 | 28 | V-0 | [176] |
| APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine/rGNO (APP/TA-CFA/rGNO) |                  |    |    |    |    |   |       |      |     |       |
| 25 P:nP:nP 4:1:0.4 | 36 | 262 | 94.4 | 35 | 3 | 3.30 | 25 | V-2 | [176] |
| Piperazine modified APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine (m-APP/TA-CFA) |                  |    |    |    |    |   |       |      |     |       |
| — — 45 1456 139.1 35 3 — 17 NR [177] |                  |    |    |    |    |   |       |      |     |       |
| Piperazine modified APP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine (m-APP/TA-CFA) |                  |    |    |    |    |   |       |      |     |       |
| 25 P:nP:nP 4:1:0.1 | 39 | 434 | 125.1 | 35 | 3 | 3.23 | 34 | V-0 | [177] |
| APP/Triazine based CFA: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine/e/rGNO (m-APP/TA-CFA/rGNO) | Piperazine modified APP/Triazine based CFA: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine/e/rGNO (m-APP/TA-CFA/rGNO) | 25 | P:nP:nP: 4:1:0.2 | 37 | 350 | 123.9 | 35 | 3 | 3.84 | 32 | V-0 | [177] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 25 | P:nP:nP: 4:1:0.4 | 37 | 397 | 125.4 | 35 | 3 | 3.34 | 30 | V-0 | [177] |
| 25 | P:nP:nP: 3.6:1:0.5 | 38 | 401 | 117.3 | 35 | 3 | 3.63 | 33 | V-0 | [177] |
| 25 | P:nP:nP: 3.04:1:0.96 | 34 | 290 | 117.1 | 35 | 3 | 4.50 | 30.5 | V-0 | [177] |
| Method (m-APP/m-APP@rGNO/TA-CFA) | P:nP | Pm | Tm | Tp | Tm-P | V-0 | Notes |
|---------------------------------|------|----|----|----|------|-----|-------|
| Piperazine modified APP/Piperazine modified APP attached with rGNO/Triazine-based CFA: synthesized by reaction of cyanuric chloride and ethanolamine and ethylenediamine | 25   | PnP 1.04:1.0. 46 | 31   | 464  | 125.8 | 35  | 3     | 2.39 | 26.5 | V-2 | [177] |
| APP/Triazine-based CA: synthesized by reaction of 2-carboxyethyl (phenyl) phosphinic acid and tris (2-hydroxyethyl) isocyanurate (APP/TA-CA) | 20   | P:N 1:1 | 38   | 83   | 41   | 35  | 3     | 4.73 | 30   | V-0 | [23]  |
| APP/Triazine-based CA: synthesized by reaction of cyanuric chloride, 2,6,7-trioxa-1-phosphabicyclo [2.2.2]octane-4-methanol and piperazine (APP/TA-CA) | 20   | P:nP 2:1 | 39   | 255  | 101  | 35  | 3     | 4.56 | 27.5 | V-0 | [27]  |
| APP/Triazine-based CA: synthesized by reaction of cyanuric chloride, 2,6,7-trioxa-1-phosphabicyclo [2.2.2]octane-4-methanol and piperazine (APP/TA-CA) | 20   | P:nP 3:1 | 37   | 253  | 98   | 35  | 3     | 4.49 | 28   | V-0 | [27]  |
| APP/Triazine-based CA: Poly(ethanedia mine-1,3,5-triazine-p-4-amino-2,2,6,6-tetramethylpiperidine) (APP/TA-CA) | 25   | PnP 2:1 | 28   | 227.9 | 62   | 35  | 3     | 4.69 | 30.3 | V-0 | [67]  |
| APP/Triazine-based CA: poly(1,3,5-triazin-2-aminoethanol | 25   | PnP 3:1 | 17   | 219  | 165  | 50  | 6     | 4.42 | 32.7 | V-0 | [36]  |
| APP/Triazine based CA: synthesized by reaction of cyanuric chloride and 2,6,7-trioxa-1-phosphabicyclo[2.2.2]octane-4-methanol and diethylenetriamine (APP/TA-CA) | 30 | P:nP 2:1 | 35 | 187 | 68 | 50 | 3 | 7.32 | 31.5 | V-0 | [178] |
| APP/Triazine based CA: synthesized by reaction of cyanuric chloride and diphenylamine and ethylenediamine (APP/TA-CA) | 30 | P:nP 2:1 | 35 | 253.7 | 91.2 | 50 | 3 | 4.76 | 32.8 | V-0 | [178] |
| APP/Triazine-based CA: synthesized by reaction of cyanuric chloride and 2,6,7-trioxa-1-phosphabicyclo[2.2.2]octane-4-methanol and diethylenetriamine (APP/TA-CA) | 30 | P:nP 2:1 | 35 | 349.9 | 91.3 | 50 | 3 | 3.45 | 34.6 | V-0 | [178] |
| APP/Triazine-based CA: synthesized by reaction of cyanuric chloride and diethyleneamine (APP/TA-CA) | 25 | P:nP 3:1 | 18 | 176 | 82 | 50 | 6 | 11.72 | 35 | V-0 | [36] |
| and γ-Aminopropyl triethoxysilane (APP/TA-CA) | APP/Triazine-based CA: synthesized by reaction of Cyanuric chloride and Ethanedi-amine and γ-Aminopropyl triethoxysilane/NOR116 (APP/TA-CA/NOR116) |
|---|---|
| | 25 P:nP 2:1:0.03 | 32 | 76 | 122 | 35 | 4 | 13.02 | 42.5 | V-0 |
| APP/Triazine-based CA: synthesized by polycondensation of 2-amino-4,6-dichloro-s-triazines and diethylenetriamine (APP/TA-CA) | | 20 P:nP 2.8:1 | 35 | 218 | 126 | 35 | 6 | 4.03 | 30.8 | V-1 |
| APP/Triazine-based CA: synthesized by polycondensation of 2-amino-4,6-dichloro-s-triazines and diethylenetriamine/Organically modified MMT (APP/TA-CA/m-MMT) | | 20 P:nP 2.8:1:0.2 | 34 | 159 | 64 | 35 | 6 | 10.58 | 33 | V-0 |
| APP/Triazine-based CA: synthesized by polycondensation of 2-amino-4,6-dichloro-s-triazines and diethylenetriamine/Organically modified MMT (APP/TA-CA/m-MMT) | | 20 P:nP 2.8:1:0.6 | 37 | 270 | 156 | 35 | 6 | 2.78 | 28.9 | NR |
| APP/Triazine-based CA: synthesized by reaction of | | 22 P:nP 4:1:0.26 | 16 | 91 | 62 | 50 | 4 | 20.84 | 29.6 | V-0 |
| APP/Triazine-based CA: N-methyl triazineethylene diamine copolymer/SiO₂ (APP/TA-CFA/SiO₂) | | 22 P:nP 4:1:0.26 | 22 | 74 | 69 | 50 | 4 | 31.67 | 29.3 | V-0 |
| APP/Triazine-based CA: synthesized by reaction of | | 25 P:nP 2:1 | 19 | 191 | 112 | 50 | 3 | 11.21 | 31.1 | V-0 |
cyanuric chloride and γ-aminopropyltriethoxy silane and trimethylamine and ethylenediamine (APP/TA-CA-ZnO)

| P:nP:nP | 2.25:1.0:12 | 18 | 430 | 132 | 50 | 3 | 4.00 | 26.1 | V-1 | [29] |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |

APP and Triazine-based CA: synthesized by reaction of cyanuric chloride and γ-aminopropyltriethoxy silane and trimethylamine and ethylenediamine (APP/TA-CA/ZnO)

| P:nP:nP | 1:1 | 44 | 313 | 235 | 35 | 6 | 3.12 | 26 | NR | [182] |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |

APP and Triazine-based IFR (APPandTA-IFR)

| P:nP:nP | 2:1 | 38 | 117 | 23.2 | 35 | 3.2 | 25.44 | 29.5 | V-0 | [31] |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |

APP and Triazine-based IFR (APPandTA-IFR)

| P:nP:nP | 3:1 | 36 | 113 | 73.9 | 35 | 3.2 | 7.83 | 30.5 | V-0 | [31] |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |

APP and Piperazine-Triazine-based CA: synthesized by reaction of cyanuric chloride and...
| System                              | Compound/Abbreviation  | Formula | P:nP | M:nP | T:nP | V0  | Nt  | Reference |
|-------------------------------------|------------------------|---------|------|------|------|-----|-----|-----------|
| APP/Piperazine-Triazine-based CA    | APP/PI-TI-CA           |         |      |      |      |     |     | [33]      |
| cluster: synthesized by reaction of |                        |         |      |      |      |     |     |           |
| cyanuric chloride and anhydrous    |                        |         |      |      |      |     |     |           |
| piperazine (APP/PI-TI-CA)           |                        |         |      |      |      |     |     |           |
|                                     |                        | 25      | 21   | 242  | 96   | 50  | 3   | 3         |
|                                     |                        |         |      |      |      |     |     | 17.5     |
|                                     |                        |         |      |      |      |     |     | NR [70]  |
| APP/Piperazine-based IFR:           |                        |         |      |      |      |     |     |           |
| Piperazine spirocyclic phosphorami- |                        |         |      |      |      |     |     |           |
| dat e (APP/PI-IFR)                 |                        |         |      |      |      |     |     |           |
|                                     |                        | 30      | 23   | 208.8| 81.41| 50  | 3   | 5.58      |
|                                     |                        |         |      |      |      |     |     | 32.5      |
|                                     |                        |         |      |      |      |     |     | V-0 [70] |
| APP/Piperazine-based IFR:           |                        |         |      |      |      |     |     |           |
| Piperazine spirocyclic phosphorami- |                        |         |      |      |      |     |     |           |
| dat e/Triazine based CFA (APP/PI-   |                        |         |      |      |      |     |     |           |
| IFR/TA-CFA)                        |                        | 30      | 23   | 116.1| 41.57| 50  | 3   | 19.66     |
|                                     |                        |         |      |      |      |     |     | 39.8      |
|                                     |                        |         |      |      |      |     |     | V-0 [70] |
| APP/ATH                            |                        |         |      |      |      |     |     |           |
|                                     |                        | 30      | 40   | 210  | 75   | 35  | 3   | 1.55      |
|                                     |                        |         |      |      |      |     |     | 24        |
|                                     |                        |         |      |      |      |     |     | V-1 [183] |
| 4,4’-diphenylmethane diisocyanate   |                        |         |      |      |      |     |     |           |
| and melamine co-microencapsulat ed |                        |         |      |      |      |     |     |           |
| APP and ATH (mc-APPandATH))        |                        | 30      | 75   | 120  | 53   | 35  | 3   | 7.20      |
|                                     |                        |         |      |      |      |     |     | 25.5      |
|                                     |                        |         |      |      |      |     |     | V-0 [183] |
| APP/MMT                            |                        |         |      |      |      |     |     |           |
|                                     |                        | 10      | 25   | 769  | 66   | 35  | 0.4| 4.11      |
|                                     |                        |         |      |      |      |     |     |           |
| APP/MMT                            |                        | 10      | 27   | 765  | 65   | 35  | 0.4| 4.53      |
|                                     |                        |         |      |      |      |     |     |           |
| APP/Modified MMT (APP/m-MMT)        |                        | 10      | 29   | 715  | 64   | 35  | 0.4| 5.29      |
|                                     |                        |         |      |      |      |     |     |           |
| APP/Modified MMT (APP/m-MMT)        |                        | 10      | 30   | 619  | 63   | 35  | 0.4| 6.43      |
|                                     |                        |         |      |      |      |     |     |           |
| APP/Nf                             |                        | 20      | 40.8 | 399  | 167.9| 35  |     | 1.49      |
|                                     |                        |         |      |      |      |     |     | 23        |
|                                     |                        |         |      |      |      |     |     |           |
| APP/organically modified BT (APP/m- |                        | 20      | 42.6 | 386  | 155.1| 35  |     | 1.75      |
| BT)                                |                        |         |      |      |      |     |     | 23        |
|                                     |                        |         |      |      |      |     |     |           |
| APP/Nf                             |                        | 20      | 40.8 | 399  | 167.9| 35  |     | 1.49      |
|                                     |                        |         |      |      |      |     |     | 23        |
|                                     |                        |         |      |      |      |     |     |           |
| APP/organically modified BT (APP/m- |                        | 20      | 42.6 | 386  | 155.1| 35  |     | 1.75      |
| BT)                                |                        |         |      |      |      |     |     | 23        |
|                                     |                        |         |      |      |      |     |     |           |
| APP/C15A                           |                        | 20      | 61   | 490  | 72.7 | 35  | 2.5| 3.97      |
|                                     |                        |         |      |      |      |     |     | 20.1      |
|                                     |                        |         |      |      |      |     |     | NR [60]  |
| OP/C15A                            |                        | 20      | 50   | 400  | 83.6 | 35  | 2.5| 3.46      |
|                                     |                        |         |      |      |      |     |     | 20.8      |
|                                     |                        |         |      |      |      |     |     | NR [60]  |
| APP/C20A/PER/MEL                    |                        | 25      | 26   | 403  | 93   | 35  | 3   | 3.31      |
|                                     |                        |         |      |      |      |     |     |           |
|                                     |                        |         |      |      |      |     |     |           |
|                                     |                        |         |      |      |      |     |     |           |
| APP/Polymer | P:nP:nP | 20A/PER/MEL | 20A/PER/MEL | 20A/PER/MEL | 20A/PER/MEL |
|-------------|---------|-------------|-------------|-------------|-------------|
| APP/PP | nP | 2.4:1:0.8 | 23 | 385 | 80 | 35 | 3 | 3.56 | 2.04 |
| APP/SiO | nP | 1.2:1:0.4 | 18 | 460 | 86 | 35 | 3 | 2.17 | 2.04 |
| APP/SiO | nP | 1.8:1:0.6 | 18 | 411 | 86 | 35 | 3 | 2.43 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 9:1 | 20 | 200 | 59.3 | 50 | 4 | 9.85 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 5:6:1 | 18 | 327 | 150.8 | 50 | 4 | 2.14 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 9:1 | 18 | 286 | 108.8 | 50 | 3 | 3.85 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 4:1 | 15 | 326 | 120.3 | 50 | 3 | 2.54 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 9:1 | 16 | 306 | 136 | 50 | 3 | 2.56 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 4:1 | 14 | 265 | 90.4 | 50 | 3 | 3.89 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 1:1 | 14 | 277 | 97 | 50 | — | 3.00 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 1:5:1 | 15 | 168 | 91 | 50 | — | 5.65 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 1:5:1 | 14 | 288 | 98 | 50 | — | 3.46 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 2:1 | 18 | 245 | 93 | 50 | — | 4.55 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 3:1 | 20 | 197 | 98 | 50 | — | 5.97 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 19:1 | 36 | 153 | 130 | 50 | — | 17.5 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 19:1 | 30 | 502 | 151 | 50 | — | 1.64 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 19:1 | 35 | 1203 | 197.6 | 50 | 6 | — | 18.2 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 7:3:1 | 32 | 343.6 | 157.2 | 50 | 6 | 4.02 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 4:1 | 31 | 316.2 | 136.9 | 50 | 6 | 4.86 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 2:5:1 | 33 | 302.1 | 121.4 | 50 | 6 | 6.11 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 1:1 | 24 | 910 | 89 | 35 | 3 | 3.89 | 2.04 |

APP/Phytic acid modified LDH (APP/m-LDH) | P:nP | 1:1 | 38 | 1284 | 214 | 50 | 6 | — | 18.2 | 2.04 |
| Phosphorus based CA: 3,9-Bis-(1-oxo-2,6,7-trioxo-1-phosphabicyclo[2.2.2]oct-4-ylmethoxy)-2,4,6,10-tetraoxa-3,9-diphosphaspiro[5.5]undecane 3,9-dioxide/MEL (P-CA/MEL) | P:nP | V | 0 | [184] |
|---|---|---|---|---|---|
| 30 | 4:1 | 18 | 198 | 175 | 50 | 6 | 3.75 | 31.6 | V-0 | [57] |
| Phosphorus based FR: Tri (1-oxo-2,6,7-trioxo-1-phosphabicyclo [2.2.2] octane-methyl) phosphate/MPyP | P:N | 1:1 | 30 | 40 | 175.2 | 90.6 | 35 | 3 | 7.05 | V-0 | [57] |
| Phosphorus-based FR: Poly(4,4-diaminodiphenyl methane spirocyclicpentaerythritol bisphosphonate)/GNO (P-IFR/GNO) | P:nP | 10:1 | 20 | 64 | 473 | 79 | 35 | — | 3.71 | — | [58] |
| Phosphorus-based IFR: Poly(4,4-diaminodiphenyl methane Obicyclicpentae erythritol phosphate-phosphate)/Znacac (PN-IFR/Znacac) | P:nP | 19:1 | 20 | 23 | 175 | 25 | 35 | 1.6 | 3.00 | 27.4 | — | [53] |
| Phosphorus-based IFR: Poly(4,4-diaminodiphenyl methane Obicyclicpentae erythritol phosphate-phosphate)/Cracac (P-IFR/Cracac) | P:nP | 19:1 | 20 | 24 | 184 | 24 | 35 | 1.6 | 3.10 | 28.2 | — | [53] |
| Phosphorus and Nitrogen-based IFR: compound containing Phosphorus and Nitrogen/Dioctadecyl dimethyl ammonium chloride modified MMT (PN-IFR/m-MMT) | P:nP | 59:1 | 30 | 20 | 173 | 102 | 50 | — | 5.20 | 38.5 | — | [56] |
| Phosphorus and Nitrogen- | P:nP | 29:1 | 30 | 22 | 192 | 127 | 50 | — | 4.14 | 38 | — | [56] |
based IFR: compound containing Phosphorus and Nitrogen /Dioctadecyl dimethyl ammonium chloride modified MMT (PN-IFR/m-MMT)

| Phosphorus and Nitrogen-based IFR: compound containing Phosphorus and Nitrogen /Dioctadecyl dimethyl ammonium chloride modified MMT (PN-IFR/m-MMT) | 30 | P:nP | 19:1 | 21 | 173 | 134 | 50 | — | 4.16 | 37.7 | — | [56] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Phosphorus and Nitrogen-based IFR: compound containing Phosphorus and Nitrogen /Dioctadecyl dimethyl ammonium chloride modified MMT (PN-IFR/m-MMT) | 30 | P:nP | 14:1 | 22 | 176 | 133 | 50 | — | 4.31 | 35.7 | — | [56] |
| Phosphorus and Nitrogen-based IFR: compound containing Phosphorus and Nitrogen /Dioctadecyl dimethyl ammonium chloride modified MMT (PN-IFR/m-MMT) | 30 | P:nP | 11:1 | 20 | 135 | 131 | 50 | — | 5.19 | 35.8 | — | [56] |
| Phosphorus and Nitrogen-based IFR: compound containing Phosphorus and Nitrogen /Dioctadecyl dimethyl ammonium chloride modified MMT (PN-IFR/m-MMT) | 30 | P:nP | 9:1 | 21 | 155 | 134 | 50 | — | 4.64 | 31 | — | [56] |
| Phosphorus and Nitrogen-based IFR: compound containing Phosphorus and Nitrogen /Dioctadecyl dimethyl ammonium chloride modified MMT (PN-IFR/m-MMT) | 30 | P:nP | 6.5:1 | 10 | 202 | 132 | 50 | — | 1.72 | 28.5 | — | [56] |
| Phosphorus and Nitrogen-based IFR: compound containing Phosphorus (22\%\) and Nitrogen (18\%) / Dioctadecyl dimethyl ammonium chloride modified MMT (PN-IFR/m-MMT) | P:nP | 5:1 | 19 | 202 | 133 | 50 | 3.25 | 25.5 | 56 | 35 | 50 | 3.25 | 25.5 | [56] |
| Phosphorus-based IFR: compound containing Phosphorus (22\%\) and Nitrogen (18\%) / Octadecyl trimethyl ammonium bromide modified MMT (P-IFR/m-MMT) | P:nP | 10:2:1 | 30 | 45 | 18 | 35 | 3 | 21.02 | 56 | 35 | 3 | 21.02 | 56 | [55] |
| Phosphorus-based IFR: compound containing Phosphorus (22\%\) and Nitrogen (18\%) / Sodium dodecyl sulfonate intercalated Ni-Al LDH (P-IFR/m-LDH) | P:nP | 10:2:1 | 32 | 55 | 16 | 35 | 3 | 19.97 | 56 | 35 | 3 | 19.97 | 56 | [55] |
| Phosphorus-based IFR: compound containing Phosphorus (22\%\) and Nitrogen (18\%) / A-POSS (P-IFR/A-POSS) | P:nP | 10:2:1 | 33 | 145 | 54 | 35 | 3 | 2.31 | 56 | 35 | 3 | 2.31 | 56 | [55] |
| Phosphorus-based IFR: compound containing Phosphorus (22\%\) and Nitrogen (18\%) / MWCNT (P-IFR/MWCNT) | P:nP | 17:6:1 | 33 | 225 | 73 | 35 | 3 | 1.10 | 56 | 35 | 3 | 1.10 | 56 | [54] |
| Material                        | Phosphorus-based IFR compound containing Phosphorus(22%) and Nitrogen(18%)/MWCNT (P-IFR/MWCNT) | Phosphorus-based IFR compound containing Phosphorus(22%) and Nitrogen(18%)/MWCNT (P-IFR/MWCNT) | Phosphoric acid/ethylenediamine (PA/EDA) | PPU-CA/APP | PPU-CA/APP | PPU-CA/APP | PPU-CA/APP | MP/PER | MP/PER | MP/PER | MP/PER | MP/PER/Organcially modified MMT (MP/PER/m-MMT) | MP/PER/Organcially modified MMT (MP/PER/m-MMT) | MMF/PER | MMF/PER/Lanthanum oxide (MMF/PER/La₂O₃) | MMF/PER/La₂O₃ | MMF/PER/La₂O₃ |
|--------------------------------|-------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------|----------|----------|----------|----------|--------|--------|--------|--------|--------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------|----------|-----------------------------------------------|-----------------------------------------------|-----------------------------------------------|
| MWCNT (P-IFR/MWCNT)           | 28 PnP 10.2:1                                                                                   | 28 PnP 7:1                                                                                      | 25 PnP 1.7:1                                                                            | 25 N/P 1:1 | 25 N/nN 2:1 | 25 N/nN 3:1 | 25 N/nN 4:1 | 25 N/nN 1.5:1 | 25 N/nN n 1.66:1.0: 11 | 25 N/nN n 1.87:1.0: 25 | 20 N/nN n 1.62:0.1 | 20 N/nN n 1.62:0.1 | 25 N/nN n 1.7:1 | 25 N/nN n 1.7:1 | 25 N/nN n 1.7:1 |
|                               | 33 145 54 35 3 2.31 — — [54]                                                                 | 31 140 79 35 3 1.53 — — [54]                                                                  | 24.7 1198.2 78.7 50 2.4 — — NR [139]                                                     | 12.8 263.1 57.1 50 2.4 3.25 — V-0 [139]                                                | 17.1 288 70 35 3 1.87 25.5 V-1 [50]                                              | 19.7 568 75 35 3 1.02 25.5 V-0 [50]                                              | 26.8 605 77 35 3 1.27 26.5 V-0 [50]                                              | 22.2 642 77 35 3 0.99 27 V-0 [50]                                              | — — 54 930 140 35 4 — — NR [64]                                             | — — 42 831 112 35 3 — — 18 NR [50]                                               | 25 380 212 50 6 2.17 29 V-2 [185]                                         | 24 265 206 50 6 3.07 34 V-0 [185]                                      |
|                               |                                                                                                 |                                                                                                 |                                                                                         | 28 250 99 35 4 2.72 — V-0 [64]                                                         | 35 305 110 35 4 2.51 — V-0 [64]                                                     | 37 400 114 35 4 1.95 — NR [64]                                                   | 42 1290 228 50 6 — — 18.1 NR [185]                                            | 25 380 212 50 6 2.17 29 V-2 [185]                                         | 24 265 206 50 6 3.07 34 V-0 [185]                                      |
|                               |                                                                                                 |                                                                                                 |                                                                                         | 25 380 212 50 6 2.17 29 V-2 [185]                                                        | 25 380 212 50 6 2.17 29 V-2 [185]                                                   | 25 380 212 50 6 2.17 29 V-2 [185]                                                   | 25 380 212 50 6 2.17 29 V-2 [185]                                                   | 25 380 212 50 6 2.17 29 V-2 [185]                                                   | — — 51 903 119.6 35 3 — — 17.5 NR [186]                                      | — — 51 903 119.6 35 3 — — 17.5 NR [186]                                      | 25 380 212 50 6 2.17 29 V-2 [185]                                         | 45 271 94.3 35 3 3.72 32 V-0 [186]                                        | 47 247 91.4 35 3 4.40 31.5 V-0 [186]                                       |
|                               |                                                                                                 |                                                                                                 |                                                                                         |                                                                                         |                                                                                         |                                                                                         |                                                                                         |                                                                                         |                                                                                         |                                                                                         |                                                                                         |                                                                                         | 50 221 85 35 3 5.63 31.5 V-0 [186]                                            |                                                                                         |                                                                                         |                                                                                         |
|                     |       | 38  | 1166 | 89.1 | 35  | 3  | —  | 17  | NR  | [187] |
|---------------------|-------|-----|------|------|-----|----|----|-----|-----|-------|
| MPP/DPER            | 30    | N:nN 3:1 | 45  | 427.6 | 79.4 | 35 | 3  | 3.62 | 28.7 | V-0   | [187] |
| MPP/EG/DPER         | 30 N:nN 1.5:1:0.5 | 20  | 218.2 | 68.7  | 35 | 3  | 3.64 | 33.2 | V-0   | [187] |
| Amino trimethylene phosphonic acid melamine salt/PER (MATMP/PER) | 25 N:nN 2:1 | 17  | 256.4 | 103.2 | 50 | 3  | 3.93 | 30.3 | V-0   | [187] |
|                    | —     | —   | 25   | 1229  | 123.6 | 50 | 3  | —   | 18.5 | NR    | [187] |
| MPyP/PER            | 25    | N:nN 3:1 | 32  | 343   | 136  | 35 | 4  | 2.13 | 29   | V-1   | [187] |
| MPyP/PER/Epoxy crosslinked β-cyclodextrin nanosponge (MPyP/PER/m-CD) | 25 N:nN 3:1:0.35 | 30  | 235  | 118   | 35 | 4  | 3.36 | 32.5 | V-0   | [187] |
|                    | —     | —   | 65   | 1417  | 128.5 | 35 | 3  | —   | —    | NR    | [66]  |
| MPyP/Phosphorus-based FR: Tri (1-oxo-2,6,7-trioxa-1-phosphabicyclo[2.2.2]octane-methyl) phosphate (MPyP/P-FR) | 30 N:P 1:1 | 40  | 175.2 | 90.6  | 35 | 3  | 7.05 | —    | V-0   | [66]  |
|                    | —     | —   | 34   | 1727  | 112   | 35 | 3  | —   | 17   | NR    | [66]  |
| MPyP/Triazine-based CFA: synthesized by reaction of cyanuric chloride and ethanalamine and ethylenediamine (MPyP/TA-CFA) | 30 N:N 3:1 | 12  | 431   | 84    | 35 | 3  | 1.88 | 29.5 | V-0   | [66]  |
|                    | —     | —   | 48   | 988   | 88.3  | 35 | 3  | —   | 17   | NR    | [39]  |
| Triazine-based CFA: synthesized by reaction of | 30 N:P 1:1 | 32  | 82.4  | 77.9  | 35 | 3  | 9.06 | 29   | V-0   | [39]  |
cyanuric chloride and piperazine/APP (TA-CFA/APP)

| Triazine-based CFA: synthesized by reaction of cyanuric chloride and piperazine/APP (TA-CFA/APP) | 30 | N:N 2:1 | 52 | 247 | 78.4 | 35 | 3 | 4.88 | 23 | V-1 | [39] |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |

| Triazine-based CA: synthesized by reaction of cyanuric chloride and piperazine/APP (TA-CA/APP) | 20 | N:P 1:1 | 38 | 83 | 41 | 35 | 3 | 4.73 | 30 | V-0 | [23] |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Triazine-based CA: synthesized by reaction of cyanuric chloride, 2,6,7-trioxa-1-phosphabicyclo[2.2.2]octane-4-methanol and piperazine/APP (TA-CA/APP) | 30 | N:N 2:1 | 36 | 456 | 96 | 35 | 3.2 | 2.47 | 25.5 | V-2 | [27] |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Triazine-based CA: synthesized by reaction of cyanuric chloride and 2,6,7-trioxa-1-phosphabicyclo[2.2.2]octane-4-methanol and diethylenetriamine/APP (TA-CA/APP) | 30 | N:P 1:1 | 32 | 166.8 | 108.2 | 35 | 3 | 3.58 | 28 | V-0 | [179] |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Triazine-based CA: synthesized by reaction of cyanuric chloride and N-aminoethylpiperazine/APP (TA-IFR/APP) | 25 | N:P 1:1 | 44 | 123 | 73.3 | 35 | 3.2 | 8.86 | 27.5 | V-0 | [31] |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Triazine-based IFR: synthesized by reaction of cyanuric chloride and N-aminoethylpiperazine/APP (TA-IFR/APP) | 25 | N:N 2:1 | 44 | 241 | 77.2 | 35 | 3.2 | 4.29 | 24.5 | V-1 | [31] |
| Reaction of cyanuric chloride and N-aminoethylpiperazine/APP (TA-IFR/APP) | — | 62 | 1221 | 265 | 35 | 6 | — | 19 | NR | [182] |
|---|---|---|---|---|---|---|---|---|---|---|
| Triazine-based IFR and APP (TAaandAPP-IFR) | 10 | N:P | 44 | 313 | 235 | 35 | 6 | 3.12 | 26 | NR | [182] |
| Triazine-based IFR and APP (TAaandAPP-IFR) | 15 | N:P | 45 | 148 | 191 | 35 | 6 | 8.30 | 29 | NR | [182] |
| Triazine-based IFR and APP (TAaandAPP-IFR) | 20 | N:P | 43 | 115 | 153 | 35 | 6 | 12.75 | 31 | V-0 | [182] |
| Piperazine-based IFR: Piperazine spirocyclic phosphoramidate/APP (PI-IFR/APP) | 30 | N:nN | 2:1 | 20 | 189.2 | 74.85 | 50 | 3 | 5.82 | 33.1 | V-1 | [70] |
| Nitrogen-based FR: compound containing nitrogen (27.5 wt.%) and Phosphorus (15.6 wt.%)/Fumed silica (NP-IFR/SiO₂) | 25 | N:nN | 49:1 | 25 | 124 | 35.1 | 35 | 4 | 19.30 | 38 | V-0 | [74] |
| Nitrogen-based FR: compound containing nitrogen (27.3 wt.%) and Phosphorus (15.6 wt.%)/Fumed silica (NP-IFR/SiO₂) | 25 | N:nN | 7:3:1 | 17 | 341 | 87.9 | 35 | 4 | 1.90 | 27 | NR | [74] |
| Nitrogen-based IFR: Poly (diallyldimethyl ammonium) and polyphosphate polyelectrolyte complexe/Polyamide-6 (N-IFR/PAn) | 25 | N:nN | 4:1 | 17 | 295.2 | 80.5 | 50 | 3 | 2.81 | 27.3 | V-1 | [75] |
| Nitrogen-based IFR: compound containing nitrogen (23%) and Phosphorus (21%)/Hollow glass microsphere (N-IFR/HGM) | 25 | N:nN | 49:1 | 16 | 93.8 | 74.4 | 50 | 3 | 7.15 | 34.5 | V-0 | [76] |
| Nitrogen (23%)- and Phosphorus (21%)-based intumescent | 25 | N:nN | 24:1 | 17 | 78.8 | 68 | 50 | 3 | 9.90 | 36.5 | V-0 | [76] |
| Flame retardant/Hollow glass microsphere (N-IFR/HGM) |  |
|---|---|
| Nitrogen-based IFR: compound containing nitrogen (23%) and Phosphorus (21%)/HGM (N-IFR/HGM) |  |
| | 25 | N:nN 11.5:1 | 12 | 61.6 | 74.2 | 50 | 3 | 8.19 | 35.5 | V-0 | [76] |  |
| Nitrogen-based IFR: compound containing nitrogen (23%) and Phosphorus (21%)/HGM (N-IFR/HGM) | 25 | N:nN 5.25:1 | 13 | 81.6 | 72.5 | 50 | 3 | 6.86 | 34.5 | V-0 | [76] |  |
| ATH/APP | 30 | M:P 1:1 | 40 | 210 | 75 | 35 | 3 | 1.55 | 24 | V-1 | [183] |  |
| 4,4′-diphenylmethane diisocyanate and melamine co-microencapsulated ATH and APP (mc-(ATHandAPP)) | 30 | M:P 1:1 | 75 | 120 | 53 | 35 | 3 | 7.20 | 25.5 | V-0 | [183] |  |
| ATH/Glass Bubble (ATH/GB) | 60 | M:M 11:1 | 31 | 212 | 53 | 50 | 4 | 22.17 | 25 | — | [81] |  |
| ATH/GB | 60 | M:M 5:1 | 36 | 190 | 49 | 50 | 4 | 31.08 | 23.4 | — | [81] |  |
| ATH/GB/Octadecylamine modified ZrP (ATH/GB/m-ZrP) | 60 | M:M:M 4.7:1:0.3 | 24 | 136 | 90 | 50 | 4 | 15.76 | 24 | — | [81] |  |
| ATH/GB/Octadecylamine modified ZrP (ATH/GB/m-ZrP) | 60 | M:M:M 4.4:1:0.6 | 24 | 152 | 91 | 50 | 4 | 13.94 | 23.2 | — | [81] |  |
| ATH/GB/Octadecylamine modified ZrP (ATH/GB/m-ZrP) | 60 | M:M:M 4.1:1:0.9 | 21 | 189 | 98 | 50 | 4 | 9.11 | 22.8 | — | [81] |  |
| ATH/Cetyltrimethyl ammonium bromide modified Fe MMT (ATH/m-MMT) | 50 | M:M 49:1 | 48 | 482 | 95.1 | 50 | 3 | 4.89 | 25.5 | NR | [80] |  |
| ATH/Cetyltrimethyl ammonium bromide modified Fe MMT (ATH/m-MMT) | 50 | M:M 15.6:1 | 49 | 412 | 90.9 | 50 | 3 | 6.11 | 27.4 | V-1 | [80] |  |
| ATH/Cetyltrimethyl ammonium bromide modified Fe MMT (ATH/m-MMT) | 50 | M:M 9:1 | 53 | 329 | 89 | 50 | 3 | 8.46 | 29 | V-0 | [80] |  |
| — — — | 26 | 1967 | 112 | 50 | 3 | — | — | — | [82] |  |
| ATH/Styrene-co-vinylbenzyl chloride modified MMT (ATH/m-MMT) | 23 | M:M | 6:6:1 | 21 | 677 | 84 | 50 | 3 | 3.12 | — | — | [82] |
|-------------------------------------------------------------|--------------------------|---------|---------|----------|----------------|---------|----|----|-------|----|---|-----------------|
| ATH/Styrene-co-vinylbenzyl chloride modified MMT (ATH/m-MMT) | 30 | M:M | 2:1 | 20 | 592 | 77 | 50 | 3 | 3.71 | — | — | [82] |
| ATH/Styrene-co-vinylbenzyl chloride modified MMT (ATH/m-MMT) | 37 | M:M | 1:17:1 | 18 | 536 | 74 | 50 | 3 | 3.84 | — | — | [82] |
| --- | — | — | — | 24 | 687 | 119 | 25 | 3 | — | 17.8 | — | [146] |
| MDH/APP/PER/MEL | 44.2 | MnM: | MnMnM | 1:3:1:0.6 | 0.56 | 43 | 121 | 58.2 | 25 | 3 | 20.79 | 28 | — | [146] |
| MDH/APP/PER/MEL | 50 | MnM: | MnMnM | 2:1:1:0.6 | 0.56 | 43 | 121 | 54.5 | 25 | 3 | 22.21 | 28.8 | — | [146] |
| MDH/APP/PER/MEL | 54.4 | MnM: | MnMnM | 3:1:0:60 | 0.56 | 44 | 104 | 53.6 | 25 | 3 | 26.88 | 30.2 | — | [146] |
| --- | — | — | — | 166 | 412 | 105 | 50 | 3 | — | 17.8 | — | [146] |
| MDH/APP/PER/MEL | 44.2 | MnM: | MnMnM | 1:3:1:0.6 | 0.56 | 217 | 68.8 | 39.3 | 50 | 3 | 20.91 | 28 | — | [146] |
| MDH/APP/PER/MEL | 50 | MnM: | MnMnM | 2:1:1:0.6 | 0.56 | 220 | 57.3 | 36.4 | 50 | 3 | 27.48 | 28.8 | — | [146] |
| MDH/APP/PER/MEL | 54.4 | MnM: | MnMnM | 3:1:0:60 | 0.56 | 232 | 54.3 | 31.2 | 50 | 3 | 35.68 | 30.2 | — | [146] |
| --- | — | — | — | 26 | 1967 | 112 | 50 | 3 | — | — | — | — | [82] |
| MDH/Styrene-co-vinylbenzyl chloride modified MMT (MDH/m-MMT) | 37 | M:M | 1:17:1 | 24 | 476 | 70 | 50 | 3 | 6.10 | — | — | [82] |
| --- | — | — | — | 30 | 1684 | 89 | 50 | 3 | — | — | — | — | [82] |
| MDH/Styrene-co-vinylbenzyl chloride modified MMT (MDH/m-MMT) | 40 | M:M | 3:1 | 24 | 471 | 80 | 50 | 3 | 3.18 | — | — | [82] |
| MDH/Styrene-co-vinylbenzyl chloride modified MMT (MDH/m-MMT) | 50 | M:M | 4:1 | 23 | 385 | 69 | 50 | 3 | 4.32 | — | — | [82] |
| MDH/Styrene-co-vinylbenzyl chloride modified MMT (MDH/m-MMT) | 60 | M:M | 5:1 | 22 | 304 | 59 | 50 | 3 | 6.12 | — | — | [82] |
| --- | — | — | — | 38 | 1425 | 121.4 | 50 | 3 | — | 17.5 | NR | [85] |
| MDH/Cetyltrimethyl ammonium bromide modified Fe-MMT (MDH/m-MMT) | 40 | MnM: | MnMnM | 39:1 | 52 | 422 | 98.1 | 50 | 3 | 5.71 | 24.9 | NR | [85] |
| MDH/Cetyltrimethyl ammonium bromide modified Fe-MMT (MDH/m-MMT) | 40 | M:M | 12.3:1 | 56 | 378 | 97.5 | 50 | 3 | 6.91 | 26.5 | NR | [85] |
| Modified MMT | Modification | Sample Code | M:M | M1 | M2 | M3 | Yield (%) | Tg (°C) | Char. (10%) | NR | Source |
|-------------|--------------|-------------|-----|----|----|----|-----------|--------|-----------|-----|---------|
| MDH/Cetyltrimethylammonium bromide modified Fe MMT (MDH/m-MMT) | 40 | M:M | 7:1 | 63 | 329 | 87.9 | 50 | 3 | 9.91 | 28.1 | V-1 | [85] |
| MDH/Cetyltrimethylammonium bromide modified Fe MMT (MDH/m-MMT) | — | — | 71 | 2283 | 218 | 35 | 1 | — | — | — | [84] |
| MDH/L,4-bis (acrylamido) diphenylsulfone crosslinked N-(4-methyl phenyl) acrylamide monomer (MDH/Cobalt chelate) | 50 | M:M | 9:1 | 72 | 619 | 306 | 35 | 1 | 2.66 | — | — | [84] |
| MDH/Cobalt chelate | 50 | M:M | 4:1 | 63 | 618 | 277 | 35 | 1 | 2.57 | — | — | [84] |
| MDH/Cobalt chelate | 50 | M:M | 2.3:1 | 53 | 776 | 236 | 35 | 1 | 2.02 | — | — | [84] |
| MDH/Cobalt chelate | 50 | M:M | 1.5:1 | 56 | 780 | 222 | 35 | 1 | 2.26 | — | — | [84] |
| Cetyltrimethylammonium bromide modified MMT/SEP (m-MMT/SEP) | 5 | M:M | 1:1 | 62 | 417 | 63.7 | 50 | 3 | 2.78 | — | — | [87] |
| MDH/Organically treated SEP (MDH/m-SEP) | 15 | M:M | 2:1 | 29 | 325 | 62.1 | 50 | 3 | 1.71 | — | — | [87] |
| MDH/Organically treated SEP (MDH/m-SEP) | 20 | M:M | 3:1 | 26 | 205 | 53.5 | 50 | 3 | 2.82 | — | — | [87] |
| MDH/cetyltrimethylammonium bromide modified MMT/SEP (MDH/m-MMT/SEP) | 15 | M:M:M | 4:1:1 | 54 | 246 | 56.3 | 50 | 3 | 4.65 | — | — | [87] |
| MDH/cetyltrimethylammonium bromide modified MMT/SEP (MDH/m-MMT/SEP) | 20 | M:M:M | 4:1:1 | 50 | 209 | 50.1 | 50 | 3 | 5.69 | — | — | [87] |
| MMT/APP | 10 | M:nM | 1.5:1 | 28 | 764 | 64 | 35 | 0.4 | 4.78 | — | — | [24] |
| MMT/APP | 10 | M:nM | 4:1 | 29 | 751 | 62 | 35 | 0.4 | 5.20 | — | — | [24] |
| Modified MMT/APP (m-MMT/APP) | 10 | M:nM | 1.5:1 | 30 | 599 | 57 | 35 | 0.4 | 7.34 | — | — | [24] |
| Modified MMT/APP (m-MMT/APP) | 10 | M:nM | 4:1 | 31 | 575 | 56 | 35 | 0.4 | 8.04 | — | — | [24] |
| Polysiloxane based FR/APP (Si-FR/APP) | 25 | M:P | 1:1 | 14 | 277 | 97 | 50 | — | 3.00 | 28.9 | V-0 | [30] |
| Polysiloxane based FR/APP (Si-FR/APP) | — | — | 25 | 981 | 147 | 50 | — | — | 17.6 | NR | — | [30] |
| NiO/AC | 15 | M:C | 1:1 | 18 | 385 | 132 | 50 | 6 | 2.88 | — | — | [108] |
| NiO/AC | — | — | 47 | 1933 | 176 | 50 | 5 | — | — | — | [189] |
| System                          | M:nM   | 5:1  | 10:1 | 12:1 | 5:1  | 10:1 | 5:1  | 10:1 | 5:1  | 10:1 | 5:1  | 10:1 |
|--------------------------------|--------|------|------|------|------|------|------|------|------|------|------|------|
| SEP/MWCNT                      | 12     | 32   | 355  | 241  | 50   | 5    | 2.70 | —    | —    | —    | —    | —    |
| Silicon/Stannous chloride      | 5      | 91   | 860.1| 193.7| 35   | —    | 4.03 | —    | —    | —    | —    | —    |
| C2O2/CO                       | 5.5    | 83   | 458.9| 78.1 | 35   | 3    | 1.07 | 20   | —    | —    | —    | —    |
| Ethylene glycol methacrylate phosphate modified C2O2/CO | 5.5    | 78   | 498.2| 75.2 | 35   | 3    | 0.96 | 19   | —    | —    | —    | —    |
| Ethylene glycol methacrylate phosphate modified C2O2/CO | 10.5   | 61   | 424.7| 74.8 | 35   | 3    | 0.88 | 20   | —    | —    | —    | —    |
| Silicon/Stannous chloride      | 5      | 25   | 364  | 194  | 50   | 6    | 2.55 | 25.8 | —    | —    | —    | —    |
| CF/MWCNT                      | 10     | 27   | 353  | 185  | 50   | 6    | 3.61 | 26.5 | —    | —    | —    | —    |
| CB/MWCNT                      | 6      | 26   | 402  | 187  | 50   | 6    | 3.02 | 23.8 | —    | —    | —    | —    |
| CB/MWCNT                      | 4      | 25   | 314  | 180  | 50   | 6    | 3.86 | 27.6 | —    | —    | —    | —    |
| CB/MWCNT                      | 8      | 27   | 361  | 166  | 50   | 6    | 3.08 | 25.7 | —    | —    | —    | —    |
| AC/CoO                       | 15     | 18   | 385  | 132  | 50   | 6    | 2.88 | —    | —    | —    | —    | —    |
| AC/CoO                       | 25.3   | 166  | 412  | 105  | 25   | 3    | —    | —    | —    | —    | —    | —    |
| PER/MEL/APP                    | 25.3   | 170  | 140  | 61.1 | 25   | 3    | 5.17 | —    | —    | —    | —    | —    |
| PER/MEL/APP                    | 25.3   | 32   | 198  | 79   | 50   | 3    | 6.96 | —    | —    | —    | —    | —    |
| Cycloextrin nanosponge/Triethylphosphosphate (CD/TEP) | 10     | 30   | 1529 | 93   | 35   | 3    | 0.63 | —    | —    | —    | —    | —    |
| CD/TEP                        | 15     | 26   | 839  | 90   | 35   | 3    | 1.03 | —    | —    | —    | —    | —    |
| CD/APP                        | 15     | 24   | 910  | 89   | 35   | 3    | 0.89 | —    | —    | —    | —    | —    |
| Phosphorus and Nitrogen elements modified lignin/Nickel acetate (m-lig/Ni(Ac)) | 20     | 31   | 330  | 69.5 | 35   | 3    | 3.25 | 26   | —    | —    | —    | —    |
| Phosphorus and Nitrogen elements modified lignin/Cobalt acetate (m-lig/Co(Ac)) | 20     | 37   | 362  | 72.8 | 35   | 3    | 3.37 | 24.5 | —    | —    | —    | —    |
Figure 15 displays the performance of different combinatorial additive systems used for PP. It can be clearly observed from the left-hand side figure that cases with FRI values above 10 (Excellent zone) are more frequent compared to all previous cases in which only one additive was used. More interestingly, the combination of additives appeared a useful strategy where very high FRI values (event more than 50) took place at intermediate loadings (25–30 wt.%). For achieving a high FRI value, the combination of several types of flame retardants is needed, for example, phosphorus, intumescent, and mineral flame retardants [150] or phosphorus, nitrogen, and mineral flame retardants [164].

**Figure 15.** FRI values as a function of combinatorial FR additives and their content in PP in long-shot (left-hand figure) and close-up (right-hand figure) views. Symbols are indicative of different types of combinatorial flame retardant used. Here: ▲ APP-13.2/PER-6.8 [68], ▼ APP-16.7/PER-8.3 [143], ▲ APP-18.7/PER-6.3 [67], ▶ APP-22.5/PER-7.5, mc-(APP-22.5&PER-7.5) [144], ♦ APP-10.5/PER-9.8/MEL-9.1, APP-15.3/PER-9.3/MEL-8.8, APP-19.1/PER-8.9/MEL-8.2 [145], ● APP-10.5/PER-9.8/MEL-9.1, APP-15.3/PER-9.3/MEL-8.8, APP-19.1/PER-8.9/MEL-8.2 [145], ▲ APP-15.3/PER-9.3/MEL-8.6, APP-14.3/PER-8.7/MEL-8.1/MDH-6.2 [146], ▼ APP-15.3/PER-9.3/MEL-8.6, APP-14.3/PER-8.7/MEL-8.1/MDH-6.2 [146], ▲ APP-12/PER-4/MEL-4/C20A-1, APP-12/PER-4/MEL-4/C20A-3, APP&MMT-12/PER-4/MEL-4/C20A-1, APP&MMT-12/PER-4/MEL-4/C20A-3 [104], ♦ m-APP-16.6/DPER-4.2/MEL-4.2, m-APP-16/DPER-4/MEL-4/SEP-1 [147], ♦ APP-13.5/PER-4.5, APP-12.75/PER-4.25/MF-1, APP-12.75/PER-4.25/MFA-1 [148], ● APP-22.5/PER-7.5 [149], ▲ APP-12.7/PER-5.3, APP-12/PER-5/T-A-FR-1 [150], ▼ APP-16.67/PER-8.33, APP-16.33/PER-8.17/NOR116-0.5 [72], ▲ APP-13.5/PER-4.5, APP-12.75/PER-4.25/G-bases-1, APP-12.75/PER-4.25/U-bases-1 [151], ▲ APP-17.2/DPER-7.8, m-APP-17.2/DPER-7.8, APP-16.2/DPER-7.8/ATH-1 [152], ▲ APP-21.4/PER-7.1, APP-20.3/PER-6.8/Kaol-1.4 [70], ▲ APP-21.75/PER-7.25, APP-19.5/PER-6.5/MMT-3, APP-19.5/PER-6.5/m-MMT-3 [153], ▲ APP-21.75/PER-7.25, APP-19.5/PER-6.5/MMT-3, APP-19.5/PER-6.5/m-MMT-3, APP-19.5/PER-6.5/m-MMT-3 [154], ▼ APP-18.75/PER-6.25, APP-18-18/PER-6/LDH-1, APP-18/PER-6/m-LDH-1 [155], ▽ APP-18.75/PER-6.25, APP-18/PER-6/m-SiR-1, APP-18/PER-6/m-SiR-1, APP-16.5/PER-5.5/m-SiR-3 [156], ▲ APP-16.67/DPER-8.3, m-APP-16.7/DPER-8.3/Z-1, m-APP-16.7/DPER-8.3/Z-1/MWCNT-0.1 [157], ▽ APP-18.75/PER-6.25, APP-18.75/PER-6.25/ALL-2 [158], ▲ APP-16.7/PER-8.3, APP-15/PER-7.5/MATO-2.5, APP-15/PER-7.5/Zn-MAO-2.5 [159], ▲ APP-16.7/PER-8.3, APP-16/PER-8/m-SEP-1, APP-15.3/PER-7.7/m-SEP-2, APP-14.7/PER-7.3/m-SEP-3, APP-14/PER-7/m-SEP-4, APP-13.3/PER-6.7/m-SEP-5 [160], ▽ APP-15/PER-5, APP-14.25/PER-4.75/OP-POSS-1, APP-14.25/PER-4.75/A-POSS-1, APP-14.25/PER-4.75/OA-POSS-1, APP-14.25/PER-4.75/T5-POSS-1 [161], ▲ APP-20/PER-10, APP-19/PER-9.5/T-RE-5, APP-19/PER-9.5/CV-5, APP-19/PER-9.5/CR-5 [162], ▲ APP-15/PER-5, APP-14.25/PER-4.75/ZnB-1, APP-14.25/PER-4.75/BPO-1, APP-14.25/PER-4.75/Bsi-1, APP-14.25/PER-4.75/LaB-1 [163], ▲ APP-
15/TA-CFA-15, TA-CFA-22.5/MPyP-7.5 [66], ○ TA-CFA-15/APP-15, TA-CFA-20/APP-10 [39], ○ TA-CA-10/APP-10 [23], ○ TA-CA-13.33/APP-6.67 [27], ○ TA-CA-20/APP-10, TA-CA-15/APP-15 [179], ○ TA-IFR-12.5/APP-12.5, TA-IFR-16.67/APP-8.33 [31], ○ TA&APP-IFR-10, TA&APP-IFR-20 [182], ○ PI-IFR-20/APP-10 [70], ○ N-IFR-24.5/SiO2-0.5, N-IFR-22/SiO2-3 [74], ○ N-IFR-20/PA6-5 [75], ○ N-IFR-24.5/HGM-0.5, N-IFR-24/HGM-1, N-IFR-23/HGM-2, N-IFR-22/HGM-3 [76], □ ATH-15/APP-15, mc-(ATH-15&APP-15) [183], □ ATH-55/GB-5, ATH-50/GB-10, ATH-47/GB-10/m-ZrP-3, ATH-44/GB-10/m-ZrP-6, ATH-41/GB-10/m-ZrP-9 [81], □ ATH-49/m-MMT-1, ATH-47/m-MMT-3, ATH-45/m-MMT-5 [80], □ ATH-20/m-MMT-3, ATH-20/m-MMT-10, ATH-20/m-MMT-17 [82], □ MDH-16.6/APP-12.7/PER-7.7/MEL-7.2, MDH-25/APP-11.5/PER-7/MEL-6.5, MDH-31.8/APP-10.4/PER-6.3/MEL-5.9 [146], □ MDH-16.6/APP-12.7/PER-7.7/MEL-7.2, MDH-25/APP-11.5/PER-7/MEL-6.5, MDH-31.8/APP-10.4/PER-6.3/MEL-5.9 [146], □ MDH-20/m-MMT-17 [82], □ MDH-30/m-MMT-10, MDH-40/m-MMT-10, MDH-50/m-MMT-10 [82], □ MDH-39/m-MMT-1, MDH-37/m-MMT-3, MDH-35/m-MMT-5 [85], □ MDH-45/Cobalt chelate-5, MDH-40/Cobalt chelate-10, MDH-35/Cobalt chelate-15, MDH-30/Cobalt chelate-20 [84], □ m-MMT-2.5/SEP-2.5, MDH-10/m-SEP-5, MDH-15/m-SEP-5, MDH-10/m-MMT-2.5/SEP-2.5, MDH-15/m-MMT-2.5/SEP-2.5 [87], □ MMT-6/APP-4, MMT-8/APP-2, m-MMT-6/APP-4, m-MMT-8/APP-2 [24], □ Si-FR-12.5/APP-12.5 [30], □ NiO-7.5/AC-7.5 [108], □ SEP-10/MWCNT-2 [189], □ C30B/3-ACBP-3, C30B-3/BUPB-3, C30B-3/MEPB-3, C30B-3/PBPA-3 [191], □ Si-3/SnCl2-2 [190], □ C20A-5/TiO2-0.5, m-C20A-5/TiO2-0.5, m-C20A-10/TiO2-0.5 [105], □ CF-5/MWCNT-5 [133], □ CB-3/MWCNT-1, CB-5/MWCNT-1, CB-5/MWCNT-3 [131], □ CB-5/C-5 [134], □ AC-7.5/NiO-7.5 [108], □ PER-10.4/MEL-9.7/APP-5.2 [145], □ PER-10.4/MEL-9.7/APP-5.2 [145], □ CD-7/TEP-3, CD-10/TEP-5, CD-7.5/APP-7.5 [135], □ m-lig-18/Ni(Ac)=2, m-lig-18/Co- (Ac)=2, m-lig-18/Zn(Ac)=2 [136].

Figure 16 shows that V-0 level in UL-94 is automatically obtained in the case of combined flame retardant systems used in PP regardless of the FRI value. However, no correlation exists between the FRI and LOI (Figure 17). The complexity of polymer–filler interaction can be considered as the main reason for diversity of properties.

![Figure 16. FRI values versus UL-94 test results. Symbols are indicative of combination of flame retardant (FR) additives used in PP. The vertical intervals in each category, i.e., V-0, V-1, V-2, and NR, are schematically representative of the amount of additive used. For example, two data distinguished by different symbols having the same or very close FRI values (horizontal quantity) in a given category (e.g., V-1), may have different vertical quantities, e.g., both reveal V-1 behavior in UL-94 test, but the upper contains more FR in PP.](image-url)
Figure 17. FRI values of PP as a function of LOI test results in long-shot (left-hand figure) and close-up (right-hand figure). Symbols are indicative of different types of blend flame retardants used. The left-side plot reveals that FRI values above 10 (Excellent zone) took place in several cases, which is in contradiction with all previous cases in which only one additive was used.

7. Conclusions and Future Perspective

This work opens new avenues to the experts working on “flame retardant polyolefins”, the title of a Special Issue entitled “Flame Retardant Polyolefins” in Polymers journal for which this work is designed and carried out. In this work, more than 150 research papers from the literature dealing with the flame retardancy of PP were analyzed, classified, and discussed in terms of flame retardancy performance. From the selected papers were extracted cone calorimetry data to calculate Flame Retardancy Index (FRI) as a measure or label of flame retardant performance. To have a comprehensive overview of flame retardant PP materials, works on PP flame retardancy were categorized in terms of additives used in classes including: phosphorus-based, nitrogen-based, mineral, carbon-based, bio-based, and hybrid combinatorial flame retardants composed of two or more additives. The analysis of efficiency of flame retardancy was performed in terms of the FRI variation as a function of wt.% of additives used. The analysis quite obviously unveiled the superiority of the combination of additives over the use of each one separately. In addition, the UL-94 and LOI values available in each class of additives were plotted in terms of the FRI so as to find possible correlation between analyses made in the literature. This work provided a pool of data on flame-retardant PP materials for future research on PP materials. It was elucidated that FRI can satisfactorily make possible classification of PP materials in terms of flame retardancy performance. The present work provides those research works that claim achieving synergistic effect of two or more flame retardants with a clear measure of flame retardant performance as Poor, Good, and Excellent labels assigned to PP materials, based on cone calorimetry data. Moreover, future works on LOI and UL-94 tests can be added to the data used here so as to draw a more detailed picture of flame retardancy behavior of PP materials. The approach can be used to make judgement about other flame retardant polyolefins. Moreover, we believe that the mechanical properties of FR polymers should also be considered in the future, but it is pertinent to the completeness of data in the literature. The importance of mechanical properties springs from the fact that highly loaded systems are prone to mechanical failure as a consequence of stress concentration. All in all, the type and the percentage of FRs in polymers affect both the mechanical and flame retardant properties of polymers; therefore, optimization of both properties is of importance.

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