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### To cite this version:

Yajie LI, Haicun YANG, Azita AHMADI-SENICHAULT, Aziz OMARI, Hongting PU - A thermal resistant and flame retardant separator reinforced by attapulgite for lithium-ion batteries via multilayer coextrusion - Polymer - Vol. 253, p.125027 - 2022



# A thermal resistant and flame retardant separator reinforced by attapulgite for lithium-ion batteries via multilayer coextrusion

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### ABSTRACT

Keywords: Attapulgite Multilayer separators Lithium-ion battery Thermal stability Flame retardancy Multilayer separators are widely used due to their wide shutdown window by combining lower melting temperature and higher melting temperature of different layers. With the development of high power lithium-ion batteries, multilayer separators equipped with effective thermal stability and flame retardancy are highly required. Herein, the poly(methyl methacrylate) modified attapulgite (ATPM) is selected as the heat resistant reinforcing component and blended with polypropylene (PP)/polyethylene (PE) respectively. Then we prepare PP(ATPM)/PE(ATPM) separators via multilayer coextrusion efficiently without multiple stretching processes, which can avoid serious separator shrinkage at elevated temperature. The intertwined ATPM could not only enhance the separator integrity, but also produce water vapor and oxide anti-flaming isolation layers at high temperatures. The as-prepared separators, referred to as MC-TIPS PP/PE/ATPM, exhibit higher thermal stability (with negligible dimensional shrinkage up to 180 °C), better flame retardancy and wider shutdown temperature window (124–183 °C) than the commercial multilayer separators. Moreover, the introduction of ester and hydroxyl groups could improve the wettability and electrolyte uptake of the separators. These properties, as well as the potential for large-scale production of multilayer coextrusion, make MC-TIPS PP/PE/ATPM an ideal choice for high-power battery separators.

### 1. Introduction

During the past decades, high power lithium-ion battery (LIB) has been vigorously developed, with applications expanding from portable electronic devices, electric vehicles to energy storage systems, which brings forward higher request for the safety performance of LIB [1]. Once LIBs are subjected to extreme conditions such as short-circuiting or overcharging [2], the exothermic chemical reactions will be initiated, which will raise internal pressure and release heat rapidly, eventually lead to thermal runaway [3,4]. Functional LIB separators or solid/gel electrolytes, which act as physical barriers between electrodes and provide pathway for lithium ions migration, have been considered to be important self-activating protection components [5,6] to prevent LIBs from thermal runaway. For example, thermotolerant biopolymer-based electrolytes [7], flame-resistant separator [8,9], thermotolerant separator [9], flame-resistant solid state organic electrolytes with superior

thermal stability [10], hybrid ceramic-polymer/ionic liquid electrolytes with high thermal stability as well as excellent Li ion-conductivity [11–16], etc. Especially, the application of polypropylene (PP)/polyethylene (PE) multilayer separators with shutdown function is the widely used strategy to protect LIBs safety due to the low cost and rich resources. Once the temperature rises up to the melting temperature of PE, the PE layer melts to close off pores, prevents ionic conduction and terminates electrochemical reaction [17]. Meanwhile, the PP layer with higher melting temperature can act as physical barrier between electrodes. However, they are flammable and their shrinkage is serious at high temperatures due to the residual stress caused by the multiple stretching process [18,19].

In order to improve the thermal stability and flame retardancy of polyolefin separators, many methods have been developed [20,21]. Separators coated with high temperature resistant organic polymers or flame-retardant additives [22], such as cellulose nanofiber [23],

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