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Short communication

Non-flammable organic liquid electrolyte for high-safety and high-energy density Li-ion batteries

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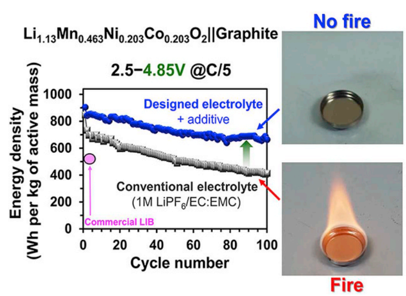
HIGHLIGHTS

- Rational design of non-flammable carbonate-based organic liquid electrolyte.
- High energy density of non-flammable full-cell with Li-rich layered oxide cathode.
- Preserved cathode structure and stable interface with non-flammable electrolyte.

GRAPHICAL ABSTRACT



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ABSTRACT

With increased energy density of rechargeable lithium-ion batteries for powering smart phones and electric vehicles and for their long range use, battery safety becomes more important than ever. This aspect motivated us to develop non-flammable liquid electrolyte that removes the risk of battery fire and explosion, which is urgently needed. Battery energy density and performance however should not be sacrificed to achieve just the safety. Here we report for the first time a rational design of non-flammable carbonate-based organic liquid electrolyte to satisfy safety, energy density and performance simultaneously. Our novel electrolyte, composed of 1 M lithium hexafluorophosphate salt and propylene carbonate and fluorinated linear carbonate co-solvents, at unmeasurable flash point does not fire representing non-flammable safe batteries but permits high-voltage stability to enable high-voltage charge of lithium-rich layered oxide cathode up to 5.0 V, high-energy density of 856 Wh per kg of cathode active mass and stable charge-discharge cycling performance of full-cell with graphite anode, in contrast to rapid performance fade of flammable conventional electrolyte system. The discovery of non-flammable carbonate-based organic liquid electrolyte opens up a new avenue to high-safety and high energy-density lithium-ion batteries for electric vehicles and advanced energy-storage applications.

1. Introduction

The global trend toward high-energy density and high-safety lithium (Li)-ion batteries for long-use smart phones and portable electronics and long-range electric vehicles (EVs) has driven battery R&D to find ways to improve current battery chemistries [1]. Commercial batteries fabricated with $LiCoO_2$ cathode and graphite anode, which are charge-discharge cycled in the voltage window of 3.0–4.2 V with non-aqueous carbonate-based organic liquid electrolyte of 1 M $LiPF_6$/ethylene carbonate (EC): ethyl methyl carbonate (EMC), are intrinsically

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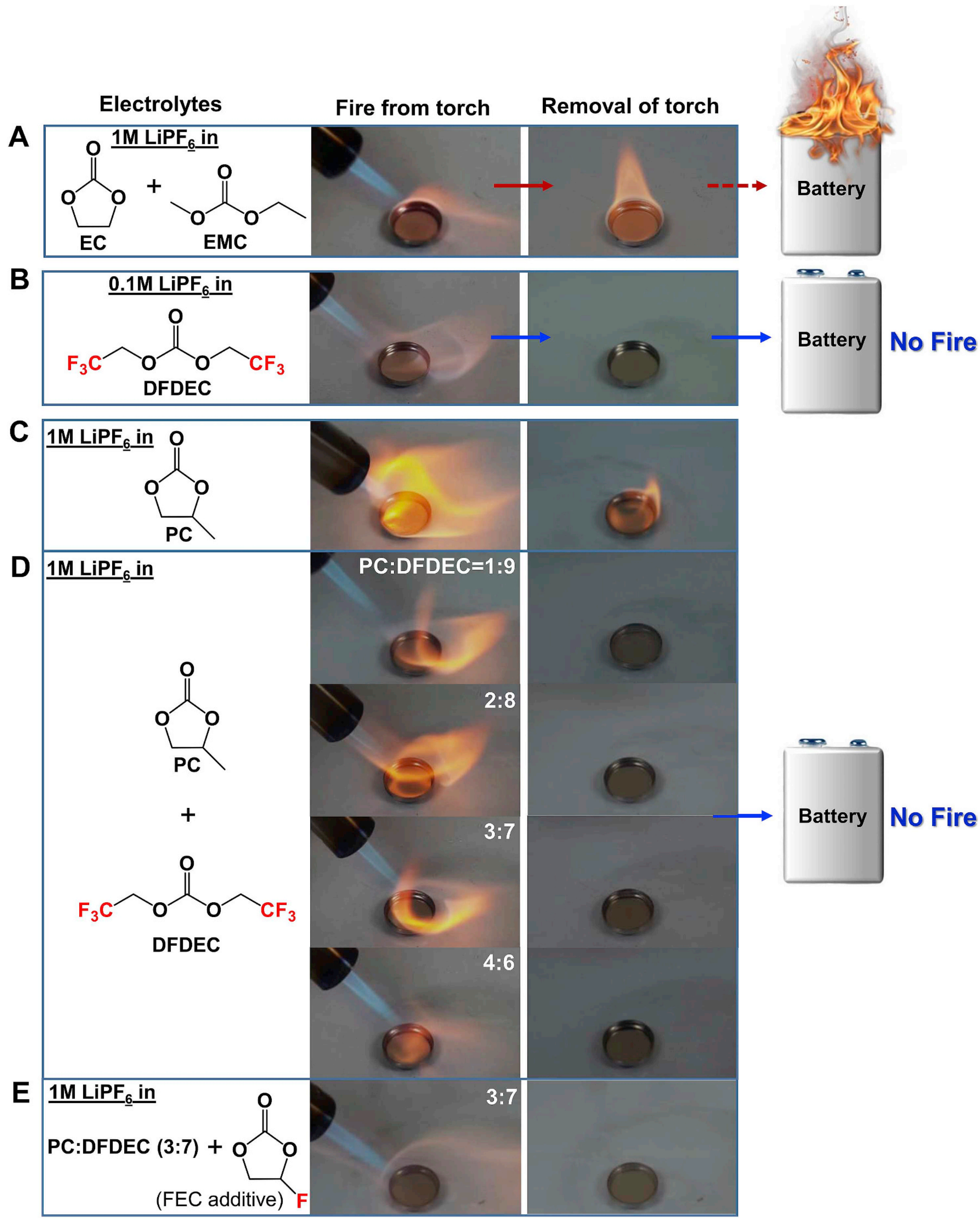

Fig. 1. Fire test results for (A) conventional electrolyte of 1 M $LiPF_6$/EC:EMC (3:7 vol ratio), (B) 0.1 M $LiPF_6$/DFDEC, (C) 1 M $LiPF_6$/PC and (D) designed electrolytes of 1 M $LiPF_6$/PC:DFDEC at volume ratios of 1:9, 2:8, 3:7 and 4:6, and (E) 1 M $LiPF_6$/PC:DFDEC (3:7) with 1 wt% FEC additive.

unsafe and always possess the risk of fire and explosion because of flammable conventional electrolyte (Fig. 1A) at the flash point of 28 °C (Fig. 2A) [2]. Organic solvents are fuels of combustion, particularly when a battery is under high current, overcharge, overheating or damaged, and when oxygen gases are evolved from metal oxide cathode. This situation is subject to thermal runaway when heat is generated faster than it can be dissipated, leading to the cell bursting and ignition on contact with oxygen in the air, and finally fire and explosion (Fig. 1A). With increased energy-density of Li-ion batteries, such dangerous event would be intensified and be a serious threat particularly to large-scale applications like EVs that contain thousands of batteries.
The safety issues motivated researchers the development of flame-retardant fluorinated electrolyte components [3,4], non-flammable inorganic (e.g. phosphate) solvents [5–7], less-flammable non-aqueous liquid electrolyte components [8,9], solid electrolyte-based all-solid-state batteries [10,11] or aqueous battery systems [12]. Battery safety issues however should be addressed without sacrificing battery energy-density and performance. To this end, the development of non-flammable carbonate-based liquid electrolyte is a promising approach and urgently needed, which holds similar battery chemistry to conventional electrolyte. Here we demonstrate our discovery of non-flammable carbonate-based liquid electrolyte of 1 M $LiPF_6$ salt in propylene carbonate