While the development of advanced electrode materials and structures is leading Li ion battery technology these days, the development of outstanding separators also improves the cell performance remarkably. Inspired by mussels’ exceptional wet-resistant holdfast onto versatile substrates, in this study, we coated polyethylene (PE) separators with mussel inspired polydopamine layers. After the polydopamine coating, PE separators become hydrophilic and thus bring various advantages in the cell operations: 1) the enhancement of power performance, 2) the adaptability with a wider range of electrolyte, particularly polar solvent for low temperature operations, 3) homogeneous Li ion flux that makes the cell operation more stable, especially for larger-scale cells and Li metal anodes. 4) the improved resistance against thermal shrinkage. While bringing a number of advantages, the coating process does not sacrifice original advantages of PE separators at all. This study suggests that mussel-inspired materials can contribute significantly to the improvement of emerging energy storage technology.