

LENRs are potentially another mechanism for producing so-called field failures that can trigger catastrophic thermal runaways in Lithium-based batteries

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Please see the following technical documents:

1. “Batteries for Sustainability – Selected Entries from the Encyclopedia of Sustainability in Science and Technology”

Ralph J. Brodd, *Editor*

Springer ISBN 978-1-4614-5791-6 (eBook)

Chapter 9 by B. Barnett et al., “*Lithium-ion Batteries, Safety*” **[25 pages of annotated quotes attached]**

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<http://www.amazon.com/Batteries-for-Sustainability-ebook/dp/B00APXDLXA>

2. LENRs in Lithium-ion batteries [68 slides]

Lewis Larsen, Lattice Energy LLC

July 16, 2010

Source URL: <http://www.slideshare.net/lewisglarsen/cfakepathlattice-energy-llc-len-rs-in-liion-battery-firesjuly-16-2010>

3. Evanescent localized superconductivity in LENR ‘patches’ [92 slides]

Lewis Larsen, Lattice Energy LLC

August 23, 2012

Source URL: <http://www.slideshare.net/lewisglarsen/lattice-energy-llc-hightemperature-superconductivity-in-patchesaug-23-2012>

4. Index to concepts and documents about Widom-Larsen theory, LENRs, and Lattice Energy [63 slides]

Lewis Larsen, Lattice Energy LLC

November 21, 2012

Source URL: <http://www.slideshare.net/lewisglarsen/lattice-energy-llcindex-to-documents-re-widomlarsen-theory-of-lenrsnov-21-2012>

LENRs might be triggers for field failures/thermal runaway events in some Lithium battery fires:

There is a heretofore little appreciated subset of Lithium-based battery problems cryptically called a “*field failure*” mode that, while much rarer than ‘plain vanilla’ safety issues such as punctures and other types mechanical damage, seem to be highly correlated with catastrophic thermal runaway events. According to a major Lithium-ion battery manufacturer in a private communication, field failures apparently occur almost randomly in roughly 1 out of every 4 to 5 million Lithium-based battery cells right off the production line, regardless of their chemistry.

LENRs and field failures in advanced batteries

This somewhat obscure field failure problem involves catastrophic thermal failure of a single battery cell. While it is often thought to be associated with internal shorts and electrical arcing within a somehow defective cell, some battery manufacturers will admit privately that this peculiar failure mode is not well-characterized and very poorly understood --- most of them are presently at a loss for ideas about exactly how to definitively mitigate such a problem. It is well known that if just a single cell in a large, multi-cell battery pack fails in this particular manner, it can potentially trigger an even more catastrophic large-scale thermal runaway event that rapidly propagates through an entire battery pack, destroying adjacent cells via thermal fratricide as well as possibly the entire interior of, for example, an all-electric motor vehicle.

This additional new source of concern about the safety of advanced Lithium-based batteries has arisen because, in the course of our company's ongoing R&D efforts, Lattice has applied the Widom-Larsen theory of Low Energy Nuclear Reactions (LENRs) on a practical level to try to help better understand the possible role of nanoscale metal dendrites and nanoparticles in certain types of failure modes that may occur in smaller Lithium-based batteries as well as in extremely large, multi-thousand-cell battery packs utilized in all-electric vehicles and some military applications.

In May 2010, academic researchers at Oxford University published a new and we think important paper that many believe implicates the involvement of Lithium metal dendrites in a significant number of Li-ion battery failures (please see R. Bhattacharyya *et al.*, "*In situ NMR observation of the formation of metallic Lithium microstructures in lithium batteries*," *Nature Materials* **9** pp. 504 - 510). What is of great concern from a safety standpoint is that nanoscale internal metal dendrites that are prone to shorting-out can grow spontaneously over time as a given battery ages and goes through many charge-discharge cycles.

A battery pack may well be perfectly safe during the first months of ordinary use; however, dendrites and other types of nanoparticulate structures grow inside over time, increasing the probability of dangerous internal electrical shorts as the battery 'ages'. *The problem is that nobody in the world has any real working experience with large multi-cell Lithium-based battery backs that have endured hard usage and vibration for periods of many years. Also, nanoscale internal metallic dendrites can potentially form and grow in almost any type of Lithium-based battery chemistry.*

Approaching battery safety from perhaps a different technical perspective than many scientists, we have become increasingly concerned that some present/future Lithium-based battery chemistries could potentially be susceptible to rare, but potentially very damaging occurrences of LENRs in isolated nanometer to micron-scale regions within some failing battery cells. Cell field failures arising from nanoscale internal shorts/arcs are thus very worrisome with regard to potentially triggering LENRs that can in turn readily initiate macroscopic, catastrophic thermal runaways.

Please see the hyperlinked Lattice presentation dated July 16, 2010: field failures are exactly the type of nanoscale event that Lattice believes could potentially lead to the creation of tiny, internal micron-scale LENR 'fireballs' that could in principle initiate large-scale macroscopic, very hot-burning metal oxidation reactions that are very capable of generating their own free oxygen inside battery casings (as described in the presentation).

Nonpublic experiments have been conducted by a large company involving custom-built Li-ion battery packs comprising 50-60 commodity 18650 Li-ion cells with a standard chemistry; the wiring interconnection architecture was ~ the same as a typical EV battery pack. According to a private communication, results from deliberately induced, catastrophic Li-ion battery *field failures* were eye-opening: anomalously high temperatures in excess of 3,000 degrees were measured and recorded before thermocouples in failing battery packs were obliterated by intense heat. A detailed explanation of exactly how such anomalously high temperatures were achieved under such conditions is still under active investigation by the company's scientists.

What is somewhat worrisome about new types of Lithium Titanate battery chemistries in a field failure mode is that Titanium metal burns at a much hotter temperature than Lithium --- at ~3,400 degrees C. So, for example, an all-electric EV cruising down a highway could potentially encounter a 3,400 degree internal Lithium and Titanium metal fire with a fast-spreading flame front that generates its own oxygen as

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it combusts materials located inside a vehicle's failing battery pack. This could create a dangerous fire that might be difficult or impossible to extinguish. Even new types of inert Argon-foam fire suppression systems such as those retrofitted in some cargo aircraft are likely to be incapable of stopping a conflagration this hot that also creates its own source of oxygen as it aggressively heats battery materials.

If a large aircraft in flight that were to experience a hypothetical good-sized Li-Ti EV-class LENR-triggered battery fire, absent a robust thermal containment system it would seem that the plane's structural integrity could potentially be compromised because (ignoring the effects of a pressure-pulse if a large battery pack's casing actually detonates) a large heterogeneous 'blob' of molten material at 3,400 degrees is certainly hot enough to melt all the way down through the aluminum or composite fuselage of an aircraft ... a disturbing possibility.

A for-now nameless engineering firm with a large battery consultancy believes that about midway through such a super-hot fire in a very large EV-class battery pack, enough excess combustible gases could potentially accumulate inside the casing just ahead of a advancing flame front to enable a powerful detonation that completes the process of battery destruction --- i.e., a large chemical explosion combined with white-hot shrapnel that can ignite other nearby combustibles.

Interestingly, as speculatively discussed in the hyperlinked Lattice presentation dated August 23, 2012, evanescent 'flickering' superconductivity may occur in micron-scale patches just before they go LENR-active and make neutrons. If in fact this behavior occurred inside a battery, nearby nanostructures holding charge might well be trying to locally 'dump' current into a superconducting patch, further exacerbating the field failure problem. Also, per the Widom-Larsen theory of LENRs some fraction of the electrons located in such a patch would get converted into neutrons via an electroweak reaction ($e + p \rightarrow n$) which locally destroys charge, thus possibly causing more nearby charge to rush-in and fill the 'gap.'

Please now refer to the attached annotated excerpts from the book chapter by Barnett *et al.*: they have written an excellent, very informative document that discusses safety issues in the context of field failure modes in Lithium-based batteries. In my opinion, it is a must-read for people interested in battery safety and well-worth the purchase price of \$143.50 for Springer's full Kindle eBook version.

I have taken the liberty to annotate Barnett *et al.*'s book chapter so that readers can easily connect blocks of text to LENR-related ideas found in this cover preface as well as in the other mentioned Lattice presentations found on SlideShare. You will find that their thinking resonates strongly with ours and that LENRs appear to be a plausible trigger for some indeterminate subset of field failure events. These can in turn potentially lead to catastrophic thermal runaway processes that presently pose a major safety risk in advanced batteries with high energy densities.

As long as it does not involve any disclosure of Lattice-proprietary technical information that we deem relevant to energy production applications, Lattice is interested and prepared to engage in fee-based consulting with other companies in regard to assessing safety issues involving LENRs in connection with field failures, battery fires, and thermal runaways.

Technical questions and inquiries are welcome.

Thank you.

Lew Larsen
January 23, 2013

**Brian Barnett, David Ofer, Suresh Sriramulu,
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Glossary

Abuse	External stress applied to a battery or cell that is not anticipated to occur under normal operating conditions and may cause the cell to experience thermal runaway.
Cascading	Process by which one cell in a battery releases sufficient heat that thermal runaway of one or more neighboring cells in a pack ensues.
Field-failure	<i>Infrequent</i> safety incident that occurs in lithium-ion cells/batteries in the field under “normal” operating conditions and does not appear to have an obvious external trigger, but is often caused by an internal short circuit. Such internal short circuits are often caused by foreign metal particles.
Safe Zone	For a given cell construction and materials, and for a given heat transfer environment, conditions of energy and power associated with an internal short such that a thermal runaway is not possible.
Thermal runaway	A process of uncontrolled heat release and rapid temperature rise.
Threshold energy	For a given cell design and heat transfer environment, an energy value dissipated in an internal short below which no thermal runaway is possible.
Threshold power	

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	For a given cell design and heat transfer environment, an internal short power below which no thermal runaway is possible.
Trigger for thermal runaway	A stimulus that initiates thermal runaway in a Li-ion cell. Examples of potential triggers include a variety of abuses as well as internal short circuits created by foreign metal particles.

Definition of the Subject and Its Importance

Safety of lithium-ion batteries is a critical topic that has not received adequate attention in the past, largely due to the fact that data regarding safety failures have been severely restricted. As a result, there are numerous misunderstandings in a field that has not received the same degree of scientific and technical rigor as other areas of lithium-ion battery technology development. However, safety of lithium-ion batteries will become even more important as lithium-ion technology enters transportation markets. Under suitable triggers, Li-ion cells can experience thermal runaway, i.e., the rapid increase in cell temperature accompanied by venting, vent-with-flame, ejection of cell parts, fire, and explosion. Safety failures of lithium-ion cells can result from a variety of triggers including overcharging, overheating, crushing, mechanical impact, and external short circuits. Safety tests have been devised for all these abuses, with varying degrees of fidelity. However, most safety incidents that have taken place with lithium-ion batteries occur due to the slow and rare development in cells of internal short circuits that mature to the point that they result in thermal runaway. Most safety tests carried out in the laboratory or factory do not replicate the conditions by which safety incidents actually occur in the field. These issues are characterized in detail, and an improved overall framework for considering lithium-ion battery safety is suggested.

Introduction

Since their introduction in 1991, lithium-ion batteries have become the dominant rechargeable battery technology for portable products and are beginning to make inroads in transportation and stationary storage applications. Lithium-ion battery technology provides the highest available energy density of any long-cycling rechargeable battery, which has been the most significant factor leading to its widespread adoption. As of this writing, over 4.5 billion lithium-ion cells (almost 40,000 MWh) [1] will be expected to be produced in 2011, the bulk of which are used in consumer electronics applications such as laptop computers and cell

phones. While the demand for such cells is increasing at the rate of 15% on a year-on-year basis, emerging applications can significantly increase the Li-ion market. It is anticipated that emerging transportation and stationary storage markets will lead to dramatic increases in the volume of Li-ion production. One recent estimate places automotive Li-ion demand in 2020 at 150,400 MWh, in contrast to 72,500 MWh projected for portable applications in that year. The transportation and stationary applications are expected to use larger capacity cells than those employed for consumer electronics applications [1].

Despite the obvious success of Li-ion technology, safety concerns remain [2]. Under suitable triggers, Li-ion cells can undergo venting, vent-with-flame, ejection of cell parts, fire, and explosion as the consequence of a process of uncontrolled heat release, termed “thermal runaway.” This risk is not surprising given the possible sources of heat release within a cell. Essentially, Li-ion batteries store a very large amount of electrochemical energy, which can be released as heat, for example, through an internal short. The electrochemical energy content of state-of-the-art Li-ion cells is by itself sufficient to raise cell temperature to 700°C if completely dissipated within the cell under adiabatic conditions. But furthermore, Li-ion cells contain energetic materials that can undergo exothermic decomposition reactions within the cell, as well as containing combustible organic solvents (with heat of combustion that is about a third of the heat of combustion of gasoline) and combustible carbonaceous anode materials, both of which can burn in atmospheric oxygen if exposed under suitable ignition conditions. The heat release from such reactions can be as much as ten times the stored electrochemical energy.

Safety failures of lithium-ion cells can result from a variety of triggers. Overcharging, overheating, crushing, mechanical impact, and external short circuit all represent forms of external triggers (often termed “abuse” conditions) that can lead to safety incidents. These external abuses have been extensively studied. Battery engineers have developed several cell-level and pack-level tests to evaluate how a cell/battery responds to the abuse, and they have also developed technologies and approaches to manage the cell and battery response to such external abuses [3–11]. Standardized tests also have been developed to assess cell and battery “tolerance” to external abuses [12, 13]. In general, this chapter shall try to draw distinctions between tests used to screen cells/batteries for their tolerance to a given trigger and the actual tolerance of a cell/battery to the given trigger (i.e., the effectiveness of the test).

Safety concerns have been heightened by highly publicized safety incidents and ensuing widespread recalls of lithium-ion batteries used in laptop computers and cell phones [14, 15]. When these rare safety incidents occur, lithium-ion batteries operating under otherwise normal conditions undergo what appear to be spontaneous thermal runaway events, often with violent flaming and extremely high temperatures. Moreover, these failures usually involve cells and cell designs that have passed extensive abuse testing, including the standardized abuse safety tests. *Most such Li-ion safety incidents in the field are not preceded by any obvious external abuse.* We refer to these spontaneous safety incidents as “field-failures”.

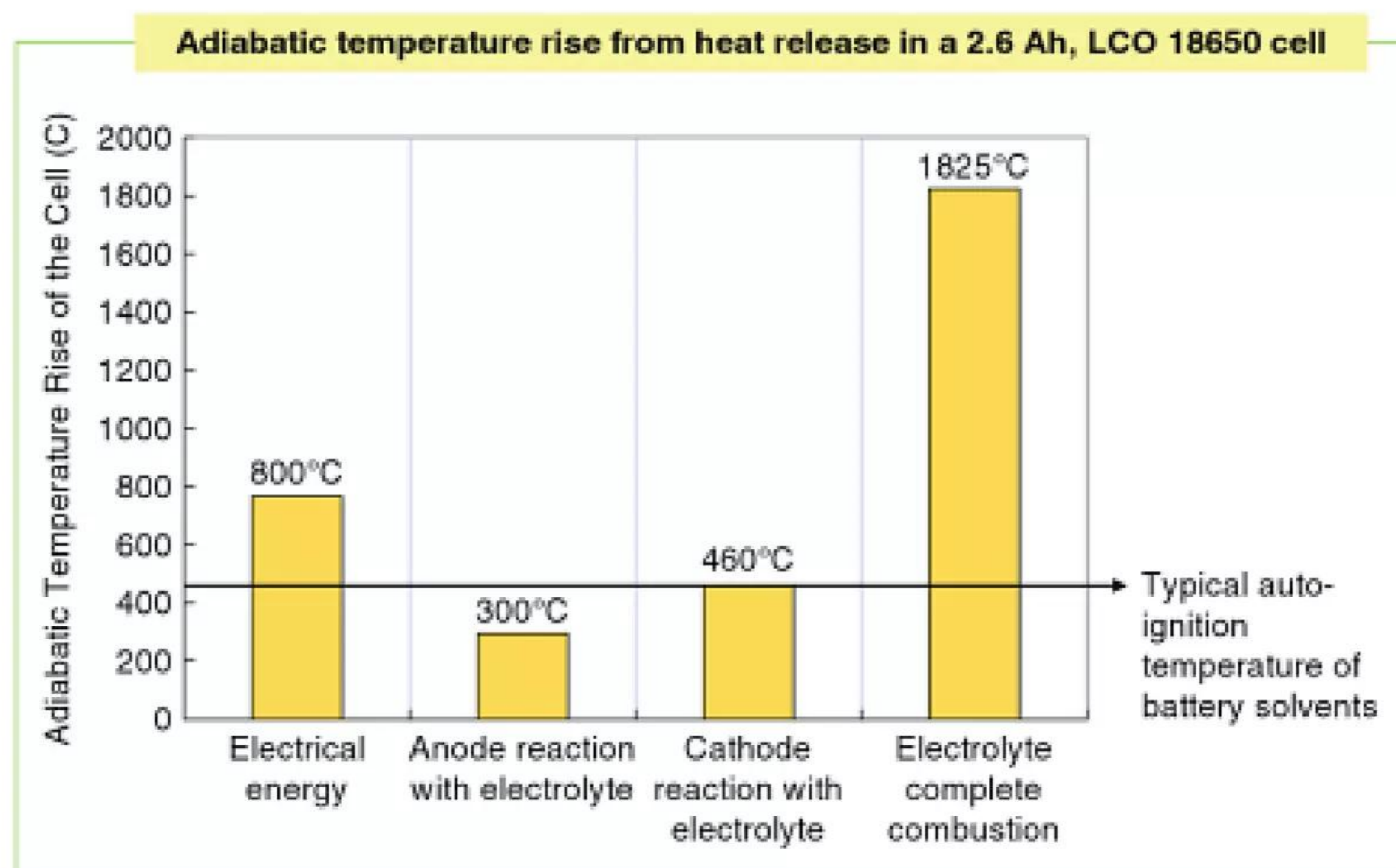


Fig. 9.1 Estimate of the adiabatic temperature rise from heat release in a 10-Wh Li-ion 18650 cell

combusted upon exposure to atmosphere; postmortem audits show that this does in fact occur in many field-failures. Graphite's heat of combustion is nearly 33 kJ/g, meaning that complete combustion of a 2.6-Ah cell's graphite content will generate about 260 kJ.

Adding graphite's combustion heat to the energies listed in Table 9.1, the total energy release by an 18650 cell can exceed 400 kJ. The heat that can potentially be released within the cell absent of atmospheric combustion, shown in Table 9.1, can itself be significant and can drive severe cell temperature increases. The highest temperatures to which cells might be driven by these processes can be estimated by considering the adiabatic temperature rise associated with the different sources of energy release discussed above, as summarized in Fig. 9.1. Release of electrochemical energy alone, as by full discharge of the cell through an internal short, can raise the cell temperature by more than 700°C under adiabatic conditions. Furthermore, the anode and cathode exothermal reactions together can also raise the cell temperature by 700°C. These temperatures are significantly higher than the autoignition temperature of the solvent, and hence, if the solvent is ejected from the cell at these temperatures, it is likely to combust outside the cell, further stimulating the development of very high temperatures, and igniting the graphitic anode with explosive combustion.

Table 9.1 and Fig. 9.1 illustrate the powerful energetics underlying thermal runaway events. An increase in temperature can stimulate significant heat release from the exothermic anode, cathode, and electrolyte decomposition reactions. If the rate of heat removal is slower than the rate of heat generation, thermal runaway will eventually occur, accompanied by enormous heat release and very high temperatures.

Triggers for Li-Ion Thermal Runaway

In general, some form of trigger, or initiating process, is required to produce a thermal runaway event. The previous section amply illustrates that triggers producing increased temperature of either the whole cell or a significant region of the cell can result in thermal runaway. [Table 9.2](#) summarizes some common triggers. Ideally, battery developers and implementers can utilize safety tests that specifically screen for a cell's or battery's tolerance to a given trigger.

The triggers that have received the greatest attention include abuse conditions such as external heating, overcharging, external shorting, impact, penetration, etc. Some of these triggers are relatively easy to anticipate as risks, and to study and to develop screening tests for, often by selecting a set of abuse conditions that are reasonably plausible. In fact, numerous standardized (or semi-standardized) abuse tests have been developed to assess abuse tolerance (a broader all-encompassing term might be "trigger tolerance"). Examples of such tests include the hot box test, the forced overcharge test, the external short circuit test, etc. [12, 13] These tests are not reviewed in detail, except to point out that they do not generally replicate the conditions in which actual safety events occur in the field. Certain other triggers present more difficult challenges with respect to development of truly effective safety screening tests. In addition, some safety tests can be "gamed," i.e., manipulated to favorably alter the outcome by carrying out the tests under certain conditions, or by modifying cells so that they can pass the tests rather than demonstrate true tolerance to the particular abuse. Situations where definition of plausible screening safety tests may *not* be possible are highlighted. [Table 9.2](#) identifies (rows 4 and 5) two such triggers.

Technologies and components such as PTCs, CIDs, vents, and safety circuits have been incorporated in today's lithium-ion cells and packs in order to manage safety with respect to many external abuse triggers.

Table 9.2 Summary of triggers that can lead to thermal runaway of Li-ion batteries

Trigger	Why can this occur?	Is this managed?
Overcharge	Defective connections, failure of charging circuit	Yes, battery management system Yes, cell-level safety devices
Overheating from external sources	Battery pack placed too close to a heat source	Yes, cell-level safety devices open the cell at suitable internal pressure
Cell crushing creating massive internal shorts	Physical abuse of battery pack	Yes, design enclosures are built more tolerant to specific abuses
<i>Internal short-circuits (a.k.a., field failures)</i>	Internal-short caused by manufacturing defects	No, new technologies needed
<i>Cascading of thermal energy release</i>	Affected cell can raise the temperature of surrounding cells	No, new technologies needed

- In field-failures, an induction period lasting on the order of minutes is often reported, during which self-heating of the battery is discernible by users/observers, prior to violent flaming and explosion.
- Postmortem examinations of actual field-failures indicate that all or portions of a cell have experienced very high temperatures. For example, cells from incidents routinely exhibit multiple localized silver-colored metallic beads. These beads are recondensed aluminum, which melts at 660°C. In some instances, there are areas in which copper current collectors have also experienced melting. Copper melts at 1,085°C.
- Neither the actual mechanism of short circuit initiation nor the mechanism by which an internal short leads to a thermal runaway is fully understood yet. After the fact, postmortems show a level of destruction that renders it impossible to ascertain the exact morphology that existed at the point of thermal runaway. We use experimental and simulation results to discuss the likely mechanism of short formation, and the factors that control thermal runaway following an internal short.

Implications for Managing Field-Failures

Most safety incidents that have been observed and evaluated involved portable computer battery packs. Although some incidents have taken place in mobile phone batteries, computers use many more cells, with cells in series and in parallel. Our discussion of the mechanism of short initiation will show that such failures are also possible in HEV, PHEV, and BEV cells. Without suitable safety technologies in place, safety incidents in HEV, PHEV, and BEV packs are likely to be more severe than those observed in laptops owing to the larger cell sizes employed (approximately 2–30 times on an energy basis) and the lower impedance of the cells. Furthermore, with roughly 75–80 cells per pack (for cells in a single string) and the total energy per pack approximately 20–200 times larger than that of computer packs, the consequences of thermal event “cascading” is a major concern. Field-failure in a vehicle pack could inflict far greater damage than that caused by notebook computer batteries.

Because the consequences of these rare safety events can be so serious, especially with increased use of lithium-ion in transportation applications, it is necessary to recognize that no manufacturing environment can always operate with zero defects. Therefore, given the recognition that foreign particles in cells can never be completely eliminated, there is a need to develop safety technologies that are “on guard” to catch their deleterious effects as early as possible. A useful perspective is to recognize that achieving Li-ion systems which are totally free from the threat of spontaneous safety events is *not* a quality problem. For example, the rate of field-failures for cells produced by the most experienced manufacturers is already at a very low level – one failure in 5–10 million cells produced, which already far exceeds six sigma manufacturing quality guidelines. These manufacturers typically

benefit from the most powerful elements of a learning curve – they have produced large numbers of cells (over a billion in several cases) over a very long period of time (more than a decade). It is reasonable to assume that less-experienced manufacturers will eventually approach similar levels, but no manufacturer will achieve zero defects in perpetuity.

Manufacturing improvements should, of course, continue to be implemented whenever possible, but it should be recognized that improvements in the cleanliness or quality control of manufacturing lines are not the fail-safe solution. Manufacturing “quality” is already maintained at a very high level. The defect rate is already very low and not readily correlated with the level of small extraneous metal particles in cells. Greatly reducing the level of particles in cells could probably only be accomplished with significant (and unacceptable) additional processing costs. (For example, consider the manufacturing quality levels that must be achieved in the semiconductor industry, and the associated costs.) Clearly, a new approach is needed to make significant progress toward improving the safety of Li-ion batteries and, in particular, to deal with the insidious role of the internal short circuit in causing lithium-ion safety events.

We have been developing tools and techniques to enable the development of new technologies to manage field-failures. In the following sections, some insights are described that have been gained from exercising these tools; namely, the likely mechanism by which a foreign metal particle forms an internal short, and the mechanism by which an internal short then stimulates thermal runaway.

Likely Mechanism of Internal Short Formation in a Field-Failure

The exact mechanism of field-failure due to foreign metal particles in cells may never be known with certainty, given the rarity of events and the extensive destruction that takes place in affected cells. The realization that field-failures are rare has a number of practical consequences for studies of internal short formation, for attempts to identify electrical signals warning of developing internal shorts, and for development of any safety technology. In particular, a reliable method(s) to generate internal shorts “similar” to those that lead to thermal runaways in the field is essential. Although not discussed in detail in this chapter, we have developed several methodologies to create internal shorts in cells. This section describes the most likely mechanism for initiation of the internal short circuit from metal particle contamination and how we have used internal short implantation methods to investigate that mechanism.

The most likely mechanism for initiation of the internal short involves the formation of a metal dendrite between the anode and cathode. In this process, a metal particle present in or on the cathode (positive) will dissolve by an electrochemical oxidation process, and the resulting metal ions will diffuse to the anode

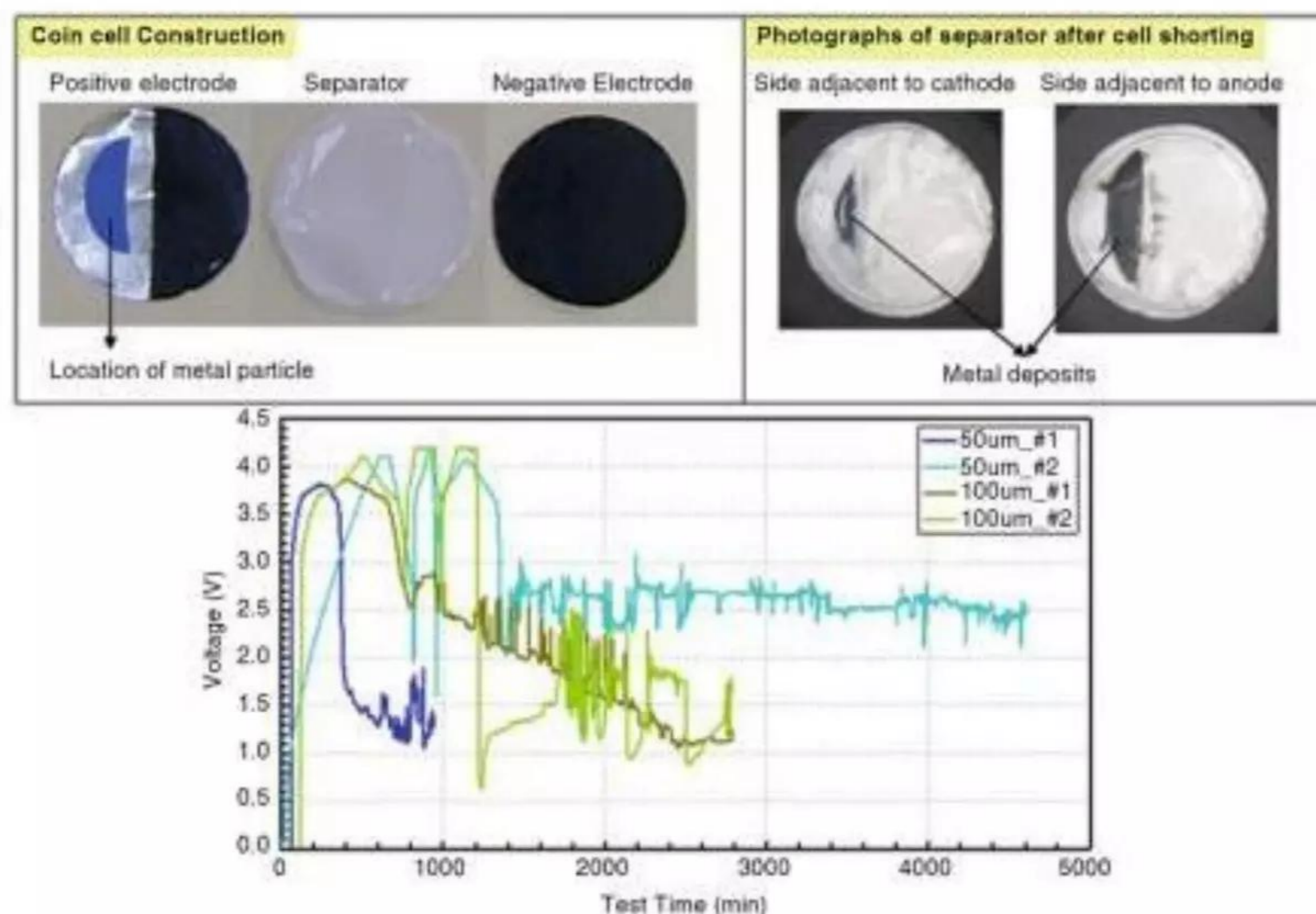


Fig. 9.2 Internal short formation in a coin cell. Photographs in the top left panel show the coin cell construction for these tests. A crescent-shaped Ni particle was placed on a scraped-off portion of the positive electrode. The cycling data on the bottom panel show the formation of an internal short circuit in the cell during normal charge/discharge cycling, for two different thicknesses of the Ni particle (two different coin cells for each particle thickness). The photographs in the top right panel are of the separator from one of the coin cell tests. The metal deposits can be clearly seen. The cathode active material was LiCoO_2 , the separator was Celgard 2325, and the anode material was MCMB2528

(negative) where they will plate, often over an extended period of time and many charge/discharge cycles, resulting in the formation of a metallic dendrite that eventually makes a durable electrical contact back to the cathode and shorts the cell. In the field, the exact location of the short in a cell is also a critical factor with respect to occurrence of thermal events. We have confirmed this mechanism by implanting particles of iron and nickel in Li-ion cells.

Figure 9.2 provides experimental evidence supporting this mechanism. The top left panel of Fig. 9.2 illustrates a cell constructed with a large (oversized relative to particles implicated in field-failures) nickel particle with thickness of either 50 or 100 μm placed on the cathode current collector. The bottom pane of Fig. 9.2 shows the subsequent cycling of this cell to the point of development of an internal short. Postmortem examination of the cell was very instructive. The top right panel of Fig. 9.2 shows deposits of plated metal adhered to the separator. Inspection of both sides of the separator shows that the deposit's area is larger on the anode side than on the cathode side, clearly indicating the direction of its growth from the anode to the cathode. Furthermore, EDAX of the anode itself shows deposition of nickel on the anode,

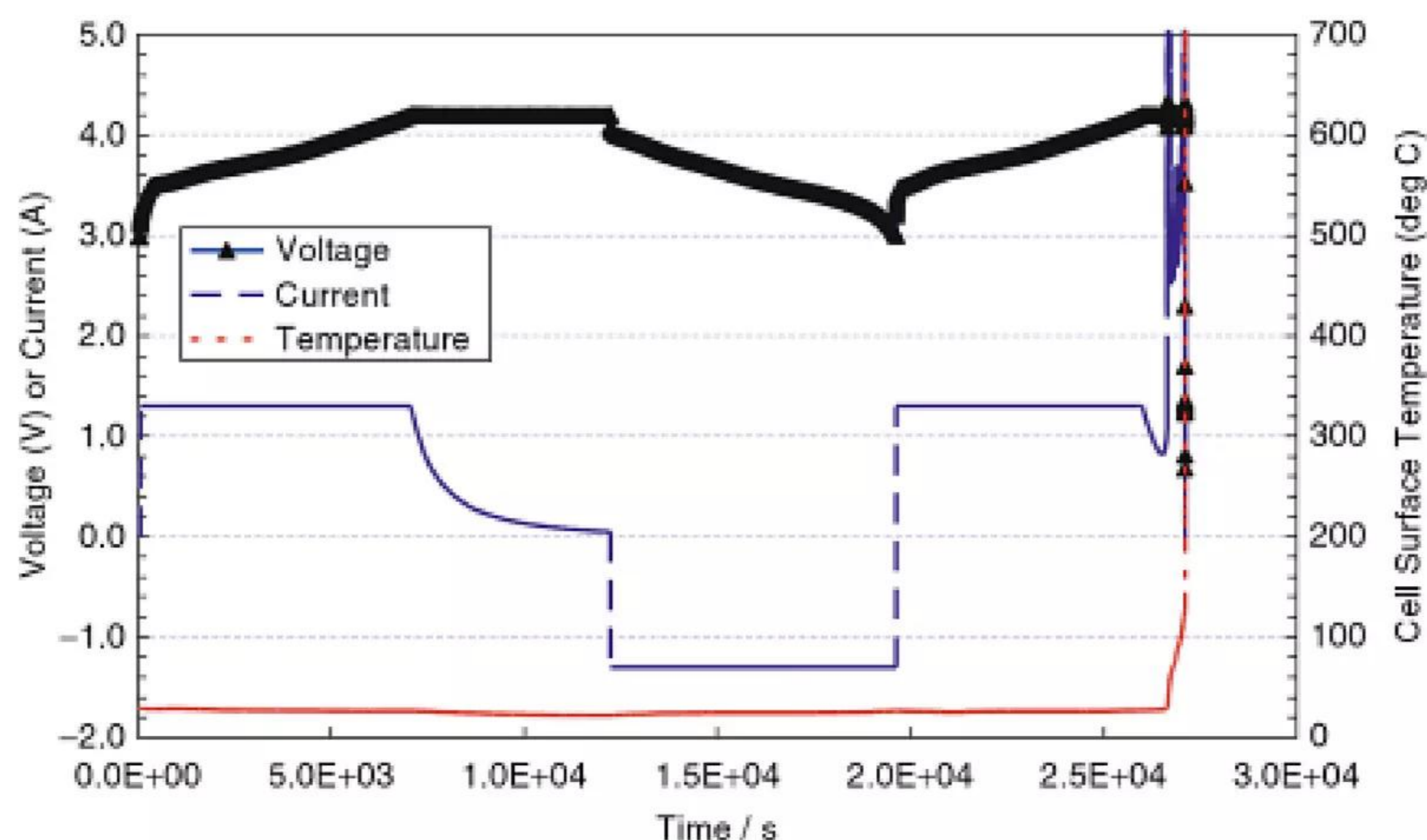


Fig. 9.3 Response of an 18650 cell following deliberate implantation of a metal particle on the cathode of the cell. In this experiment, a fully discharged commercial 18650 cell was removed from its can. The jelly roll was partially unwound, and a metal particle was placed on the cathode side. The jelly roll was then placed in a suitably sized can and normally cycled. The cell experienced thermal runaway during normal charge/discharge cycling

which of course was not present prior to cell assembly. Taken together, these results clearly show the operative mechanism for formation of internal shorts. Metal particles on (and possibly in) the cathode dissolve and plate out on the anode, growing back through the separator, leading to an internal short.

It is not surprising that a nickel metal particle present in the cathode would result in shorting of the Li-ion cell. The standard potential for Ni/Ni^{2+} is 2.9 V (vs Li/Li^+), implying that Ni will dissolve at typical cathode potentials >3.5 V (vs Li/Li^+) and plate at typical anode potentials <1.5 V (vs Li/Li^+). Similar behavior is also expected of Fe; the standard potential for Fe/Fe^{2+} is 2.6 V (vs Li/Li^+). Separate cyclic voltammetry experiments on Ni and Fe foils in Li-ion battery electrolytes also confirmed this mechanism. This mechanism also explains why internal shorts can develop even on open circuit and explains how the shorts can “grow” over time and not be present for some time after manufacture of the cells. Plating and dendrite growth is a stochastic process, and induction of the overall cell shorting process is also likely to be so. Thus, several factors can limit the rate of dendrite growth and probably even its occurrence.

It has also been experimentally confirmed that metal particles placed on the cathode in 18650 cells can cause internal short circuits capable of inducing thermal runaway (sample result shown in Fig. 9.3). These experiments will be detailed in a future publication. Using this sort of test platform, the resistance of shorts and the evolution of that resistance during tests can be estimated. It has also been possible to evaluate variations in behavior of various metals and variations

in the nature of deposits formed with different metals. Judicious placement of particles can also lead to short formation and cell failure after multiple charge/discharge cycles (as opposed to during the initial charge). The delayed short formation facilitates efforts to identify precursor signals that can warn of development of an internal short in the field.

Several factors can determine how a short might grow from a foreign metal particle, and hence determine the magnitude and rate of local i^2R heating as the cell discharges through the short circuit. These factors include: the nature of the metal, the size and shape of the particle, the rate capability of the cell, separator permeation and thermal properties, charge/discharge history of the cell, and temperature distribution/history. At this moment, there is insufficient experimental data to comment on the relative contributions of these factors in determining the rate and magnitude of heat release through the short circuit. However, we are performing specially designed experiments to probe these factors and will discuss some of them in a forthcoming publication.

Other Possible Mechanisms of Internal Shorting

There are various ways in which an apparently normal cell might develop an internal short in the field [39, 40]. However, the nature of the internal shorts formed by these different routes, and the extent to which they may or may not be likely to induce thermal runaway, might be very different. Mechanisms cited in the literature or generally discussed by battery engineers that implicate foreign metal particles are summarized below:

- *Lithium deposition/dendrites* form during cell operation for a number of possible reasons, including charge processes that drive the anode to the lithium potential or as a result of lithium imbalance in the cell. Unintentional overcharge due to a faulty BMU can also result in lithium deposition on the anode, with repeated overcharging leading to lithium dendrite growth. There is also evidence reported for foreign metal particles on the anode surface serving as nucleation sites for lithium deposition. There is no doubt that lithium deposition occurs in lithium-ion cells under a variety of circumstances. We suspect that lithium dendrites can lead to internal shorts, resulting in premature cell failure. However, lithium dendrites are less likely to dissipate sufficient power to enable thermal runaway than are dendrites of higher melting point metals. Our simulations show that the temperature of the dendrite itself can significantly exceed the melting temperature of lithium metal (180°C). Hence, it is less likely that lithium deposits/dendrites could support powers similar to those possible with metal contaminants such as Ni or Fe (melting temperatures >1,400°C).
- *Particle migration and separator puncture.* A metal particle in the cell (presumably in/on the anode) can migrate somewhat as a result of the volume changes that occur during charge and discharge and can translate sufficiently to pierce the

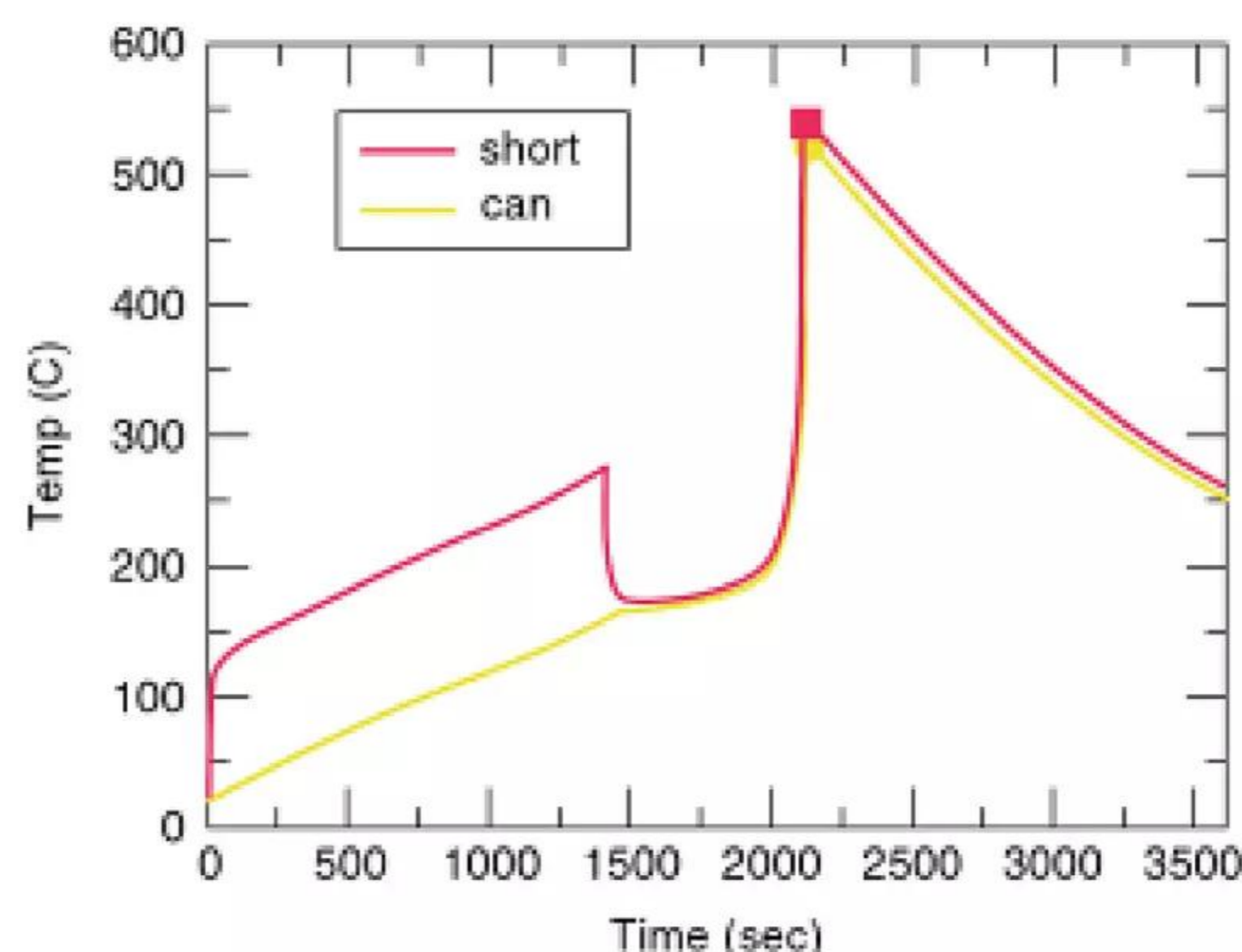


Fig. 9.5 Temperature history in the vicinity (~ 2 mm away) of the short circuit and at the can surface for a 10-W short circuit that is active for 1,410 s in a 2.6-Ah 18650 cell

cumulative heat release and extent of reaction for the anode and cathode decomposition reactions. A surface heat transfer film coefficient of $10 \text{ W/m}^2\text{-K}$ was employed, corresponding to nearly quiescent ambient conditions. The initial temperature and ambient temperature were both set to 20°C .

The results shown in Figs. 9.5–9.7 can be considered in terms of three distinct time periods. In the first time period, the temperature in the vicinity of the short circuit is higher than that at the outer extremities of the cell, owing to heat generation from the short circuit (which has been placed near the center of the cell). The internal short is a localized heat source, or hot spot. In practice, this heat source can be driven from other cells in the battery pack or just from the shorted cell itself in the case of an isolated cell. In addition to electrical heating, heat release from the anode and cathode decomposition reactions, occurring wherever critical onset temperatures in the cell have been exceeded, also contributes to the temperature rise. As shown in Fig. 9.6, the iso-temperature contours are oblong in this time frame due to significantly higher thermal conductivity in the axial direction of the jelly roll compared to the radial direction.

Once the short power is reduced to zero at 1,410 s, the cell temperature quickly becomes uniform, as heat transfer internal to the cell equalizes the temperature within the cell.

In the second time period (from about 1,410–2,000 s), there is no longer a contribution from the local heat source because the internal short has been turned off. The temperature continues to increase, albeit at a much slower rate. In this time frame, the rate of heat release from the decomposition reactions is only slightly higher than the rate of heat loss at the cell surface, resulting in a gradual rise of the cell temperature. As can be seen from the results in Fig. 9.7, it is primarily the exothermal anode decomposition reactions that contribute to this heat release; the

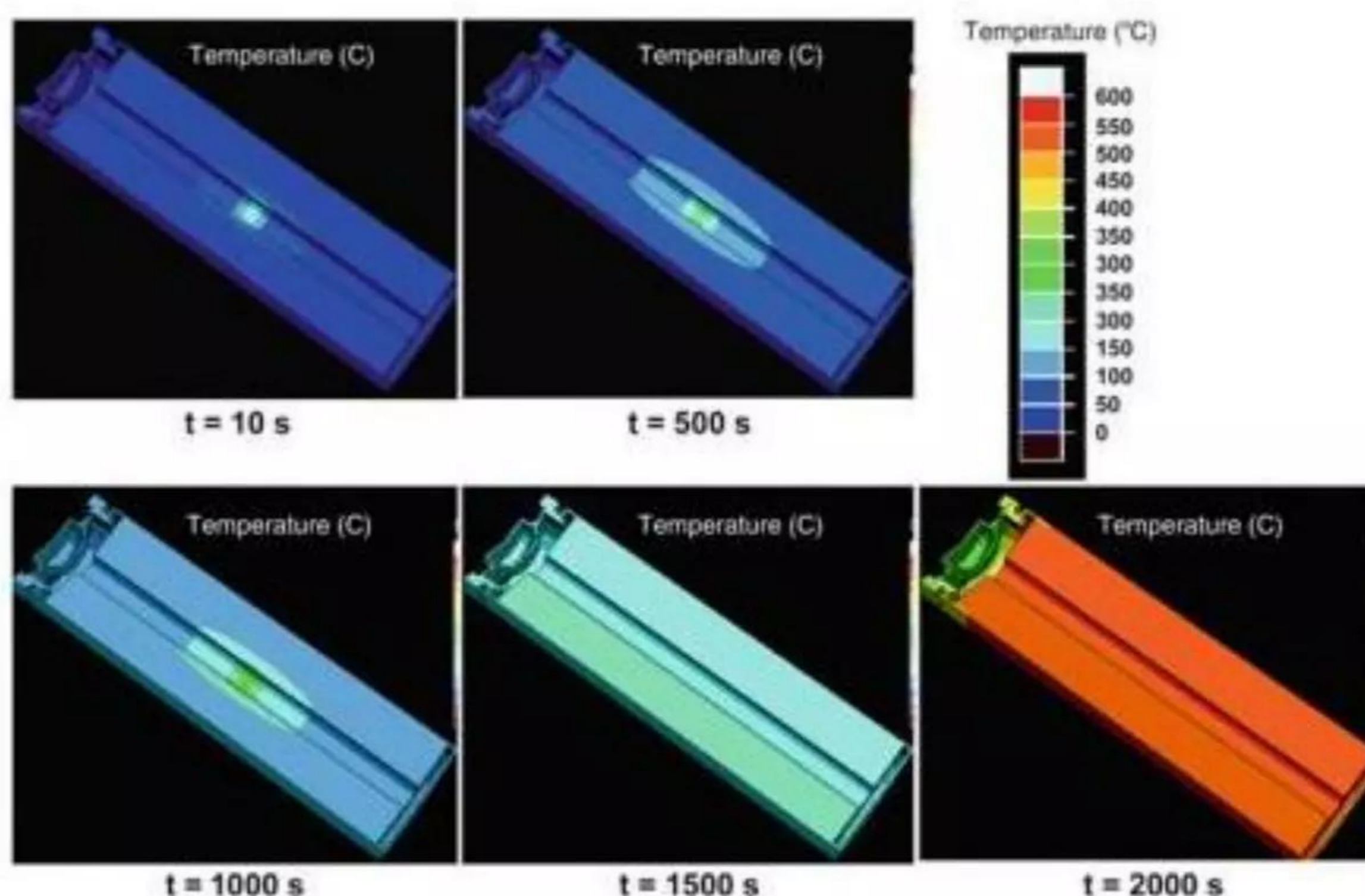


Fig. 9.6 Temperature distributions corresponding to the simulation in Fig. 9.5 showing the progressive change in temperature in a cell following the formation of an internal short circuit that leads to eventual thermal runaway

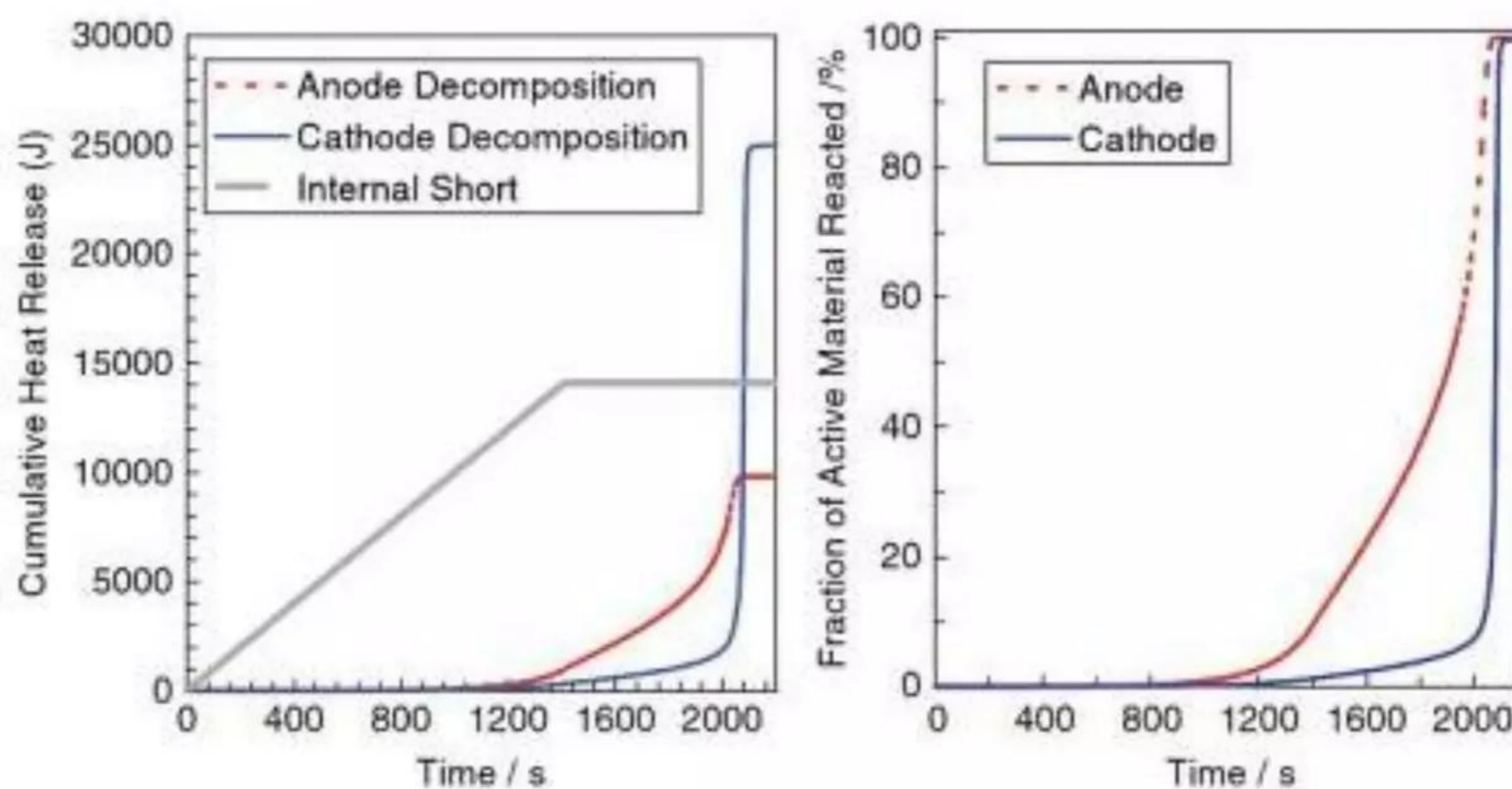


Fig. 9.7 Cumulative heat release (plot on the left) and the extent of reaction (plot on the right) for the anode and cathode decomposition reactions, corresponding to the simulation results in Fig. 9.5

energy contributed from the cathode decompositions is insignificant in this time frame. The anode decomposition reactions assume a triggering role because their rates (and hence the associated heat release) become significant at a lower temperature than the cathode decomposition reactions, and hence, the anode reactions

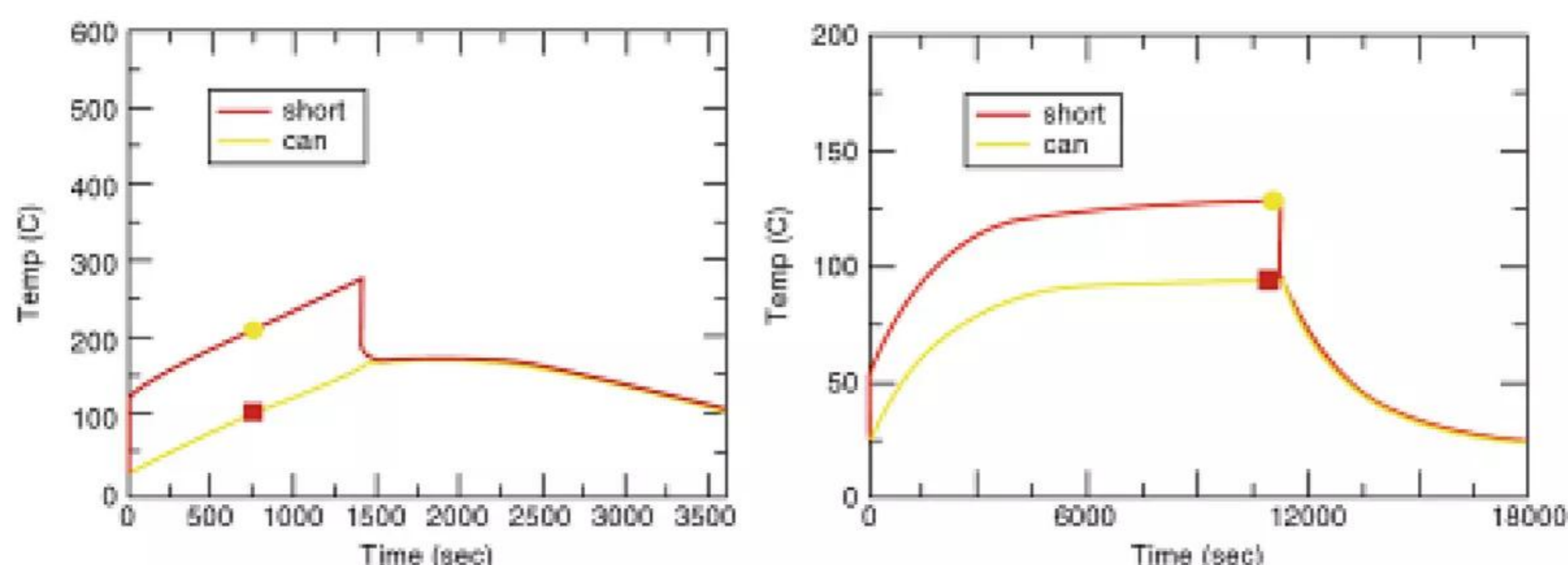


Fig. 9.8 Simulated temperature profiles for two conditions where thermal runaway does not occur. The plot on the left corresponds to the case where a 10-W short was active for 1,400 s. The plot on the right corresponds to a short power of 3.5 W, which was active until the cell completely discharged through the short circuit without causing thermal runaway

proceed to a greater extent than the cathode reactions in this time frame. The particular example and corresponding set of conditions being discussed here have been chosen to illustrate a condition in which a sufficient percentage of the material in the cell has been elevated to a temperature that is just above the “point of no return” on the path toward thermal runaway. In an equivalent simulation in which the internal short is turned off just 10 s sooner at 1,400 s, discussed below and shown in Fig. 9.8, no thermal runaway ensues. The substantial length of time for which an internal short can be active prior to inducing thermal runaway suggests that there may be sufficient time to intervene and prevent a thermal runaway if adequate signals indicating development of an internal short can be identified.

In the third time period, beyond about 2,000 s, the cell begins to heat up dramatically and goes into thermal runaway. Self-heating of the cell accelerates because the rates of the anode and cathode decomposition reactions accelerate with rising temperature and because the extent of active material in the cell that contributes to this heat generation also increases. The combination of these factors results in an unstable increase in temperature which is known as thermal runaway. The rapid rise in temperature is roughly what witnesses report when they have been in proximity to lithium-ion field-failures. Note not only the very rapid rise in temperature in Fig. 9.5 once thermal runaway has initiated, but also the general range of temperatures that are realized as a result of the runaway. The latter reflects the general range of temperature known to occur in safety incidents because of the evidence (previously cited) that is obtained from cell postmortems.

The above discussion illustrates the complexity of thermal runaway induced by an internal short. Furthermore, the rapid rise in temperature at ~2,000 s illustrates one of the reasons that material-level heat release from DSC or ARC measurements by themselves are not likely to be relevant predictors of relative safety of various cathode materials. Note how in Figs. 9.6 and 9.7 the progression to a thermal runaway is inevitable beyond about 1,410 s, at which point the short has been turned off in this simulation, and note how the cell quickly traverses a wide range of temperatures (in this case, from about 150°C to about 550°C) as runaway proceeds.

As mentioned above, this model has also been used to explore the conditions under which thermal runaway will not occur. Figure 9.8 shows results for two conditions where thermal runaway does not occur. The figure on the left corresponds to a 10-W short, otherwise identical to the one depicted in Fig. 9.5, which is active for 1,400 s, whereas the figure on the right corresponds to a 3.5-W short that is active until the cell completely discharges through the short. All other conditions are identical to the simulations corresponding to Figs. 9.5–9.7.

Comparing the results in Fig. 9.5 with the left hand plot of Fig. 9.8 shows that the energy dissipated in the short is a key factor in determining thermal runaway. Essentially, when the 10-W short is active for 1,400 s (Fig. 9.8), thermal runaway does not occur. But when the 10-W short is active for only 10 s longer (Fig. 9.5), thermal runaway occurs. Thus, there is a minimum energy (or “threshold energy”) that must be dissipated in the short before thermal runaway will occur. The specific value of that threshold energy is related to cell design, materials employed, and operative heat transfer conditions. Of course, slight variations in any of these parameters might result in a different threshold energy.

The right-hand plot in Fig. 9.8 shows a case in which short power has been reduced in successive simulations until thermal runaway does not occur no matter how long the short is maintained in an active state (a “threshold power”). Note the parallel lines in Fig. 9.8 beyond about 5,000 s, corresponding to the state in which the rate of heat generation in the cell is exactly equal to the rate of heat loss from the outer surface of the cell. Since the experiment is bounded by the total energy of the cell, the short essentially terminates when there is no capacity remaining in the cell. As can be seen in the figure, the cell temperature then decays to room temperature.

The Concept of a “Safe Zone”

The results presented in Figs. 9.5, 9.7, and 9.8 have been chosen from a larger number of simulations because, in combination, they illustrate how a new figure can be constructed from simulations to demarcate a “Safe Zone” for a given cell construction and cell materials and for the associated heat transfer conditions. Figure 9.9 illustrates the construct for a generic 18650 cell. In this figure, various short powers are represented on the x-axis. The y-axis corresponds to the relative energy dissipated in the short as a percentage of the total electrochemical energy in the cell.

Figures 9.5, 9.7, and 9.8 represent conditions that can be placed on this chart. The 10-W short begins at $t = 0$ corresponding to zero energy release on the y-axis and moves vertically with increasing time. At 1,400 s, there has been insufficient energy input to stimulate a thermal runaway, but at 1,410 s, thermal runaway ensues. A threshold energy has been exceeded. Running a series of such simulations, a family of such situations can be constructed, demarcating a threshold energy curve on the plot.

Similarly, Fig. 9.8 (right) has already demonstrated existence of a threshold power below which no thermal runaway is possible, no matter how long the short is

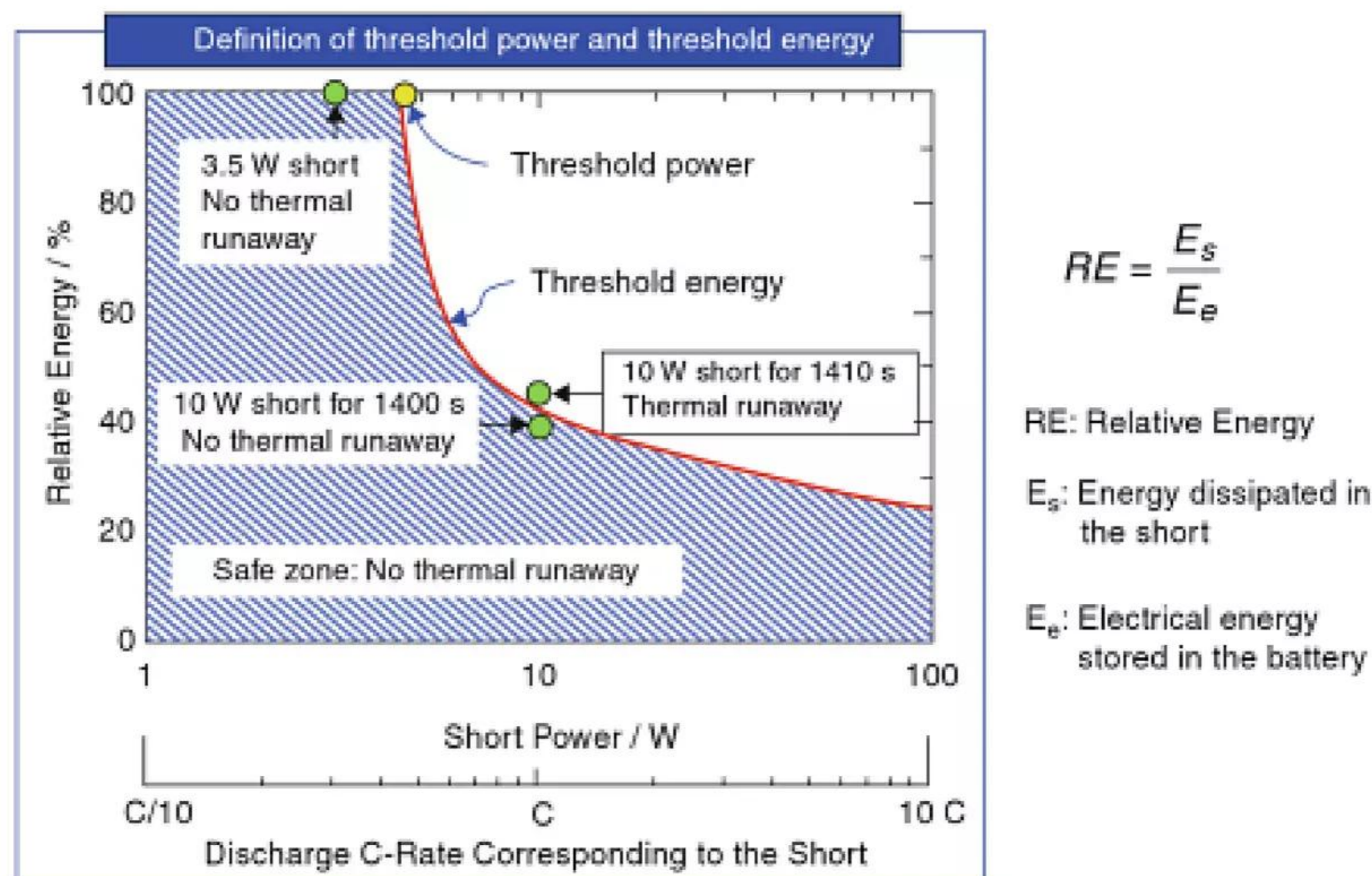


Fig. 9.9 The Safe Zone construct

in place. The existence of a threshold power implies that there will be a finite range of short resistance capable of dissipating sufficient localized heat to stimulate thermal runaway. If short resistance is above this critical range, insufficient current is supplied to the short. The critical short resistance range will be defined by the characteristic of the cell and pack.

Note in Fig. 9.9 how the threshold energy curve rises sharply as short power is reduced to values approaching the threshold power. The resulting cross-hatched area is a “Safe Zone.” Internal shorts with power or energy characteristics within this region will not induce thermal runaway. In practice, the threshold power is a more critical parameter, since in the field there are no practical means to interrupt an internal short once it has been initiated. Implications of the threshold energy curve’s shape will be discussed in a future publication.

Note that a “Safe Zone” plot can be constructed for any lithium-ion cell, representing the particular cell size and design (i.e., form factor, capacity, electrode dimensions, etc.) and materials employed, as well as the particular heat transfer conditions that apply. One can use Safe Zone plots to compare the relative safety of various cells and/or materials, as well as to evaluate the relative safety of cells under various heat transfer conditions.

Illustrative Applications of the Safe Zone Concept

The Safe Zone concept allows a quantitative assessment of safety in a way that has not previously been possible. There are numerous ways that the Safe Zone concept

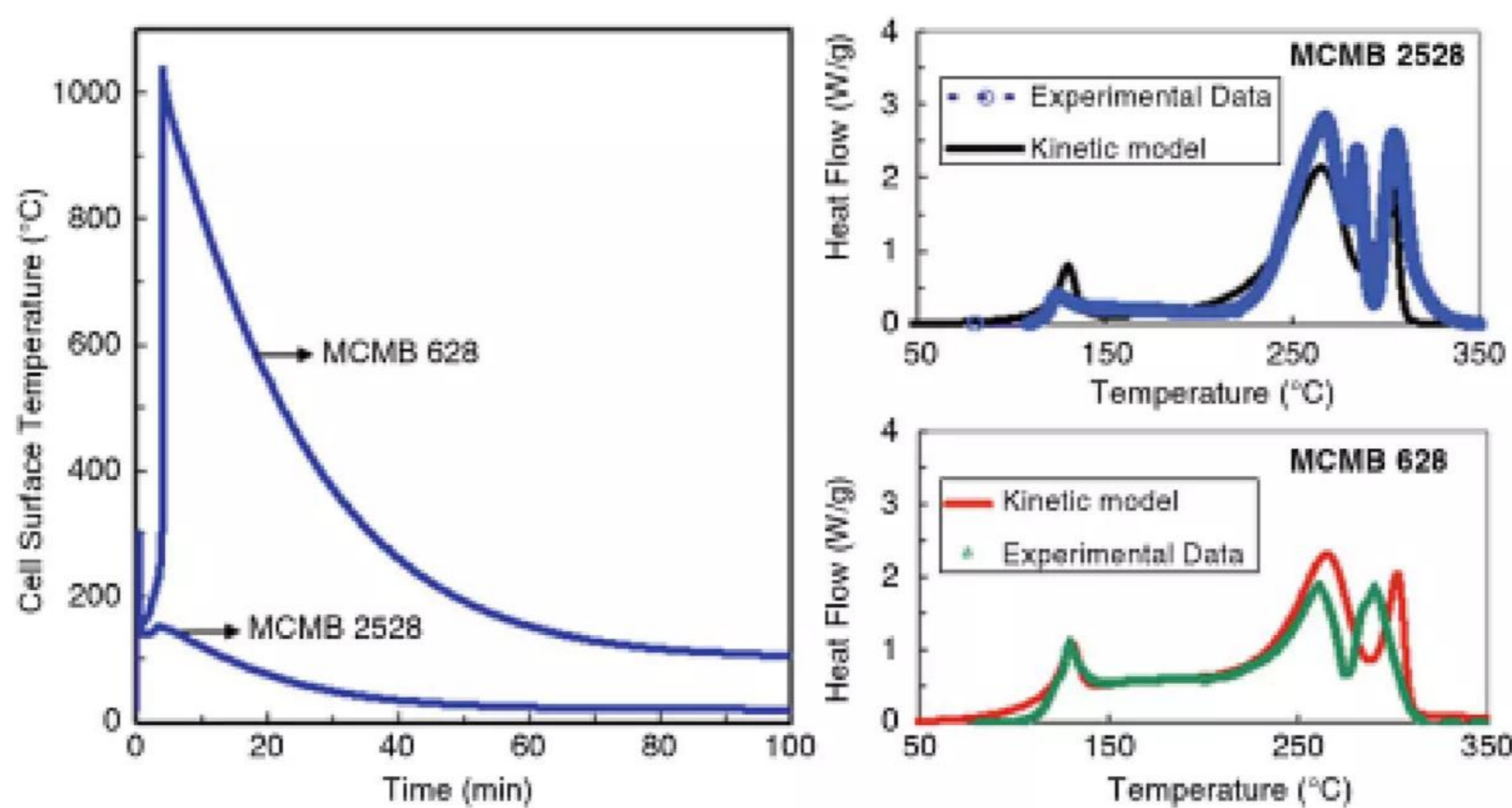


Fig. 9.14 Simulation results showing the effect of the choice of anode material on thermal runaway. The plot on the *left* shows the cell surface temperature time dependence for two different anode submodels

includes the relative volume of the jelly roll in which the temperature had risen past the point at which the separator would shut down (130°C).

The right hand charts in [Fig. 9.14](#) show DSC data for these two anode materials and the associated anode models that were employed in the simulations. The results of the simulations are shown on the left hand side of the figure. The results are striking – a simple change in anode material has a dramatic impact on the safety of this cell, with the cell employing MCMB 628 going into thermal runaway while the cell employing MCMB 2528 does not. DSC data on the right of [Fig. 9.14](#) suggest the reasons for this result. The MCMB 628 shows significantly higher heat release between 100°C and 200°C than the MCMB 2528. This example shows the extent to which the anode material acts as the trigger for thermal runaway when internal shorts and field-failures occur. Furthermore, the cell surface temperature profiles on the left side of [Fig. 9.14](#) illustrate another critical factor that is not well appreciated: the cell transitions rapidly into thermal runaway well below 200°C – well before any significant exothermic decomposition of cathode material has taken place.

Differences Between Abuse Tolerance and Field-Failures

The distinction between abuse tolerance and field-failure (internal shorts) is important for a number of reasons. It helps bring into focus the extent to which historic testing for safety is focused on abuse testing, even though the actual safety events that occur in the field are quite different. From a mechanistic perspective, the triggers are fundamentally different and the resulting responses

Abuse Tolerance	Field Failures
<ul style="list-style-type: none"> ♦ Predictable ♦ Common to all cells ♦ Can/should be evaluated at the cell level by standard tests ♦ Various chemistries can/should be evaluated for relative abuse tolerance ♦ Time constants relatively long, entire thermal mass of cell is heated uniformly ♦ Can be augmented by protection devices ♦ Externally-driven 	<ul style="list-style-type: none"> ♦ Not predictable, no warning ♦ One-in-a-million (or less) ♦ Difficult to evaluate at the cell level, or through QC ♦ Cell temperature is non-homogeneous, starting with "hot spot" ♦ Much higher temperatures can occur <i>quickly, locally</i> ♦ PTC, CID, shutdown separators, electronic controls are not effective – problem is internally driven.

Fig. 9.15 Summary of the key differences between abuse tolerance and field-failures

are different. The general distinctions between abuse tolerance and field-failure are summarized in [Fig. 9.15](#) and discussed below.

Field-failures are overwhelmingly attributable to internal short circuits that occur during otherwise "normal" operation, i.e., there is no external abuse or trigger. In contrast, heat exposure or forced overcharge are examples of an external abuse driving a potentially unsafe situation. The response to these abuse conditions should be identical for all cells of the same cell design employing the same materials.

In response to most abuses (note that penetration- and impact-based abuses are exceptions), a cell's temperature throughout an abuse is homogeneous, until the cell fails or the abuse is terminated. All cells of the same cell design and same materials should respond to an abuse in a roughly identical manner. In contrast, field-failure involves a nonhomogeneous temperature distribution in a cell's response to an internally driven trigger – the internal short, or hot spot.

The internal short represents a particularly insidious failure mechanism not only because it takes place during otherwise normal operation but also because it is self-driven and cannot be protected against by typical safety protection devices such as PTCs, CIDs, etc. It is also how most safety incidents are initiated in the field, and yet the battery industry does not have adequate tests to "predict" the risk for such failures.

The lack of availability of suitable tests for the internal short should not be a surprise – any event that takes place rarely, is stochastic *and* exhibits some form of incubation process, arguably does not exhibit a detectable precondition at the point of manufacture and certainly does not exhibit a predisposal to the problem *in every cell*. Thus, the practice of removing a few cells from the manufacturing line to test for safety is fine for abuse triggers (for abuse tolerance) but is clearly inappropriate as a measure of susceptibility to development of an internal short in the field.

Perhaps most challenging is the tension between the desire of those in the battery community to have a test that they can use to screen cells for field-failure (the internal short) and the statistical impossibility of picking cells to test that have the necessary preconditions for possible internal short-induced thermal runaway, putting aside the issue of the extent to which actual charge/discharge, voltage/time, and temperature effects may impact the probability of development of an internal short. That tension has led many across the battery and application/user communities to advocate that tests such as nail penetration, round bar crush impact tests, or other custom tests mentioned earlier, be employed to screen cells for tolerance to internal shorts. There are many reasons why this approach is an inappropriate surrogate for field-failure. One reason is that the condition that leads to internal shorts leading to thermal runaway is not present in fresh cells. In addition, it is statistically impossible to identify cells that actually have some form of precondition *and* for which that precondition has matured to the point at which an internal short exists. Most importantly, the nature of the internal shorts produced in such tests is nothing like that of the shorts involved in field-failures. These abuse tests often involve multiple shorts and hard shorts; conditions that are not representative of how internal shorts occur in the field. Both modeling and actual observations have shown that a number of different variable parameters can be critical to whether a given short will cause runaway in a given cell, and thus, the possibility of thermal runaway is a complex, multidimensional response surface. The single protocol, “one size fits all” approach of internal short circuit abuse tests means that such tests fix a number of these parameters and can therefore be gamed. Thus, relative safety under internal short circuit abuse test conditions is not likely to be relevant to the tendency for field-failure.

Nevertheless, the nail penetration test, the round bar crush test, and the drop test do reflect potential triggers (in this case, abuses) that must be considered, especially for transportation applications. In such applications, it is easy to imagine impact conditions that might lead to multiple internal shorts in cells. It is clearly desirable to understand the tolerance of cells to such abuses. The challenge, however, is to understand how representative such tests may be for the wide variety of impacts (speeds, angles, penetrations, etc.) that might occur in the field (putting aside the issue of potential gaming of the tests). Similar problems have been addressed with regard to transport of large quantities of flammable liquid in vehicles’ gasoline tanks.

This abuse tolerance versus field-failure framework is also a useful way to characterize testing work carried out by various researchers, or tests that purport to address safety. For example, various tests can be assessed with respect to whether they reflect or measure a property of all cells of the same design and materials (an abuse), or whether they reflect a rare condition that develops after time in the field.

Figure 9.16 also captures the difficult challenge of validating any new safety technology that is developed. How can one validate a safety technology intended to address events that occur on the order of once for every 5–10 million cells without producing and testing many millions of cells? The challenge is fundamental, and thus, it is important to have available means of producing cell/battery failures that are arguably “similar” to those that occur in the field. For these reasons, verification of the failure mechanism is also important.

Future Directions

Abuse tolerance in lithium-ion cells is critical to public safety, and a number of screening tests for lithium-ion cells and batteries have been devised. Battery developers and users then test cells and batteries to screen for tolerance with respect to the given abuse, and then design cells and batteries with improved abuse tolerance.

In contrast, safety in lithium-ion cells/batteries requires deliberate new strategies to deal with the possible occurrence of internal shorts that can progress to thermal runaway. These strategies need to recognize the extent to which the safety tests most typically utilized in the battery industry do not reflect the most common and most dangerous mechanism by which safety incidents actually occur in the field – field-failures.

The battery community does not utilize an overall strategy to intervene, for safety’s sake, as temperature rises in a lithium-ion cell or battery in advance of a thermal runaway. Although literature publications may consider the relative temperatures at which particular battery materials decompose, there is no consideration given to how these temperatures may be prevented.

The work presented in this chapter clearly shows that when thermal runaway occurs, it proceeds so fast that cell temperature progresses from less than about 150–180°C to over 600°C almost instantly. This result also indicates the fallacy in imputing safety advantages to one battery material that exhibits an onset temperature for decomposition (as captured, e.g., in DSCs) that is somewhat higher than that of another battery material (e.g., a cathode material with an onset temperature of 220°C is suggested to result in a safer cell than one with an onset temperature of 190°C). For example, the assumption is often made that cathode materials with higher onset temperatures will automatically make a cell safer or eliminate safety incidents. This work illustrates why that view has limited validity; first because anode decomposition is generally the lower temperature trigger for initiating thermal runaway subsequent to internal heating generated by an internal short, and second, because higher cathode decomposition temperature can at best increase the threshold energy for inducing thermal runaway (not the threshold power), but cannot eliminate the threat altogether.

Looking to the future, we suggest that abuse tolerance and field-failures be managed as distinct concerns. With both categories, a reasonable strategy involves

identifying and pursuing all efforts to prevent the final runaway process from maturing.

In that sense, a new approach to safety should also recognize one additional factor; namely that lithium-ion cells are no longer useful when their temperature exceeds about 100–110°C, and thereafter, they should/must be removed from service. Abuse tolerance can be further enhanced by focusing all efforts on identifying and shutting down all offending safety-related triggers by, for example, discharging the cell as soon as a cell temperature exceeds about 110–120°C. If an abuse/trigger is removed reasonably quickly, the cell simply cools down, and no safety event occurs.

In the case of field-failures (the internal short), this approach does not work because the cell skin temperatures may not reach these levels until thermal runaway is inevitable and/or in progress. Because it can be self-driven, thermal runaway from internal shorts must be detected during charge/discharge cycles prior to the final charge cycle that results in an internal short “mature” enough to lead to a thermal runaway. Therefore, it is imperative to identify early warning signals and to develop sensors that can warn of developing internal shorts well before a thermal runaway initiates and that are capable of integration at low cost into battery management systems.

We hope this chapter encourages the battery community to pursue more fundamental work into safety testing as well as understanding of safety-related processes in lithium-ion batteries. Work is needed to develop more sophisticated models, to measure material properties that contribute to enhanced safety, to develop improved methods of calorimetry, and to develop new safety technologies that will help assure safety of lithium-ion batteries. With greater visibility of the issues, perhaps this field will be taken up in graduate schools as an important topic that should yield rich problems and productive theses.

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Ralph J. Brodd *Editor*

Batteries for Sustainability

Selected Entries from the Encyclopedia of
Sustainability Science and Technology

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