**Image-based Modeling of Coupled Electro-Chemo-Mechanical Behavior of Li-ion Battery Cathode Using an Interface-Modified Reproducing Kernel Particle Method**

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**Abstract**

An interface-modified reproducing kernel particle method (IM-RKPM) is introduced in this work to allow for a direct model construction from image pixels of heterogeneous polycrystalline Li-ion battery microstructures. The interface-modified reproducing kernel (IM-RK) approximation is constructed through scaling of a kernel function by a regularized distance function in conjunction with strategic placement of interface node locations. This leads to RK shape functions with either weak or strong discontinuities across material interfaces, suitable for modeling various interface mechanics. With the placement of a triple junction node and distance-based scaling of kernel functions, the resulting IM-RK shape function also possesses proper discontinuities at the triple junctions. This IM-RK approximation effectively remedies the well-known Gibb’s oscillation in the smooth approximation of discontinuities. Different from the conventional meshfree approaches for interface discontinuities, this IM-RK approach is done without additional degrees of freedom associated with the enrichment functions, and it is formulated with the standard procedures in the RK shape function construction. This work focuses on identifying the accuracy and convergence properties of IM-RKPM for modeling the coupled electro-chemo-mechanical system. A linear patch test is formulated and numerically tested for the electro-chemo-mechanical coupled problem with a Butler-Volmer boundary condition representing the physical conditions in Li-ion battery microstructures. This is followed by verification of the optimal rates of convergence of IM-RKPM for solving the coupled problem with higher order solutions. The image-based modeling of Li-ion battery microstructures in the numerical examples demonstrates the applicability of the proposed method to realistic Li-ion battery materials modeling.

**Keywords**

*Reproducing Kernel Particle Method, image-based modeling, electro-chemo-mechanical coupling, interface-modified reproducing kernel approximation, Li-ion battery*

**Article Highlights**

* IM-RKPM for weak-strong discontinuities
* Coupled electro-chemo-mechanical patch test
* Image-based modeling of microstructures

# Introduction

Li-ion batteries are a predominant power source for modern life, renowned for powering everything from electric vehicles to consumer electronics. They must reliably withstand countless (dis)charging cycles throughout their lifetimes; however, these materials are vulnerable to lifetime capacity fade, fatigue, and degradation by electrode particle cracking with regular usage [1]. On the microstructure-scale, a Li-ion battery cathode is composed of many individual particles, each containing multiple grains, all covered in a binder layer for enhanced electrical activity [2–7]. Randomized grain orientations combined with anisotropic material properties lead to mismatches across grain boundaries and result in stress buildup during normal (dis)charging [2, 3].

Investigations of diffusion-driven stresses, analogous to thermal stresses, in Li-ion battery electrode particles have been modeled in the early days using finite differences with just one parabolic equation to model the electrochemical reaction [8]. The mechanical behavior of Li-ion battery cathodes, driven by an electrochemical reaction, is described by a set of highly coupled, nonlinear parabolic and elliptic partial differential equations [9–11]. At the macro-scale, full electrode or cell models, such as the multi-scale multi-domain (MSMD) model, have been proposed to account for subdomain geometries and local degradation resulting from heterogeneous electrostatic potential and thermal effects [12, 13]. The pseudo-two-dimensional (P2D) model has been introduced for electrochemistry calculation by leveraging porous electrode theory to model the electrode as spherical particles immersed in electrolyte with the transport treated as largely unidirectional [9, 14–17]. Additional work has further simplified the P2D model [18, 19]. Microstructural models, including electrode particle models, have been used to model the coupled electrochemistry iteratively and compute diffusion-based stresses on realistic microstructural meshes [20, 21]. These works, however, do not examine the convergence of numerical methods for solving the coupled multi-physics battery problems.

Previous methods to model the coupled chemo-mechanical behaviors at various scales largely rely on mesh-based methods, like the finite element method (FEM) [8, 20, 21]. Mesh-based methods become tedious in generating high-quality meshes for microstructure modeling, constructing approximations with arbitrary order of continuity and discontinuities, performing adaptive mesh refinement, and avoiding mesh entanglement in large deformation. For example, many mesh-based methods additionally require high levels of refinement to correctly model complex Li-ion battery material geometries [22–24]. To alleviate the aforementioned difficulties, an enhancement of the reproducing kernel particle method (RKPM) is introduced in this work for image-based modeling of coupled electro-chemo-mechanical behavior of Li-ion battery cathodes. RKPM [25–29] has shown to be effective for adaptive refinement [30], modeling as-built material microstructures [31–34], and for incorporating strong and weak discontinuities into the approximation [35–40, 34]. Some meshfree frameworks enrich solutions with special jump functions along material interfaces, but additional unknowns must be solved for [35]. Derivative discontinuity enrichment functions have been implemented for bi-material [36] and tri-material [37] interfaces through reproducing condition coupling, coined the interface-enriched RK approximation. Other methods also include a weighted reconstructing RK approximation [40], which constructs a high-order approximation by a locally-weighted sum of low-order RK approximations over multiple subdomains to selectively minimize the effects of the subdomain with the discontinuity and hence suppressing numerical oscillations.

In this work, the model encompasses two levels of coupling: full coupling of the lithium concentration and electrostatic potential fields as well as staggered coupling of the electrochemistry and mechanics. The electrochemical fields are coupled along the entire boundary through the Butler-Volmer interface condition, a mixed boundary condition which controls the reaction kinetics of the system [9–11]. An interface-modified RK (IM-RK) approximation is introduced for a natural embedment of discontinuities along grain boundaries and at triple junctions. This modification is made in the RK approximation without introducing enrichment functions and thus requires no additional degrees of freedom in solving PDEs. A linear patch test is formulated and numerically verified for the electro-chemo-mechanical coupled problem. Convergence properties of RKPM for the coupled problem with Butler-Volmer boundary conditions are evaluated. Finally, image-based modeling of Li-ion battery cathode microstructures with transient electro-chemo-mechanical coupling is investigated.

The paper is organized as follows. Section 2 introduces the governing equations of the coupled electrochemistry and mechanics under consideration. Section 3 presents an IM-RK shape function for modeling interface discontinuities. Discretization of the coupled problems and numerical implementation are detailed in Section 4. This is followed by numerical investigation of accuracy, convergence, and image-based modeling of Li-ion battery microstructures in Section 5. Concluding remarks are given in Section 6.

# Basic Equations

The multi-physics problem investigated in this work can be separated into two main categories: electrochemistry and elasticity, where the model equations are taken as from Plett [10]. The electrochemical part of the model considers the fully coupled lithium concentration, , and solid-phase electrostatic potential, , while the mechanical displacement, , and deformation is due to the chemically induced eigenstrain. Let be a domain with the spatial dimension and be the boundary of with the Dirichlet boundary and the Neumann boundary , where the Dirichlet and Neuman boundaries in each field are not necessarily consistent for all three fields in general, i.e. , where the second subscript , , refers to the field variables described above. This work considers only a single polycrystalline cathode particle surrounded by an electrolyte; although this electrolyte is not explicitly modeled, it is assumed to have constant concentration and potential. Individual grains exhibit random orientations and strong anisotropy in both diffusivity and mechanical expansion/contraction.

## Electrochemistry

### Lithium transport

Throughout charging and discharging a Li-ion battery, Li-ions move between the cathode and the anode and throughout the microstructures. The transport of solid-phase lithium is expressed as

|  |  |  |
| --- | --- | --- |
|  |  | (1) |
|  |  | (2) |
|  |  | (3) |

where is the lithium concentration field, is the outward surface normal vector, is the surface current density defined in (11), and is Faraday’s constant. Here, the surface flux is applied along the entire exterior of the particle, that is, .

The flux term, , is assumed to follow Fickian diffusion in equation (3), where is the concentration-dependent diffusivity tensor rotated to each grain’s principal orientation. In two dimensions, the tensor is taken as

|  |  |  |
| --- | --- | --- |
|  |  | (4) |
|  |  | (5) |

where , is a 2D rotational tensor, and (5) shows its Voigt notation, and is the angle of the principal grain orientation. Strong anisotropic diffusion is represented through a large ratio between and . The lithium concentration field is initialized as a constant value and is considered as the stress-free state. Because lithium diffusion is guided by the electric potential energy, the lithium transport equation is fully coupled with the electrostatic potential equation through the particle’s external boundary, as discussed in Section 2.1.3.

With the strong form of the lithium transport given in (1), (2), and (3), the corresponding weak form reads as: Given , find ,

|  |  |  |
| --- | --- | --- |
|  |  | (6) |

The dependence of in (11) with respect to the concentration and the electrostatic potential , as to be discussed in the next section, makes this a nonlinear equation. Another important feature of this system is that it is time dependent. The uniqueness of the electrochemical system of equation depends on the combination of the time dependent concentration equations and the Butler-Volmer boundary condition (11). Uniqueness is discussed further in Section 2.1.4.

### Solid-phase electrostatic potential

The scalar electrostatic potential, , corresponds to the amount of work needed to move a unit charge against an electric field from a reference point to a specified point and is governed by the following Poisson equation:

|  |  |  |
| --- | --- | --- |
|  |  | (7) |

where is the solid-phase conductivity, which is taken as a constant value. The following flux boundary term is applied along the entire external boundary of the cathode particle:

|  |  |  |
| --- | --- | --- |
|  | . | (8) |

As mentioned before, is the surface current density and is the outward surface normal vector. Equations (2) and (8) are coupled by this term through the Butler-Volmer interface condition discussed in the next section. The applied current density, , is a parameter that dictates the rate of reaction by offsetting the current density in the coupled electrochemistry. This applied current density is dependent on the charge rate, , a normalized measure of how fast a battery is charging or discharging, as shown below:

|  |  |  |
| --- | --- | --- |
|  |  | (9) |

where is the maximum theoretical lithium concentration, is Faraday’s constant, 3600 is the number of seconds in an hour, and these quantities are normalized by the volume-to-surface-area ratio of the particle.

For the electrostatic potential strong form given in equations (7) and (8), the corresponding weak form reads as: Given , find ,

|  |  |  |
| --- | --- | --- |
|  |  | (10) |

### Again, the dependence of in (11) with respect to the concentration and the electrostatic potential makes (6) and (10) a coupled nonlinear system to be solved simultaneously by Newton’s method, as discussed in Section 2.3 and Section 4. Even though the potential equation has pure Neuman boundary condition, uniqueness is maintained though the coupling to the lithium transport equation (6) via the Butler-Volmer equation (11).

### Butler-Volmer reaction kinetics

The Butler-Volmer interface condition governs the electrochemical reaction kinetics that take place between the electrolyte and immersed cathode particle, and in effect couples the lithium concentration conservation and electrostatic potential balance. The highly nonlinear Butler-Volmer reaction with respect to concentration and electrostatic potential , acts along the cathode particle exterior and is expressed as:

|  |  |  |
| --- | --- | --- |
|  |  | (11) |

In the above equation, is the current density, is the concentration-dependent exchange current density given in Appendix A, and are rate control parameters at the anode and cathode respectively, is Faraday’s constant, is the universal gas constant, and is temperature. Much of the complexity and ill-conditioned nature of the electrochemical system is due to the high nonlinearity of this boundary condition. The Butler-Volmer relation in (11) defines the flux boundary conditions presented in equation (2) and equation (8). Aside, when , the Butler-Volmer relation simplifies to . For this work, a constant temperature with is assumed. The Butler-Volmer kinetics depend on both the electrostatic potential and concentration fields along the external particle boundary with overpotential, , expressed as

|  |  |  |
| --- | --- | --- |
|  |  | (12) |

where is the concentration-dependent equilibrium potential given in Appendix A and is the theoretical maximum Li-ion concentration for the electrode material. The Butler-Volmer relation is a nonlinear Neumann condition where the surface flux is a function of electrostatic potential and concentration fields. Recall that this boundary type is distinct from the traditional Robin mixed-type boundary condition which linearly combines Dirichlet and Neumann boundary terms.

### Uniqueness of the electrochemical system

Due to the lack of essential boundary conditions, it is not immediately obvious that the electrochemical system given in (1)-(10) has a unique solution, and, while a full proof of uniqueness for nonlinear Robin-type boundary conditions for coupled systems is a bit out of scope for this paper, a heuristic argument is provided.

First, recognize that the diffusion equation with pure Neumann boundary conditions is unique given a proper initial condition. Second, the Poisson equation is unique up to a constant with pure Neumann boundary conditions. The diffusion and Poisson equations are analogs to the lithium transport and potential equations if the Butler-Volmer boundary condition is excluded. The coupling between these two systems provided by the Butler-Volmer boundary condition is enough to pin down the constant in the potential equation.

This is true as long as the linearized version of the Butler-Volmer equation is included in Newton’s method. If the previous iteration’s concentration and potential are used to fix the Butler-Volmer to a constant value (reducing it to a Neumann and not a Robin condition), then the system is no longer unique. Likewise, if the time dependent term is removed from the lithium transport equations. This is the reason for the non-uniqueness of the linear patch test in Section 5.1.

In summary, the combination of time dependence of the conservation of Li-ions and nonlinear boundary coupling ensure solution uniqueness, despite the fully Neumann boundary, see [41] for details.

### Nonlinear electrochemical material properties

Many of the electrochemical material properties exhibit strong nonlinearity and concentration dependence. Nonlinear material profiles with respect to normalized concentration have been adopted for the lithium diffusivity [42], equilibrium potential [43], and exchange current density [43], as shown in **Fig. 1** and expressed in Appendix A. These functions are based on experimental data and approximated using a high order interpolating polynomial.

Chart, line chart, histogram

Description automatically generated

**Fig. 1** Nonlinear, concentration dependent material property profiles: (a) diffusivity , (b) equilibrium potential , and (c) exchange current density

## Mechanical deformation

### Diffusion-induced elastic deformation

Consider the following diffusion-induced linear elasticity subjected to diffusion-induced strain :

|  |  |  |
| --- | --- | --- |
|  |  | (13) |
|  |  | (14) |
|  |  | (15) |

where is the stress tensor, is the material constitutive tensor, and is elastic strain defined as:

|  |  |  |
| --- | --- | --- |
|  |  | (16) |
|  |  | (17) |
|  |  | (18) |

where is the diffusion-induced strain tensor, is the lithium concentration at the stress-free state, which for this paper is taken as the initial lithium concentration, and . Traction-free boundary conditions are employed over the full boundary to allow free expansion in the domain, and eigenvalue shifting method is employed to suppress rigid body modes. In 2D, the anisotropic tensor is taken as

|  |  |  |
| --- | --- | --- |
|  | (no sum on ). | (19) |

Stresses are induced purely due to the grain misalignment and anisotropic expansion and contraction.

The weak form of the elastostatic balance equation is given below: Given , find ,

|  |  |  |
| --- | --- | --- |
|  |  | (20) |

### Nonlinear mechanical material properties

The expansion of Li-ion cathode materials on the continuum scale is correlated to the measured expansion of unit cell lattice parameters. The expansion coefficients and are computed as follows:

|  |  |  |
| --- | --- | --- |
|  |  | (21) |
|  |  | (22) |

where and act along the principal and secondary grain orientations, respectively. The expansion coefficients are computed based on the normalized change in lattice parameters and , whose nonlinear concentration-dependent curves are visualized in **Fig. 2** and detailed further in Appendix A [44]. These functions are based on experimental data and approximated using a high order interpolating polynomial. The change in lattice parameters and are taken with respect to the initial lattice parameters and measured at the initial stress-free Li-ion concentration , found in Table 2. For all cases, plane strain is assumed.

Chart, line chart

Description automatically generated

**Fig. 2** Nonlinear, concentration dependent lattice parameters in are used to determine expansion in the (a) primary and (b) secondary lattice directions

## Incremental equations

The nonlinearities in the aforementioned conservation equations are solved by Newton’s method, where the incremental weak forms are discussed below. Recall the lithium diffusion and electrostatic potential weak forms; they are restated below for convenience:

|  |  |  |
| --- | --- | --- |
|  |  | (23) |
|  |  | (24) |

Define the following terms with the iteration counter :

|  |  |  |
| --- | --- | --- |
|  |  | (25) |
|  |  | (26) |
|  |  | (27) |
|  |  | (28) |
|  |  | (29) |

The lithium diffusion transport equation and its corresponding incremental equation are

|  |  |  |
| --- | --- | --- |
|  |  | (30) |
|  |  | (31) |

Similarly, the electrostatic potential and its corresponding incremental equation are

|  |  |  |
| --- | --- | --- |
|  |  | (32) |
|  |  | (33) |

where

|  |  |  |
| --- | --- | --- |
|  |  | (34) |
|  |  | (35) |
|  |  | (36) |
|  |  | (37) |
|  |  | (38) |

The IM-RK approximation and discretization (given in Section 3) are introduced in the incremental equations (23)-(38). The resulting matrix equations of the coupled problem are detailed in Section 4.

# Interface-Modified RK Shape Function for Modeling of Polycrystalline Material Microstructures

Smooth approximation functions, like the standard RK shape functions with smooth cubic B-spline kernels, suffer from Gibbs oscillations when approximating discontinuities. As such, RK shape function modification is needed to properly address discontinuities across grain boundaries. Many enrichment strategies have been previously proposed, as seen in [34, 37–39]. In this work, the discontinuities are introduced into the RK approximation through a simple scaling of kernel functions near material interfaces. Together with discretization point placement strategies at interfaces and an imposition of reproducing conditions, weak and strong discontinuities can be embedded in the RK shape functions, and the extension to general multi-dimensions is straightforward.

To apply an interface modification to RK shape functions, as will be discussed in Section 3.1, grain boundary identification and discretization are first considered. In this approach, the distance-based kernel function scaling is introduced [34]. In this work, interface node placement is further introduced to yield RK approximation functions with weak or strong discontinuities at material interfaces as well as at triple junctions in the grain microstructures. The overall representation of the proposed IM-RK approximations to be constructed is illustrated in **Fig. 3**.

## Reproducing kernel approximation

Consider a -dimensional domain discretized by points with nodal coordinates for node , as illustrated in **Fig. 3**.

Diagram

Description automatically generated

**Fig. 3** RK discretization with overlapping supports, RK shape function for bulk nodes, and IM-RK shape functions for grain boundary nodes and neighboring nodes

The reproducing kernel (RK) approximation is constructed such that a function is approximated as

|  |  |  |
| --- | --- | --- |
|  |  | (39) |

Here, is the RK shape function corresponding to node evaluated at , and is the generalized nodal coefficient of node . The RK shape function, , is a correction of a kernel function, , which is defined over a compact support, , in the multi-dimensional form as:

|  |  |  |
| --- | --- | --- |
|  | . | (40) |

The correction function, , is expressed as a linear combination of basis vectors with coefficients , where with , , and the basis functions are expressed in a vector given as

|  |  |  |
| --- | --- | --- |
|  | . | (41) |

The following reproducing conditions are introduced to solve for :

|  |  |  |
| --- | --- | --- |
|  |  | (42) |

which can be equivalently written as

|  |  |  |
| --- | --- | --- |
|  |  | (43) |

Substituting the RK shape function definition (40) into the reproducing equations (43), the coefficient vector is obtained as:

|  |  |  |
| --- | --- | --- |
|  |  | (44) |

with the moment matrix defined as

|  |  |  |
| --- | --- | --- |
|  |  | (45) |

and . After substituting back into (40), the RK shape function is obtained,

|  |  |  |
| --- | --- | --- |
|  | . | (46) |

In the RK approximation described above, the kernel function determines the continuity in the approximation, while the order of monomial basis determines the order of completeness in solving PDEs. Unless otherwise specified, the following cubic B-spline kernel function is used:

|  |  |  |
| --- | --- | --- |
|  |  | (47) |

It is noted that the RK shape functions do not necessarily possess the Kronecker delta properties Imposition of essential boundary conditions are achieved with additional effort, such as the commonly used Nitsche’s method [45, 29]. In this work, the modification of RK shape functions to be discussed in the next section also offers a convenient means for imposing essential boundary conditions if the prescribed function value on the boundary is a constant field.

## Interface-modified kernel functions

Let be the set of all RK nodes and be the set of interface nodes. First, the RK kernel functions are modified by a regularized Heaviside function associated with the material interface [37]:

|  |  |  |
| --- | --- | --- |
|  |  | (48) |

where is the scaled kernel function, is the regularized Heaviside scaling function, and is the distance function from evaluation point to the nearest material interface. The regularized Heaviside function is taken as

|  |  |  |
| --- | --- | --- |
|  |  | (49) |

with Macaulay brackets signifying . Note that kernels associated with the interface nodes are not scaled with and remain unmodified, as shown in **Fig. 4**.

Chart, diagram

Description automatically generated

**Fig. 4** Scaled 1D kernel functions and nodes (black) around interface at x=0.5 (dashed line): (a) cubic B-spline kernels, (b) two hyperbolic tangent kernel scaling functions, and , and (c) scaled kernel functions

It is easily observed that the domains of influence of all kernel functions except the kernel functions associated with the interface nodes terminate at the interface.

## Interface-modified RK (IM-RK) approximation

The interface modified RK shape function is constructed as follows [34]:

|  |  |  |
| --- | --- | --- |
|  |  | (50) |

The coefficient vector, , is obtained by applying the reproducing conditions in equation (43) by following a similar procedure as in equations (44) and (45):

|  |  |  |
| --- | --- | --- |
|  | . | (51) |

is the modified moment matrix, defined as

|  |  |  |
| --- | --- | --- |
|  |  | (52) |

Consequently, the IM-RK shape functions take the following form by substituting the modified moment matrix and coefficient vector back into equation (50) to yield:

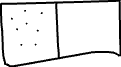
|  |  |  |
| --- | --- | --- |
|  |  | (53) |

Since interface modified kernels terminate along the interface, careful consideration must be taken to ensure each evaluation point near the interface is covered by sufficiently many non-collinear kernels (kernel function centers not along the same line) to avoid moment matrix singularity [29]. Due to the minimal kernel coverage condition, three options can be considered for the modified moment matrix inversion in equation (53): (i) a pseudo-inverse, (ii) a quasi-linear RK approximation [46], or (iii) regularization of scaling function: to barely include nodes near the interfaces in the kernel support coverage to avoid moment matrix singularity. Here, the third option is employed.

Due to the regularization , the derivatives of the IM-RK shape functions can be computed directly as follows:

|  |  |  |
| --- | --- | --- |
|  |  | (54) |

where



|  |  |  |
| --- | --- | --- |
|  |  | (55) |

Let , , , , , and is the material interface. The weak and strong discontinuities on in the IM-RK shape function can be achieved by the placement of RK discrete points at .

*Case 1*. with weak discontinuity for .

This is achieved by placing RK discrete points on , that is, . It can be easily shown that since for ,

for , , ,

for according to (52),

for , , according to (53).

*Case 2.* with strong discontinuity on .

This is achieved by letting , that is, without placing RK discrete points on . Since no RK discrete points are placed on , let , we have

,

,

where and are positions adjunct to the opposite sides of . Since, then , and hence. Therefore, , .

It is also obvious to show that by placing RK discrete nodes on , defined in (52) remains continuous on , hence , as shown in Case 1.

Diagram

Description automatically generated

**Fig. 5** Comparison of standard RK and IM-RK shape functions with weak discontinuity in 1D: (a) standard RK shape functions with unscaled cubic B-spline kernels, (b) corresponding RK shape function derivatives, (c) IM-RK shape functions with scaled kernel to yield a weak discontinuity at , and (d) corresponding IM-RK shape function derivatives

Placing a node on the interface, a demonstration of the IM-RK shape function with weak discontinuity on the interface is provided in **Fig. 5**. Note that IM-RK shape functions still maintain the partition of unity and polynomial reproducibility properties. Due to the termination of all neighboring kernel functions at the material interface, the enforcement of reproducing conditions forces the interface RK shape function to exactly recover the Kronecker delta property in this one-dimensional case and simultaneously introduce a weak discontinuity at the interface. Notably, the IM-RK approximation introduces a weak discontinuity into the shape functions even when smooth cubic B-spline kernels are used.

Chart

Description automatically generated

**Fig. 6** Supports, shape functions, and shape function derivatives of IM-RK shape functions with weak discontinuities associated with grain boundary neighboring nodes (yellow and blue) and grain boundary node (black): (a) bird’s eye view of IM-RK supports, (b) rotated view of IM-RK shape functions, (c) x-derivative of IM-RK shape functions, (d) y-derivative of IM-RK shape functions

The two-dimensional IM-RK shape functions with placement of nodes on the grain boundaries and triple junctions yield weak discontinuities near a grain boundary as illustrated in **Fig. 6** and near a triple junction as shown in **Fig. 7**. Regarding triple junction IM-RK shape function construction, two aspects are carefully considered: the distance and moment matrix calculations. First, the distance by closest point projection is always unique due to the grain convexity at triple junction locations. Second, to avoid moment matrix singularity at the triple junction, kernel support sizes of the surrounding grain boundary points are chosen to ensure sufficient kernel coverage at triple junctions.

Chart

Description automatically generated

**Fig. 7** Supports, shape functions, and shape function derivatives of IM-RK shape functions with weak discontinuities associated with grain triple junction neighboring nodes (yellow, blue, pink), triple junction node (black), and grain boundary nodes (purple green, cyan): (a) bird’s eye view of IM-RK supports, (b) rotated view of IM-RK shape functions, (c) x-derivative of IM-RK shape functions, (d) y-derivative of IM-RK shape functions

Note that in two dimensions, the Kronecker delta properties of the IM-RK shape functions are not fully recovered on the domain boundary due to the coverage of multiple kernel functions associated with the boundary points. Nevertheless, it can be shown that a constant boundary value can still be directly imposed by the IM-RK shape functions with constant nodal coefficients.

## Arbitrary weak and strong discontinuity in the IM-RK approximation

In this section, it is further demonstrated how the weak and strong discontinuities can be achieved naturally in the IM-RK approximation by using the kernel scaling and proper placement of discretization points at the material interface. Let in a domain be the material interface where a weak or strong discontinuity is to be generated. Two domain discretizations are considered: (i) with a node placed at the interface location and (ii) without a node placed at the interface location. In both cases, cubic B-spline kernel functions are used for all nodes in IM-RK shape function construction as shown in **Fig. 8**.

Chart, diagram, histogram

Description automatically generated

**Fig. 8** The RK nodes (black dots) and the associated smooth kernel functions in a domain with an interface at (dashed line): (a) a node is placed at the interface location and (b) no node is placed at the interface location

The IM-RK approximation procedures presented in Section 3.3 are followed to construct the IM-RK shape functions for both discretization cases. As seen in **Fig. 9**, by controlling the node placement at the interface, the different types of discontinuity in the resulting IM-RK approximation are achieved. An IM-RK approximation with weak discontinuity is obtained when a node is placed right on the interface, while an IM-RK approximation with strong discontinuity is achieved when no node is placed at the interface.

To demonstrate the effectiveness of IM-RK shape functions for approximating weak and strong discontinuities, two target functions (TF1 and TF2) are considered for a least-squares-based function approximation using IM-RK shape functions. The target functions feature distinct types of discontinuities at

Diagram, histogram

Description automatically generated

**Fig. 9** IM-RK shape function construction: (a) kernel scaling function, (b) scaled kernels, and (c) IM-RK shape functions with weak and strong discontinuities obtained using discretizations with and without node placement at the interface location (dashed line)

Both the IM-RK shape functions with weak discontinuity and strong discontinuity can accurately represent the weak discontinuity in TF1 to machine precision error as seen in **Fig. 10**, whereas standard RK shape functions generate error on the order of . For the strong discontinuity in TF2, only IM-RK with strong discontinuity can represent the strong discontinuity to machine precision; both standard RK and IM-RK with weak discontinuity yield errors on the order of , as depicted in **Fig. 11**. A plate with circular inclusion modeled by IM-RK with weak discontinuity is demonstrated in Appendix B. Note that for solving PDEs with weak discontinuities, employment of IM-RK with strong discontinuities can be used but needs to be implemented in conjunction with Nitsche’s method [45] to enforce compatibility on the material interface. In this work, IM-RK with weak discontinuities are employed for Li-ion battery microstructure modeling as discussed in Section 5.

Diagram, engineering drawing

Description automatically generated

**Fig. 10** Target function 1 approximation and error comparison using standard RK, IM-RK with weak discontinuity, and IM-RK with strong discontinuity

# Numerical Implementation

In this work, IM-RK with weak discontinuity is employed for modeling discontinuous gradients in lithium concentration, electrostatic potential, and displacements in Li-ion battery microstructures. Let the field variables and their increments be approximated by the IM-RK shape functions

|  |  |  |
| --- | --- | --- |
|  |  | (56) |
|  |  | (57) |
|  |  | (58) |

Diagram

Description automatically generated with low confidence

**Fig. 11** Target function 2 approximation and error comparison using standard RK, IM-RK with weak discontinuity, and IM-RK with strong discontinuity

where is the IM-RK approximation function. The respective gradient fields are expressed as:

|  |  |  |
| --- | --- | --- |
|  |  | (59) |
|  |  | (60) |
|  |  | (61) |

where the gradient matrices are given as follows:

|  |  |  |
| --- | --- | --- |
|  |  | (62) |

Using backward Euler for time integration of the lithium transport equation, and considering the nonlinearities of with respect to and of with respect to and , the incremental form of the coupled electrochemistry equations in (31) and (33) at iteration of time step is given as:

|  |  |  |
| --- | --- | --- |
|  |  | (63) |

where

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  | |  | (64) | |
|  | |  | (65) | |
|  | |  | (66) | |
|  | |  | (67) | |
|  | |  | (68) | |
|  | |  | (69) | |
|  | |  | (70) | |
|  | |  | (71) | |
|  |  | | | (72) | |

The maximum Li-ion concentration, , is given in Table 2. Since the nonlinearity of this problem entirely stems from the electrochemistry model and the mechanical equilibrium considered is linear, the corresponding mechanical matrix equation is separately given as follows:

|  |  |  |
| --- | --- | --- |
|  | *,* | (73) |

where

|  |  |  |
| --- | --- | --- |
|  |  | (74) |
|  |  | (75) |

As previously mentioned, the electro-chemo-mechanical model is separated into two sub-models with the solution workflow illustrated in **Fig. 12**. First, the electrochemical field variables are initialized as constant fields. These electrochemical field initializations are selected by defining a constant concentration representative of a specified charge state in the battery material (0-100% of ) and using the Butler-Volmer interface condition to back calculate the corresponding electrostatic potential initialization. The transient Li-ion conservation equation is advanced in time and a Newton-Raphson iteration is used to solve the incremental fully coupled equations until a convergence tolerance is achieved for each incremental field variable. Once the convergence criteria are sufficiently satisfied, the concentration field is passed to the mechanical model in a staggered manner to compute the chemically induced displacements and strains. This process is repeated until the final time is reached.

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**Fig. 12** Solution procedure flowchart

# Numerical Results

In this section, the convergence properties of RKPM for solving the static electro-chemo-mechanical coupled problem is first verified by solving a linear patch test and a manufactured problem. The performance of IM-RKPM is further examined by modeling a pixel image of a Li-ion battery microstructure. Unless specified otherwise, RK and IM-RK shape functions are constructed using cubic B-spline kernels with circular support and normalized support side of , linear basis, with convergence tolerances of for each fields’ nodal coefficient increments. The stiffness matrices and force vectors are integrated with 3x3 Gauss quadrature in the following test problems, except for the linear patch test, where 10x10 quadratures are used to pass patch test. Backward Euler is used for time integration of the lithium transport equation.

## Linear patch test for coupled static electro-chemo-mechanical fields

The linear patch test here refers to the verification of a numerical method’s ability to obtain a linear solution exactly if the exact solution of the three coupled fields is linear. This should not be confused with the fact that the coupled electro-chemo-mechanical system remains nonlinear due to the Butler-Volmer boundary conditions. As such, the solutions are obtained iteratively. Passing the linear patch test is a sufficient condition for achieving a 2nd order rate of convergence in error norms and a 1st order rate of convergence in error norms. The governing equations and the associated boundary conditions are defined based on the manufactured linear, time-independent solutions for lithium concentration(), electrostatic potential (), and displacements (). To ensure uniqueness of the static coupled problem with Neumann-type Butler-Volmer boundary conditions, an eigenvalue shifting technique is employed to suppress arbitrary constant eigenmodes.

In the following, the manufactured polynomial fields are denoted with a superscript and with arbitrary coefficients in a domain as follows:

|  |  |  |
| --- | --- | --- |
|  |  | (76) |
|  |  | (77) |
|  |  | (78) |
|  |  | (79) |

The flux boundary conditions for the electrochemical fields given in equations (2) and (8) are prescribed with the following manufactured boundary fluxes:

|  |  |  |
| --- | --- | --- |
|  |  | (80) |
|  |  | (81) |

where is computed by evaluating equation (11) with and . For this test, material properties are taken as: , , and and follow nonlinear profiles detailed in Appendix A. Faraday’s constant is taken as . The manufactured solutions are selected such that they are physically representative leading to stable treatment of the exponential in the Butler-Volmer equation:

|  |  |  |
| --- | --- | --- |
|  |  | (82) |
|  |  | (83) |

In crafting the boundary conditions of this coupled patch test, the following conditions are considered: (i) boundary conditions are Neumann-type to reflect the Butler-Volmer relation and (ii) boundary conditions are consistent with the manufactured fields. Both and are solved simultaneously using a Newton iteration of the incremental equations given in equations (31) and (33), with constant initial guesses of and .

Once the Newton iteration converges, the updated concentration field is used to compute the chemically induced eigenstrain for the mechanical equation to solve for the corresponding displacement field. With the expansion coefficients of and , the analytical displacement field is:

|  |  |  |
| --- | --- | --- |
|  |  | (84) |

The RK discretization with 11x11 nodes in a domain is considered. 10x10 Gauss integration is employed for error norms and force vector and stiffness matrix integration. The normalized error norms for function and the approximated are defined as follows:

|  |  |  |
| --- | --- | --- |
|  |  | (85) |
|  |  | (86) |

The normalized error norms for vector function and the approximated are defined as follows:

|  |  |  |
| --- | --- | --- |
|  |  | (87) |
|  |  | (88) |

where is the normalized error norm and is the normalized error semi-norm, respectively. The errors of all three fields and their respective gradients are shown in Table 1.

Table 1. Coupled patch test normalized error norms

|  |  |  |
| --- | --- | --- |
| Field |  |  |
|  |  |  |
|  |  |  |
|  |  |  |

**Remark 4.1**

Incorporation of the exponential Butler-Volmer boundary condition requires proper selection of material parameters to avoid nonphysical environments that could lead to solution iteration divergence. As such, physical material profiles and constants are used for variables involved directly in the Butler-Volmer expression (. Since diffusivity and conductivity are not involved in the Butler-Volmer calculation, their values can be arbitrarily selected.

## Convergence properties

A problem with a higher-order manufactured solution is designed herein to study the convergence properties of IM-RKPM in solving the coupled system over the domain using the following manufactured fields:

|  |  |  |
| --- | --- | --- |
|  |  | (89) |
|  |  | (90) |

where , , , and .Again, these values were chosen to avoid extremely large, nonphysical gradients in the Butler-Volmer expression in equation (11). The corresponding boundary conditions are applied following equations (80) and (81). Eigenvalue shifting is employed for removal of zero energy modes. Isotropic diffusion is considered with constant , , and and are prescribed following nonlinear profiles described in **Fig. 1** and Appendix A. Here, is computed with , and the corresponding manufactured displacement field takes the following form:

|  |  |  |
| --- | --- | --- |
|  | . | (91) |

Calendar

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**Fig. 13** IM-RKPM solutions with : (a) , (b) , (c) , and (d)

**Chart, line chart

Description automatically generatedFig. 14** Convergence plots for relative error norms of all field variables: (a) norm and (b) semi-norm . At this scale, all lines in (b) overlap

Rigid body motion is suppressed in the displacement field allowing for free expansion. For all cases explored, and is selected. A relatively large target function frequency is selected to simulate higher frequency modes in the multi-grain microstructures. This convergence study includes cases with 256, 1024, 4096, and 16384 nodes. In the following study, the frequency parameters in (91) are chosen, and the IM-RKPM results using 16384 nodes are shown in **Fig. 13**.

For all fields, super-convergent rates are observed for relative error norms and semi-norms, as shown in **Fig. 14**. Due to the chemically-induced eigenstrain, error accumulation is expected from the concentration field to the displacement field, where larger error in the mechanical field, , compared to that of the electrochemical fields, and is observed.

## Mechanical modeling of heterogeneous microstructure

Statistically generated cathode microstructural images constructed from a database of CT-scan images [4, 47] are used for model construction. A coarsened image with 51x51 pixels is taken from the test particle geometry illustrated in **Fig. 15**, spanning a domain where each grain is assigned a unique grayscale value as the grain ID.

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**Fig. 15** Statistically-driven cathode particle microstructural image of a (a) 362x362 pixel cross section of the full particle representation generated using the procedure in [47] and a (b) test particle constructed from a 51x51 pixel subset of the full particle image

**Fig. 16** displays the image-based geometry colorized by grain ID subjected to a vertical displacement at the top surface, free horizontal deformation at top and bottom surfaces, and a fixed bottom surface midpoint to suppress rigid body motion. The elastic modulus and Poisson ratio are varied across the different grains as shown in **Fig. 16** to introduce material heterogeneity. Essential boundary conditions are enforced through Nitsche’s method [45] using a normalized penalty parameter of , where is the maximum Young’s modulus, and is the maximum kernel support size associated with the essential boundary.

Diagram

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**Fig. 16** A heterogeneous image-based test particle with varying elastic modulus and Poisson ratio is loaded under the prescribed displacement conditions

Map

Description automatically generated with medium confidence

**Fig. 17** Colorized discretization of image-based microstructure with zoomed-in view near two triple junctions with (a) a highly refined grain-boundary-fitted FEM mesh and (b) a coarse nodal distribution for RKPM and IM-RKPM

A highly refined grain-boundary-fitted FEM mesh with 4-node bilinear elements is constructed based on the raw image pixels to conform to grain boundary node locations. This mesh, shown in **Fig. 17**, contains 62,852 elements and 63,354 nodes, and its FEM solution is taken as the reference for RKPM and IM-RKPM solutions. For the purpose of comparison with the FEM solution, 4,195 points obtained from a much coarser mesh are used for RKPM and IM-RKPM discretization as shown in **Fig. 17**. Adaptive refinements near triple junctions are introduced in the RKPM discretizations. It is noted that the FEM’s highly refined mesh leads to a considerably smoother boundary representation.

A picture containing graphical user interface

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**Fig. 18** Displacement field predictions in using FEM, RKPM, and IM-RKPM

**Fig. 18** shows that all methods yield similar displacement distributions. Strain and stress distributions are plotted in **Fig. 19** and **Fig. 20**, respectively. Overall strain jumps across grain boundaries in RKPM are slightly diffused. On the contrary, IM-RKPM sharply captures strain discontinuities across the grain boundaries despite the coarse nodal spacing used, yielding good agreement with the highly refined FEM solution.

Graphical user interface

Description automatically generated

**Fig. 19** Strain field predictions using FEM, RKPM, and IM-RKPM with zoomed-in views of two triple junction regions

All methods predict stress singularities at the triple junctions. Stress concentrations near triple junctions and other sharp corners are well captured while using IM-RKPM. This verification study illustrates how IM-RKPM can be used with a coarse discretization to obtain results on par with those of a highly-refined FEM solution. Due to the employment of FEM approximation, FEM stress solutions are less smooth than those obtained by IM-RKPM.

Graphical user interface

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**Fig. 20** Stress field predictions in using FEM, RKPM, and IM-RKPM with zoomed-in views of two triple junction regions

## Image-based electro-chemo-mechanical modeling of Li-ion battery cathode microstructure

A comparison of the standard RKPM and IM-RKPM methods is demonstrated through a coupled transient electro-chemo-mechanical simulation of a realistic microstructure. The 51x51 pixel image of the test particle geometry shown in **Fig. 15** is again used for model verification. For the discretization, the pixel points are used as the RKPM discretization points with given material grain IDs corresponding to the pixel value in the image. Grain boundary nodes are assumed at the midpoint between two adjacent pixels with different grain IDs as illustrated in **Fig. 21**. Particle boundary nodes are assigned along the entire exterior of the test microstructure. Denoting grain boundary nodes enables introduction of weak discontinuities into the IM-RK shape functions as discussed in Section 3. The distance function in equation (48) is computed for all evaluation points with respect to their nearest grain boundaries. Additional “dummy” boundary nodes were added in-between existing boundary nodes for a more accurate distance function calculation, but they are not included in the shape function construction nor the degrees of freedom in the solution. Gauss integration cells are constructed between adjacent nodes and conform to the grain boundaries using triangular integration cells.

Chart

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**Fig. 21** Grain boundary nodes (black filled markers) separating adjacent grains with their ID assignments shown by color for pixel points (colored filled markers).

Principal material orientations are assigned for each grain, as depicted in **Fig. 22**. Strong anisotropic diffusion is represented by Anisotropic mechanical expansion coefficients are defined in equations (21) and (22) with nonlinear material profiles given in Appendix A. The lithium concentration and electrostatic potential fields are initialized with constant values and , considered as the stress-free state. For this example, , although traditionally this value is dependent on the charge rate (C-rate) as well as the geometry of the electrode. Additional electrochemical material constants and ambient conditions are summarized in Table 2. An elastic modulus of and Poisson ratio of are used. The coupled electrochemical solution converges in 4 Newton iterations for nearly all time steps. The simulation is run until with a time step of . Eigenvalue shifting is used to suppress the rigid translational and rotational mechanical modes.

Table 2. Summary of electrochemical material parameters

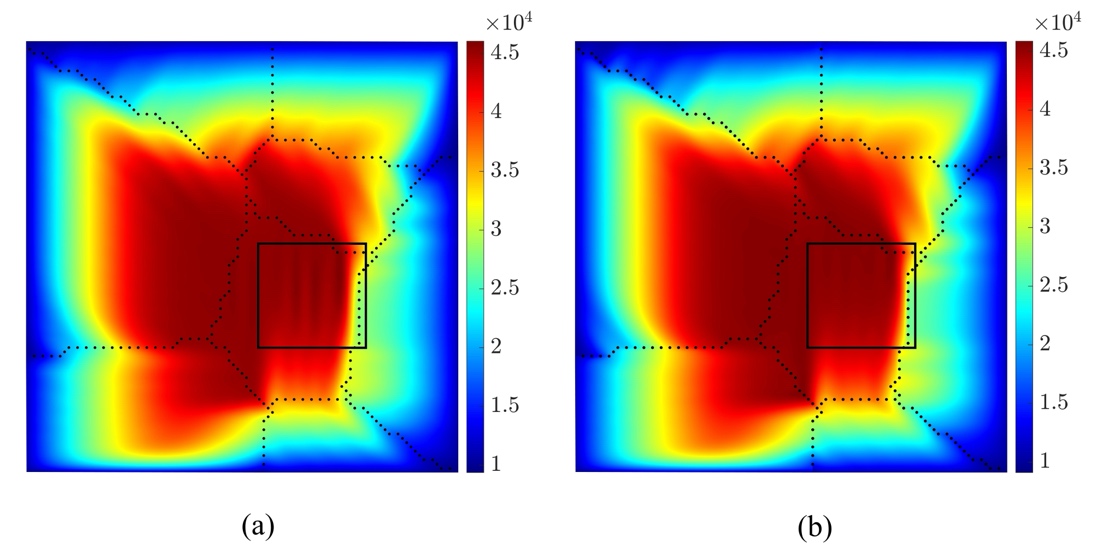
|  |  |  |  |
| --- | --- | --- | --- |
| Symbol | Variable Description | Value | Units |
|  | Reaction rate parameters at the anode |  |  |
|  | Reaction rate parameter at the cathode |  |  |
|  | Solid-phase electrical conductivity |  |  |
|  | Maximum theoretical lithium concentration |  |  |
|  | Initial stress-free lithium concentration |  |  |
|  | Faraday’s constant |  |  |
|  | Universal gas constant |  |  |
|  | Temperature |  |  |

A picture containing chart

Description automatically generated

**Fig. 22** Principal material orientation assignments denoted by arrow direction for each uniquely colored grain

After 500 seconds, the concentration field shows significant diffusion along the various principal grain orientations. Similar to stress fields in the previous study (see **Fig. 20**) the concentration field in **Fig. 23** computed with RKPM shows oscillations near an interface between grains with distinct principal directions. These oscillations are largely mitigated when using the IM-RKPM method.



**Fig. 23** The concentration field in computed after 500 seconds using (a) RKPM and (b) IM-RKPM. Grain boundary nodes are overlaid with black points

Graphical user interface, chart, surface chart

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**Fig. 24** Strain fields computed after 500 seconds using RKPM (top) and IM-RKPM (bottom). Cross sections and are labeled in red for corresponding strain cross-sectional plots in **Fig. 25**. Grain boundary nodes are overlaid with black points

**Chart, line chart

Description automatically generated**

**Fig. 25** Strain cross-sectional plots for comparison of RKPM and IM-RKPM solutions (a) at and (b) at

Diagram

Description automatically generated

**Fig. 26** Stress fields in computed after 500 seconds using RKPM (top) and IM-RKPM (bottom). Grain boundary nodes are overlaid with black points

At the final stage, strains are plotted in **Fig. 24** with cross-sectional strain distributions along cross sections and (labeled in **Fig. 24**) further visualized in **Fig. 25**, showing a reduced strain oscillation and sharper strain jumps across the grain boundaries in the IM-RKPM solutions. Final stress contours are shown in **Fig. 26**. For both strains and stresses, similar oscillation reduction and sharper jumps across grain boundaries are observed in IM-RKPM when compared with standard RKPM. Both full field plots are annotated with black boxes and gray circles to indicate corresponding areas of reduced oscillations. For example, when standard RKPM is used for the simulation, the resulting stress fields yield non-physical jumps in across vertical interfaces and in across horizontal interfaces, circled in gray in **Fig. 26**. This behavior is attributed to the Gibbs oscillations observed near these boundaries. When IM-RKPM is used, these non-physical stress discontinuities are largely suppressed.

# Conclusions

The interface-modified reproducing kernel particle method (IM-RKPM) introduced in this work allows for a direct transition from image pixels to node-based discretization without geometry reconstruction and mesh generation in the conventional modeling procedures. The RK kernel functions are scaled by a regularized distance function from evaluation points to the nearest grain boundaries. When kernel scaling is used in combination with strategic placement of interface node locations, the resulting IM-RKPM can account for both weak and strong discontinuities, making it effective to tackle discontinuities in the grain boundary mechanics. These discontinuous RK approximations significantly reduce Gibbs oscillations near grain boundaries. Under the IM-RKPM framework, adaptive refinement near grain boundaries and triple junctions is straightforward while maintaining desired consistencies and discontinuities. Further, no enrichment functions and the associated degrees of freedom are needed in the IM-RK approximation, making the IM-RKPM formulation easy to implement under the standard RKPM framework.

The coupled electro-chemo-mechanical system is modeled with IM-RKPM, and its convergence properties are investigated. A linear patch test is formulated for the coupled problem, incorporating the Butler-Volmer boundary condition which represents the critical physics in the Li-ion battery operations. With RKPM passing the linear patch test of the coupled problem, optimal rates of convergence are achieved for all fields in the electro-chemo-mechanical coupled problem with higher-order manufactured solutions.

In addition to the weak discontinuities across the grain boundaries, Li-ion battery microstructures exhibit stress concentrations near the triple junctions, and they can be properly captured via adaptive refinement in IM-RKPM in the vicinities of these junctions. The strain and stress oscillations near triple junctions in the RKPM solution are largely reduced in the IM-RKPM solution. IM-RKPM results also compare well with the solution obtained from a highly refined grain-boundary-fitted FEM model, demonstrating the effectiveness of the proposed IM-RKPM for image-based modeling of Li-ion battery microstructures.

As this work focuses on the grain boundary solution enhancement and the overall convergence of the coupled problem with IM-RKPM, Gauss integration is used to examine the proper convergence properties of the method without the influence of nodal integration and stabilizations of the coupled system. Nodal integration methods, like the stabilized conforming nodal integration (SCNI) [48] and the associated naturally stabilized nodal integration (NSNI) [49], can be employed for further computational efficiency.

# Appendix A: Nonlinear material property profiles

In the electro-chemo-mechanical system of Li-ion battery materials, as the normalized concentration varies, many of the material properties also change accordingly. The equations used to generate the nonlinear material property profiles shown in **Fig. 1** and **Fig. 2** following [20] are given here. The nonlinearities of the exchange current density and lattice parameters and with respect to the normalized concentration are characterized by the 10th order polynomials with the coefficients listed in Table 3.

The nonlinear diffusivity and equilibrium potential profiles are represented using piecewise polynomials. The piecewise polynomial coefficients are given for diffusivity in Table 4 and for equilibrium potential in Table 5. Each row of piecewise polynomial coefficients pertains to a given interval of .

Table 3. Characterized polynomial coefficients with respect to for lattice parameters and and exchange current density [43, 44]

|  |  |  |  |
| --- | --- | --- | --- |
| Polynomial | coefficients | coefficients | coefficients |
|  |  |  |  |
|  |  |  |  |
|  |  |  |  |
|  |  |  |  |
|  |  |  |  |
|  |  |  |  |
|  |  |  |  |
|  |  |  |  |
|  |  |  |  |
|  |  |  |  |
|  |  |  |  |

Table 4. Piecewise polynomial coefficients with respect to for diffusivity in [42]

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Interval | coefficients | coefficients | coefficients | coefficients |
|  |  |  |  |  |
|  |  |  |  |  |
|  |  |  |  |  |
|  |  |  |  |  |
|  |  |  |  |  |
|  |  |  |  |  |

Table 5. Piecewise polynomial coefficients with respect to for equilibrium potential in [43]

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Interval | coefficients | coefficients | coefficients | coefficients |
|  | 15.5276001364 | -6.5923311959 | -2.4960428818 | 5.6141783138 |
|  | 895.3135766304 | -33.6334506526 | -3.3486811169 | 5.3141783138 |
|  | -81.9793520542 | 10.4802301423 | -3.3516406933 | 5.2234296539 |
|  | 43.1929468100 | -5.9671112557 | -3.1630077379 | 4.9964228573 |
|  | 19.7924511525 | 0.2951859850 | -3.0606415847 | 4.6636439177 |
|  | -14.4065560642 | 4.5837966472 | -2.4078308531 | 4.3803240703 |
|  | 2.0308297558 | 1.2433285041 | -1.9232682056 | 4.1709723954 |
|  | 23.2107688484 | -0.9515644065 | -1.6136776121 | 3.9931096896 |
|  | 12.3318762496 | 0.9815583839 | -1.1076674280 | 3.8454370532 |
|  | -21.4709897243 | 3.6635330598 | -0.5413994637 | 3.7568177705 |
|  | -1.7180081860 | -0.7023601791 | -0.4528225435 | 3.7178421650 |
|  | -151.6070578413 | 5.2166938899 | -0.6448348249 | 3.6638183006 |
|  | -234.9780625093 | -49.0200087926 | -1.2602183697 | 3.6256674119 |
|  | -4304.650123480 | -199.562648108 | -4.15180267650 | 3.5598529149 |
|  | -10492.91639164 | -1948.39800276 | -13.0443209531 | 3.4381460848 |
|  | 154108.9641688 | -6491.33193384 | -33.3152697102 | 3.3229029154 |
|  | 85857831.55525 | -111367.986619 | -77.8486949008 | 3.0956434993 |
|  | -301.1173472459 | 260.223469449 | -43.0111734725 | 2.9922846494 |

# Appendix B: Plate with circular inclusion subjected to an eigenstrain

The infinite plate with a circular inclusion subjected to an eigenstrain [50] is investigated for comparison of the standard RKPM and IM-RKPM. In the numerical analysis, a plate with finite dimension is modeled, as shown in **Fig. 26**, where the circular inclusion is subjected to a uniform dilational eigenstrain of . Due to the symmetry of the geometry and loading, only a quarter model is used. Symmetric boundary conditions are prescribed for on and on , and the analytical displacement fields are prescribed for all other boundaries of the finite domain with Nitsche’s method [45] using a normalized penalty parameter of , where is the maximum Young’s modulus and is the maximum kernel support size associated with the essential boundary. A plane strain assumption is made. The dimension of the quarter domain is , and the inclusion radius is .

Chart

Description automatically generated with low confidence

**Fig. 27** Problem set up for a plate with a circular inclusion (domain 1) in a matrix (domain 2)

The analytical solution [50] gives the following displacements:

|  |  |  |
| --- | --- | --- |
|  |  | (92) |
|  |  | (93) |

and strains:

|  |  |  |
| --- | --- | --- |
|  |  | (94) |
|  |  | (95) |
|  |  | (96) |

where the constant is defined as:

|  |  |  |
| --- | --- | --- |
|  |  | (97) |

The Lamé parameters, and , are related to the elastic modulus, , and Poisson ratio, , by

|  |  |  |
| --- | --- | --- |
|  |  | (98) |
|  |  | (99) |

The model discretization in **Fig. 27** includes 224 nodes, with 4x4 Gauss quadrature for domain integration and 4-point Gauss quadrature for boundary integration. Linear basis with cubic B-splines and normalized support sizes of are used for both RK and IM-RK shape functions.

Chart

Description automatically generated with low confidence

**Fig. 28** RKPM discretization of the quarter model

In the first analysis, material properties for the inclusion (domain 1) and the matrix (domain 2) are , , , and . The results are displayed in **Fig. 28**. As expected, the smooth nature of the standard RK approximation yields significant Gibbs oscillations in the radial and hoop strains near the interface. IM-RKPM properly captures the weak discontinuity in the radial displacement and reduces oscillations in the strain fields.

Graphical user interface, chart

Description automatically generated

**Fig. 29** Comparison of , , and using standard RK and IM-RK approximations, plotted along the radial line when , , , and

The numerical test was repeated with a larger stiffness ratio of , instead of the ratio of from the previous case. As seen in **Fig. 29**, the increased stiffness ratio leads to larger, more pronounced Gibbs oscillations for the standard RK approximation. The introduction of the IM-RK approximation is still able to greatly reduce oscillatory interface behavior in the strain fields.

Graphical user interface, chart, line chart

Description automatically generated

**Fig. 30** Comparison of , , and using standard RK and IM-RK approximations, plotted along the radial line y=x when , =0.28, =900, and =0.33

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**References**

1. Palacín MR, De Guibert A (2016) Why do batteries fail? Science 351:1253292. https://doi.org/10.1126/science.1253292

2. Chen Z, Danilov DL, Eichel R-A, Notten PHL (2022) Porous Electrode Modeling and its Applications to Li-Ion Batteries. Adv Energy Mater 12:2201506. https://doi.org/10.1002/aenm.202201506

3. Quinn A, Moutinho H, Usseglio-Viretta F, Verma A, Smith K, Keyser M, Finegan D (2020) Electron Backscatter Diffraction for Investigating Lithium-Ion Electrode Particle Architectures. Cell Rep Phys Sci 1:100137. https://doi.org/10.1016/j.xcrp.2020.100137

4. Furat O, Finegan D, Diercks D, Usseglio-Viretta F, Smith K, Schmidt V (2021) Mapping the architecture of single lithium ion electrode particles in 3D, using electron backscatter diffraction and machine learning segmentation. J Power Sources 483:229148. https://doi.org/10.1016/j.jpowsour.2020.229148

5. Nitta N, Wu F, Lee JT, Yushin G (2015) Li-ion battery materials: present and future. Mater Today 18:252–264. https://doi.org/10.1016/j.mattod.2014.10.040

6. Manthiram A (2020) A reflection on lithium-ion battery cathode chemistry. Nat Commun 11:1550. https://doi.org/10.1038/s41467-020-15355-0

7. Mohamed N, Allam NK (2020) Recent advances in the design of cathode materials for Li-ion batteries. RSC Adv 10:21662–21685. https://doi.org/10.1039/D0RA03314F

8. Zhang X, Shyy W, Marie Sastry A (2007) Numerical Simulation of Intercalation-Induced Stress in Li-Ion Battery Electrode Particles. J Electrochem Soc 154:A910. https://doi.org/10.1149/1.2759840

9. Doyle M, Fuller T, Newman J (1993) Modeling of Galvanistic Charge and Discharge of the Lithium/Polymer/Insertion Cell. J Electrochem Soc 140:. https://doi.org/10.1149/1.2221597

10. Plett GL (2015) Battery Management Systems, Volume I: Battery Modeling. Artech House

11. Richardson GW, Foster JM, Ranom R, Please CP, Ramos AM (2022) Charge transport modelling of Lithium-ion batteries. Eur J Appl Math 33:983–1031. https://doi.org/10.1017/S0956792521000292

12. Kim G-H, Smith K, Lee K-J, Santhanagopalan S, Pesaran A (2011) Multi-Domain Modeling of Lithium-Ion Batteries Encompassing Multi-Physics in Varied Length Scales. J Electrochem Soc 158:A955. https://doi.org/10.1149/1.3597614

13. Schmidt A, Oehler D, Weber A, Wetzel T, Ivers-Tiffée E (2021) A multi scale multi domain model for large format lithium-ion batteries. Electrochim Acta 393:139046. https://doi.org/10.1016/j.electacta.2021.139046

14. Jokar A, Rajabloo B, Désilets M, Lacroix M (2016) Review of simplified Pseudo-two-Dimensional models of lithium-ion batteries. J Power Sources 327:44–55. https://doi.org/10.1016/j.jpowsour.2016.07.036

15. Chada T, Suther B, Rife D, Subramanian V, Biswas P (2017) Model Based Analysis of One-Dimensional Oriented Lithium-Ion Battery Electrodes. J Electrochem Soc 164:E3114–E3121. https://doi.org/10.1149/2.0141711jes

16. Mai W, Colclasure A, Smith K (2019) A Reformulation of the Pseudo2D Battery Model Coupling Large Electrochemical-Mechanical Deformations at Particle and Electrode Levels. J Electrochem Soc 166:A1330–A1339. https://doi.org/10.1149/2.0101908jes

17. Zhao Y, Stein P, Bai Y, Al-Siraj M, Yang Y, Xu B-X (2019) A review on modeling of electro-chemo-mechanics in lithium-ion batteries. J Power Sources 413:259–283. https://doi.org/10.1016/j.jpowsour.2018.12.011

18. Marquis S, Sulzer V, Timms R, Please C, Chapman SJ (2019) An Asymptotic Derivation of a Single Particle Model with Electrolyte. J Electrochem Soc 166:A3693–A3706. https://doi.org/10.1149/2.0341915jes

19. Dao T-S, Vyasarayani C, McPhee J (2012) Simplification and order reduction of lithium-ion battery model based on porous-electrode theory | Elsevier Enhanced Reader. J Power Sources 198:329–337. https://doi.org/DOI: 10.1016/j.jpowsour.2011.09.034

20. Allen JM, Weddle PJ, Verma A, Mallarapu A, Usseglio-Viretta F, Finegan DP, Colclasure AM, Mai W, Schmidt V, Furat O, Diercks D, Tanim T, Smith K (2021) Quantifying the influence of charge rate and cathode-particle architectures on degradation of Li-ion cells through 3D continuum-level damage models. J Power Sources 512:230415. https://doi.org/10.1016/j.jpowsour.2021.230415

21. Chen W-X, Allen JM, Rezaei S, Furat O, Schmidt V, Singh A, Weddle PJ, Smith K, Xu B-X (2024) Cohesive phase-field chemo-mechanical simulations of inter- and trans- granular fractures in polycrystalline NMC cathodes via image-based 3D reconstruction. J Power Sources 596:234054. https://doi.org/10.1016/j.jpowsour.2024.234054

22. Nguyen T-T, Réthoré J, Yvonnet J, Baietto M-C (2017) Multi-phase-field modeling of anisotropic crack propagation for polycrystalline materials. Comput Mech 60:289–314. https://doi.org/10.1007/s00466-017-1409-0

23. Quey R, Dawson PR, Barbe F (2011) Large-scale 3D random polycrystals for the finite element method: Generation, meshing and remeshing. Comput Methods Appl Mech Eng 200:1729–1745. https://doi.org/10.1016/j.cma.2011.01.002

24. Lim H, Battaile CC, Bishop JE, Foulk JW (2019) Investigating mesh sensitivity and polycrystalline RVEs in crystal plasticity finite element simulations. Int J Plast 121:101–115. https://doi.org/10.1016/j.ijplas.2019.06.001

25. Liu WK, Jun S, Zhang YF (1995) Reproducing kernel particle methods. Int J Numer Methods Fluids. https://doi.org/10.1002/fld.1650200824

26. Chen J-S, Pan C, Wu C-T, Liu WK (1996) Reproducing Kernel Particle Methods for large deformation analysis of non-linear structures. Comput Methods Appl Mech Eng 139:195–227. https://doi.org/10.1016/S0045-7825(96)01083-3

27. Liu WK, Chen Y, Jun S, Chen JS, Belytschko T, Pan C, Uras RA, Chang CT (1996) Overview and applications of the reproducing Kernel Particle methods. Arch Comput Methods Eng 3:3–80. https://doi.org/10.1007/BF02736130

28. Chen J-S, Hillman M, Chi S-W (2017) Meshfree Methods: Progress Made after 20 Years. J Eng Mech 143:04017001. https://doi.org/10.1061/(ASCE)EM.1943-7889.0001176

29. Belytschko T, Chen JS, Hillman M (2023) Meshfree and Particle Methods: Fundamentals and Applications, 1st ed. Wiley

30. You Y, Chen J-S, Lu H (2003) Filters, reproducing kernel, and adaptive meshfree method. Comput Mech 31:316–326. https://doi.org/10.1007/s00466-003-0434-3

31. Yang JP, Chi S-W, Chen J-S (2013) Image-Based Multiscale Modeling of Porous Bone Materials. In: Li S, Qian D (eds) Multiscale Simulations and Mechanics of Biological Materials. Wiley, Oxford, UK, pp 377–401

32. Basava RR, Chen J-S, Zhang Y, Sinha S, Sinha U, Hodgson J, Csapo R, Malis V (2014) Pixel Based Meshfree Modeling of Skeletal Muscles. In: Zhang YJ, Tavares JMRS (eds) Computational Modeling of Objects Presented in Images. Fundamentals, Methods, and Applications. Springer, Cham, pp 316–327

33. Zhang H, Gao Z, Xu L, Yu X, Wong KCL, Liu H, Zhuang L, Shi P (2018) A Meshfree Representation for Cardiac Medical Image Computing. IEEE J Transl Eng Health Med 6:1800212. https://doi.org/10.1109/JTEHM.2018.2795022

34. Wang Y, Baek J, Tang Y, Du J, Hillman M, Chen JS (2023) Support vector machine guided reproducing kernel particle method for image-based modeling of microstructures. Comput Mech. https://doi.org/10.1007/s00466-023-02394-9

35. Krongauz Y, Belytschko T (1998) EFG approximation with discontinuous derivatives. Int J Numer Methods Eng 41:1215–1233. https://doi.org/10.1002/(SICI)1097-0207(19980415)41:7<1215::AID-NME330>3.0.CO;2-#

36. Wang D, Chen J-S, Sun L (2003) Homogenization of magnetostrictive particle-filled elastomers using an interface-enriched reproducing kernel particle method. Finite Elements in Analysis and Design 39:765–782. https://doi.org/10.1016/S0168-874X(03)00058-1

37. Chen JS, Kotta V, Lu H, Wang D, Moldovan D, Wolf D (2004) A variational formulation and a double-grid method for meso-scale modeling of stressed grain growth in polycrystalline materials. Comput Methods Appl Mech Eng 193:1277–1303. https://doi.org/10.1016/j.cma.2003.12.020

38. Wang D, Sun Y, Li L (2009) A Discontinuous Galerkin Meshfree Modeling of Material Interface. Comput Model Eng Sci 45:57–82. https://doi.org/10.3970/cmes.2009.045.057

39. Roth MJ, Chen J, Danielson KT, Slawson TR (2016) Hydrodynamic meshfree method for high‐rate solid dynamics using a Rankine–Hugoniot enhancement in a Riemann‐SCNI framework. Int J Numer Methods Eng 108:1525–1549. https://doi.org/10.1002/nme.5266

40. Sun CT, Guan PC, Jiang JH, Kwok OLA (2018) The weighted reconstruction of reproducing kernel particle method for one-dimensional shock wave problems. Ocean Eng 149:325–340. https://doi.org/10.1016/j.oceaneng.2017.12.017

41. Díaz JI, Gómez-Castro D, Ramos AM (2019) On the well-posedness of a multiscale mathematical model for Lithium-ion batteries. Advances in Nonlinear Analysis 8:1132–1157. https://doi.org/10.1515/anona-2018-0041

42. Verma A, Smith K, Santhanagopalan S, Abraham D, Yao KP, Mukherjee PP (2017) Galvanostatic Intermittent Titration and Performance Based Analysis of LiNi 0.5 Co 0.2 Mn 0.3 O 2 Cathode. J Electrochem Soc 164:A3380–A3392. https://doi.org/10.1149/2.1701713jes

43. Colclasure AM, Dunlop AR, Trask SE, Polzin BJ, Jansen AN, Smith K (2019) Requirements for Enabling Extreme Fast Charging of High Energy Density Li-Ion Cells while Avoiding Lithium Plating. J Electrochem Soc 166:A1412–A1424. https://doi.org/10.1149/2.0451908jes

44. Dolotko O, Senyshyn A, Mühlbauer MJ, Nikolowski K, Ehrenberg H (2014) Understanding structural changes in NMC Li-ion cells by in situ neutron diffraction. J Power Sources 255:197–203. https://doi.org/10.1016/j.jpowsour.2014.01.010

45. Nitsche J (1971) Über ein Variationsprinzip zur Lösung von Dirichlet-Problemen bei Verwendung von Teilräumen, die keinen Randbedingungen unterworfen sind. AbhMathSeminUnivHambg 36:9–15. https://doi.org/10.1007/BF02995904

46. Yreux E, Chen J (2017) A quasi‐linear reproducing kernel particle method. Int J Numer Methods Eng 109:1045–1064. https://doi.org/10.1002/nme.5319

47. Furat O, Petrich L, Finegan DP, Diercks D, Usseglio-Viretta F, Smith K, Schmidt V (2021) Artificial generation of representative single Li-ion electrode particle architectures from microscopy data. npj Comput Mater 7:105. https://doi.org/10.1038/s41524-021-00567-9

48. Chen J-S, Wu C-T, Yoon S, You Y (2001) A stabilized conforming nodal integration for Galerkin mesh-free methods. Int J Numer Methods Eng 50:435–466. https://doi.org/10.1002/1097-0207(20010120)50:2<435::AID-NME32>3.0.CO;2-A

49. Hillman M, Chen J-S (2016) An accelerated, convergent, and stable nodal integration in Galerkin meshfree methods for linear and nonlinear mechanics. Int J Numer Methods Eng 107:603–630. https://doi.org/10.1002/nme.5183

50. Mura T (1987) Micromechanics of Defects in Solids, 2nd ed. Springer Dordrecht