

# ONTOLOGY ASSISTED MODELLING OF GALVANIC CORROSION OF MAGNESIUM

DANIEL HOECHE<sup>1</sup>, NATALIA KONCHAKOVA<sup>1</sup>, MIKHAIL L. ZHELUDKEVICH<sup>1;2</sup>,  
THOMAS F. HAGELIEN<sup>3</sup> AND JESPER FRIIS<sup>4</sup>

<sup>1</sup> Institute of Surface Science, Helmholtz-Zentrum Geesthacht, Max-Planck Str. 1, Geesthacht, 21502, Germany, daniel.hoeche@hzg.de, natalia.konchakova@hzg.de, mikhail.zheludkevich@hzg.de

<sup>2</sup> Faculty of Engineering, Kiel University, Kaiserstrasse 2, 24143 Kiel, Germany

<sup>3</sup> SINTEF-Ocean, NO-7465 Trondheim, Norway, thomas.f.hagelien@sintef.no,

<sup>4</sup> SINTEF-Industry, NO-7465 Trondheim, Norway jesper.friis@sintef.no

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**Abstract.** Multi-physics and multiscale modelling frameworks, especially for the computer based analysis of surface damage and corrosion, requires not only robust computational tools but also an efficient data-centric architecture for handling information exchange at different modelling scales. The issue to exchange data provided by different computational solvers as well as required and used in different programming languages forms a request in specific formats signifying a strong non-uniformity for an easy nexus with other solvers. This non-uniformity has created a need to focus on intermittent state-of-the-art data-centric software tools, which aim to bridge data exchange technology, to ensure heterogeneity across a wide range of solvers [1]. Moreover, data organization in the form of ontological representation and metadata structures are necessary to be prepared as a standard for a coherent representation of information regardless of the diverse nature of data formats specific to a scientific discipline. A domain ontology based on the European Materials & Modelling Ontology (EMMO) for galvanic corrosion is outlined and connected to corresponding concepts in a galvanic corrosion model. This fundamental work provides and discusses the concept, underlying terminology and working mechanism of a data-centric architecture for exchanging and interfacing data-flow between data sources/sinks and solvers. It realizes an ontological representation of physics and chemistry of galvanic corrosion. Thus, it is a nuclei for further improvement's of interoperability between complex corrosion related phenomena and models. It paves the way for accurate computational corrosion engineering.

## 1 INTRODUCTION

Damage and corrosion are huge issues for engineering of multi-material structures and for the construction sector, which have a high economic impact [2]. An accurate service-life prediction and maintenance action are challenging scientific and industrial problems on which researchers and manufacturing specialists are working together. Successful predictions of corrosion and extension of service-life of en-

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gineering structures and lightweight multi materials systems can bring till 8% of world  $CO_2$  emission and contribute sufficiently to the EU Green deal course. Modelling and simulation provide fundamental support in this process. Currently some modelling approaches are developed to analyse corrosion of materials surface, damage state and failure due to the materials degradation. Advanced simulation techniques provide the access to different software and data, which should be available and exchangeable in numerical analysis of the complex non-linear multi-scale modelling problem of corrosion, bridging different simulation tools on different simulation steps and levels [4]. Moreover, the computation challenge includes different issues with homogenization, accuracy, real-time validation and other functional complexities [1]. Unfortunately, exchange of data provided by different computational solvers is very often strong non-uniform due to use of different programming languages and application of different modelling stiles. To solve this software engineering and scientific issue and support an easy nexus between modeling codes and data with different solvers, the standardisation of modelling workflows, in form of materials modelling data table (MODA) description, data organization and providing ontological representation of models and used or generated data are required [3]. Moreover, the ontological based modelling and simulation would provide support CAD, predictive maintenance and contribute to the detail analysis of risk management in multi-functional constructions.

In this paper, the authors would like to demonstrate an ontology based modelling approach applied to a simple example of predictive modelling of galvanic couple of magnesium based components in light weight structural design. The degradation of galvanic coupled materials is a well known problem, especially for constructors. Due to the dissolution of the magnesium alloys in aggressive environments in contact with the more noble elements, structural and quality issues occur. For that purpose it would be nice to have a tool to compute the degradation depending on the environmental constraints.

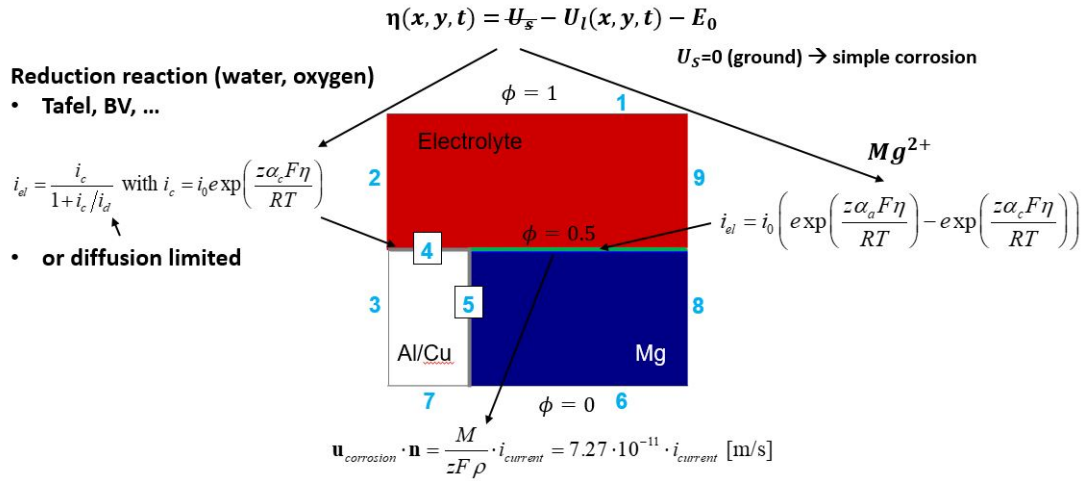
In this study, we simulate the degradation process between pure Mg and a 6060 aluminium alloy or bare copper on a very simple geometry by application of the Level-Set method mathematically coupled to the electrochemical fundamental equations. The investigation includes the ontology of the simple galvanic corrosion model. Both contact partners were chosen to show the influence of varying overpotentials. In the literature nice investigations by Desphande [5, 6, 7], Murer [8], Jia [9, 10] or Lacroix [11] can be found using similar approaches. The real world objects and their properties are related to concepts in a domain ontology for galvanic corrosion based on the European Materials & Modelling Ontology (EMMO) [12]. This study will give a base for further investigations and offer some possibilities to implement the involved chemical and physical processes on a “volumetric” surface [1].

## 2 Ontology for corrosion modelling

### 2.1 Problem description domain: computation of galvanic corrosion

For the simulation of current distributions, the Laplace equation is solved and coupled to the Level-Set equations according [13]. The electrode kinetics was described by the Buter-Volmer equation and by a modified version according Murer et.al [14]. Figure 1 shows the considered geometry and materials domains.

The fundamental approach is to set the Level-Set variable  $\phi$  at the magnesium surface to 0.5. Boundary numbers and Level-Set initials are indicated in the Figure 1.  $\eta = U_s - U_l - E_0$  is the over potential, which depends on the electrode (extern) potential  $U_s$  (here  $U_s = 0$ ), the electrolyte potential  $U_l$  and the equilibrium potential  $E_0$ .  $i$  is the current. The boundary conditions (BCs) are presented in Table 1.



**Figure 1:** Materials domain, geometry, boundaries and applied materials relations used for simulation. The blue numbers label the different boundaries in the model.

**Table 1:** Boundary conditions at the considered geometry.

Boundary	Laplace	Level-Set
1	$i = 0$	$\phi = 1$
2	symmetry	symmetry
3	symmetry	–
4	$\eta$	symmetry
5	$\eta(\phi > 0.5)$	
6	$U_s = 0$	$\phi = 0$
7	$U_s = 0$	–
8	$i = 0$	symmetry
9	$i = 0$	symmetry

### 2.1.1 Electrochemistry

The model is based on very simple assumptions by neglecting convection, diffusion and concentration gradients. Thus, the Nernst equation which describes the electrolyte is reduced to the Laplace equation  $\Delta U = 0$  in the simplified model and the following equations occur:

$$\nabla \mathbf{j}_{ls} = Q_{ls} \text{ and } \mathbf{j}_{ls} = -\sigma \cdot \nabla U_{l,s} \quad (1)$$

Here  $l$  and  $s$  indicate the electrolyte respectively the electrode,  $\mathbf{j}$  describes the current density vector,  $U$  the electrical potentials and  $\sigma$  the effective conductivity.  $Q$  as a source term becomes zero. At the electrode-electrolyte interface, the standard Butler-Volmer equation in Mg and the modified Butler-Volmer equation according Murer et.al [14] in Al and Cu, were used to explain and describe the kinetics (empirical). The required parameters were taken from the experimental studies and fittings.

$$i_{el} = i_0 \left[ \exp\left(\frac{z\alpha_a F \eta}{RT}\right) - \exp\left(\frac{z\alpha_c F \eta}{RT}\right) \right] \quad (2)$$

$z$  is the electron number,  $F$  is the Faraday constant,  $T$  is room temperature,  $R$  is the molar gas constant,  $\eta$  is the overpotential,  $i_0$  is the exchange current, and  $\alpha_a, \alpha_c$  are the anodic and cathodic coefficient, respectively. The modified equation just describes the cathodic branch and looks like:

$$i_{el} = \frac{i_c}{1 + i_c/i_d} \quad \text{with} \quad i_c = i_0 \exp\left(\frac{z\alpha_c F \eta}{RT}\right) \quad (3)$$

The difference results from  $i_d$  which describes the current limitation due to diffusion (reduction) processes mainly based on oxygen. This effect was discussed by Hoeche and Isakovic in [20]. The fitting parameters, used for the simulation and ontology, are presented in Table 2.

**Table 2:** Parameters for electro-chemical analysis.

Material	$E_0$ [V]	$i_0, i_d$ [mA/cm <sup>2</sup> ]
cp-Mg	-1.65	0.035/-
Al-alloy	-0.62	-0.0009/- 0.013
Cu	-0.22	-0.0073/- 0.090

### 2.1.2 Free surface

In order to model the surface tracking, the Level-Set method was applied. A function  $\phi(x, y, t)$  was defined over the entire domain to describe the interface, according to the following criteria:

$$\phi(x, y, t) = \begin{cases} > 0.5 & \text{electrolyte} \\ = 0.5 & \text{interface} \\ < 0.5 & \text{magnesium} \end{cases} \quad (4)$$

The interface movement can be described by a simple partial differential equation, which can be solved simultaneously numerically with the Laplace equation. Olsson et.al. [15] used the following expression:

$$\frac{\partial \phi}{\partial t} + \mathbf{u}_{corr} \cdot \nabla \phi = \gamma_L \nabla \cdot \left( \varepsilon ((\nabla \phi \cdot \mathbf{n}) \mathbf{n}) - \phi(1 - \phi) \frac{\nabla \phi}{|\nabla \phi|} \right) \quad (5)$$

Here the vector  $\mathbf{u}_{corr}$  is the speed of the surface tracking, which is considered in this case as the corrosion rate. The speed is a result of the Laplace equation and the normal vectors of the interface  $\mathbf{n}$ . The interface thickness  $\varepsilon$  was set to  $\varepsilon = 3h$  (where  $h$  is the mesh size).  $\gamma_L$  is a stabilization parameter that determines the repetition of re-initialization for each time step. This parameter has the unit [m/s] and is in the order of the maximum velocity of approximately  $10^{-7} - 10^{-9}$  appearing from the corrosion rate. For accurate computation tuning becomes necessary. The interface normal vector is defined by  $\mathbf{n} = \frac{\nabla \phi}{|\nabla \phi|}$  at  $\phi = 0.5$  and the surface velocity for Mg is according to Faraday's law:

$$\mathbf{u}_{corr} \cdot \mathbf{n} = \frac{M}{zF\rho} \cdot i_{current} = 7.27 \cdot 10^{-11} \cdot i_{current} \text{ [m/s]} \quad (6)$$

where  $M$  is the atomic mass,  $z$  is the electron number,  $F$  is the Faraday constant and  $\rho$  is the density of the corroding Mg.  $F = 96,485.34 \text{ C mol}^{-1}$ ,  $M = 23.98 \text{ g mol}^{-1}$ ,  $z = 2$ ,  $\rho = 1770 \text{ kg m}^{-3}$  for the corroding  $\alpha$ -phase. A smoothed continuously differentiable function was defined to accommodate the boundary conditions at the interface:

$$\delta = 6 \nabla \phi \cdot |\phi(1 - \phi)| \quad (7)$$

Thereby, it is possible to transform boundary phenomena to volumetric ones. Due to the smeared out interface, the material parameters along the interface were described by Eq. (8), whereas  $f$  corresponds to the different physical properties (e.g.  $\sigma$ ) of the electrolyte and the magnesium.

$$f = f_{Mg}(\phi < 0.5) + f_{el}(\phi > 0.5) \quad (8)$$

The initial level set function  $\phi_0$  was set according to the model build up in Figure 1.

### 2.1.3 Experimental data

Experimental input data needed by the modelling, were produced from 99.5% pure magnesium, aluminium 6060 and 99.5% grade copper. The galvanic couple was adjusted by blocks of 10x12x20 cm which were joined by plastic screws. After grinding and cleaning, the samples were placed in a foursquare acrylic glass container with an agitator filled with 0.1% and 3.5% sodium chloride, respectively. The assay was pressed from inside to the wall. On the other side of the glass, a camera was installed which documented the experiments. After a certain time of immersion, samples were dried and cleaned by chromic acid to remove corrosion products.

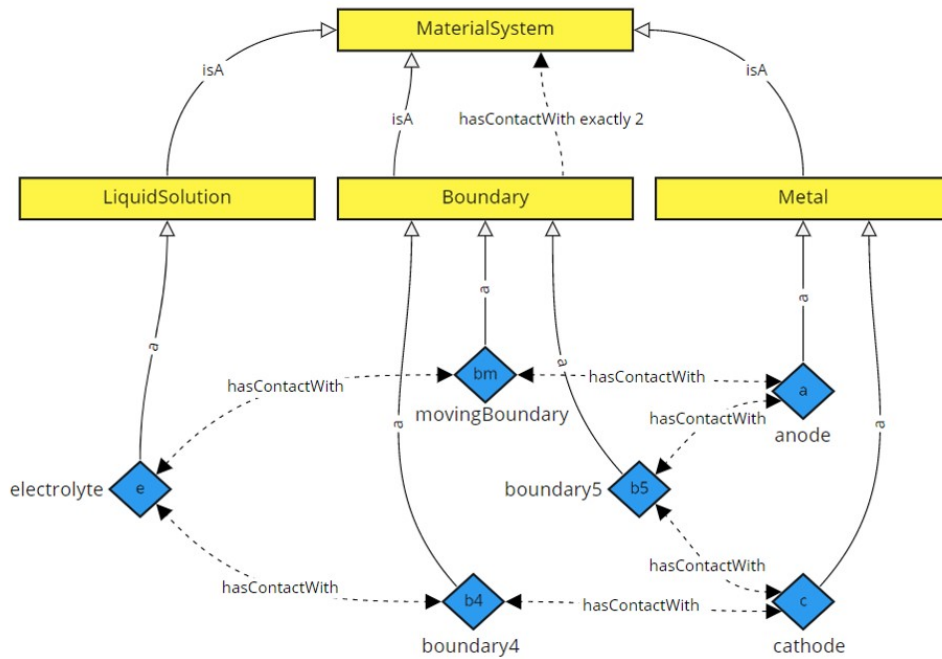
Potentiodynamic polarisation was performed in 0.1% and 3.5% NaCl solution, respectively, at a starting pH of 6.5. The corrosion cell (333 ml) with a three electrode set-up consisted of an Ag/AgCl reference, a Pt counter electrode and the specimen as working electrode. The electrolyte temperature was  $22 \pm 0.4^\circ\text{C}$  and the electrolyte was stirred during the experiments.

## 2.2 Ontologising galvanic corrosion

In order to enable a semantic description of the galvanic corrosion model, we will in this section outline a domain ontology for galvanic corrosion that the real world objects and physical quantities in our corrosion model can be related to.

Figure 2 shows the ontologisation of the three domains and the three internal boundaries in Figure 1 as individuals. Their corresponding material subclasses are also shown. The boundaries are related to the domains via *hasContactWith* relations (illustrated as double-headed arrows to emphasise that it is symmetric). For simplicity are the external boundaries not included in the figure.

Figure 3 shows how the anode, cathode and electrolyte individuals are related to their corresponding formulation of the Laplace equation for the three domains ( $da$ ,  $dc$ ,  $de$ ) with *hasModel* relations. These



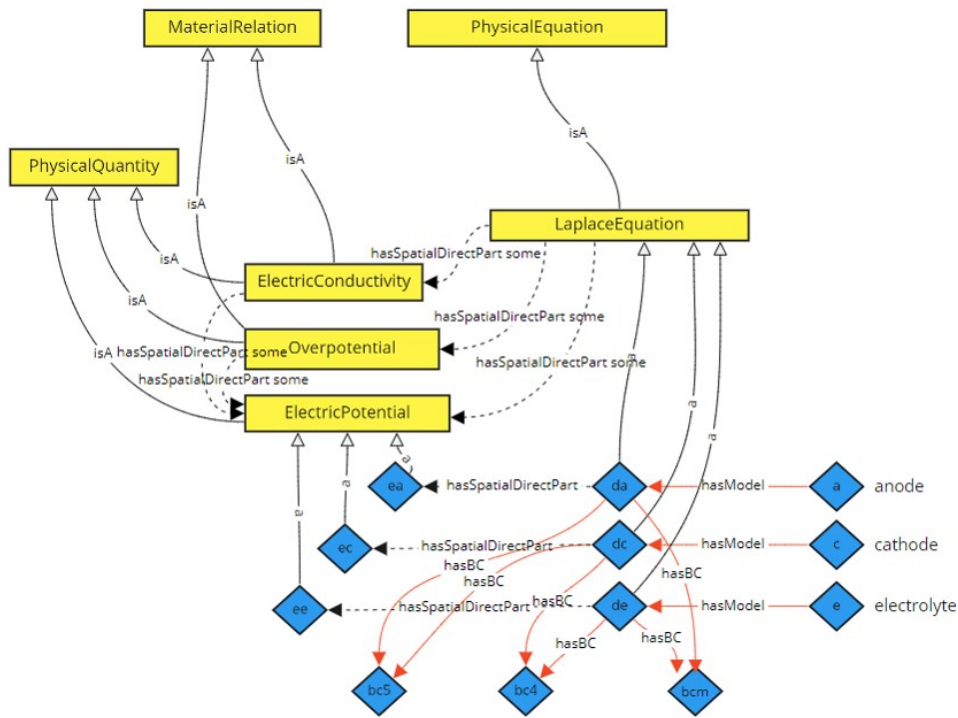
**Figure 2:** Real world material classes (yellow boxes) and individuals (blue diamonds) considered galvanic corrosion model. The individuals refer to the domains (anode: Mg, cathode: Cu and electrolyte) and some selected boundaries (bm: Mg/electrolyte, b4: Cu/electrolyte and b5: Mg/Cu) in Figure 1.

instances of the Laplace equation are related to their boundary conditions via *hasBC* relations and to their corresponding solutions (ea, ec, ee), which are instances of ElectricPotential, via *hasSpatialDirectPart* relations. Spatial direct parthood is used, because EMMO allows one to decompose a physical equation into its spatial direct parts, being operators, numbers and physical quantities.

A simplified view of the ontological representation of the boundary conditions is illustrated in Figure 4. *hasModel* relations are used to relate the three inner boundaries (b5, b4, bm) to their corresponding boundary conditions (bc5, bc4, bcm). These boundary conditions are material relations and have overpotential individuals (n5, n4 and nm) as one of their spatial direct parts. The boundary condition *bc4* is further classified as a modified interface current equation and the boundary condition *bcm* is sub-classified as a standard interface current equation, both of these being material relations.

### 2.3 Efficient data centric architecture

Input data and simulation state are represented by a data model which is mapped to EMMO. This ensures variables are consistent with the physical equations, materials relations and boundary conditions. The data-models are represented as entities with a set of properties. Each properties can have a data representation shape, type (string, float, int, boolean etc) and a unit. In addition, the attribute can be mapped to specific observation methods in the EMMO that allows for documenting how the specific data point was obtained through direct- or indirect measurement or calculation. For example, the electric potential can be described as a 2D scalar field with the shape corresponding to the grid, with the unit Volt. The semantic interoperability platform SOFT [1] allows for instantiating the data model and manages data



**Figure 3:** Ontological representation of the Laplace equation.

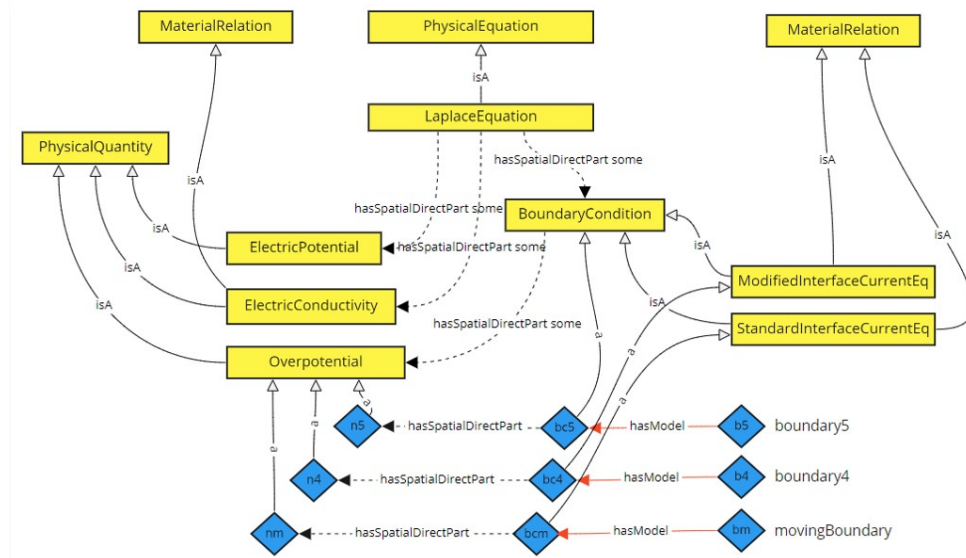
input/output to and from various sources, while ensuring a consistent internal representation.

## 2.4 Simulation and Experimental Results.

Potentiodynamic polarization measurements of the used samples reveal the electro-kinetic parameters for the modeling. The corresponding graph, included the polarization curves, the determined parameters of the uncoupled bare metal samples measurements and fitted theoretical curves were also presented in [20]. In case of copper and aluminum they match the investigations of Clark et.al [16].

The fits according Murer [14] describe the kinetics over a wide range. Tafel approximation doesn't fulfill this. In this case the current  $i_d$  limits the cathodic kinetics mainly due to (e.g.  $4e^-$  on Cu) oxygen reduction. Observations and the fitted values are in agreement with previous studies on Cu [17], [18] or on Al [19]. For very high polarization the so called hump phenomena occurs, which has not considered in the current study. Since the electrical conductivity is a very important parameter, it has been measured before and after the galvanic tests. Table 3 shows no significant variation leading to assume a constant (55/1.9 [mS/cm]) in the simulation.

The modelling results are demonstrated in Figures 5 - 6. The computation itself needs the direct coupling of the mentioned equations. Time steps, reinitialization, the mesh size and the interface width have to be set up very careful. For an optimized computation the following results occur. As shown in Figure 5 the electrolyte potential distribution varies mainly due to the low conductivity. Electrolyte potential (color), current flow lines (arrows) and surface deformation (red line corresponds to  $\phi = 0.5$ ) on the systems



**Figure 4:** Ontological representation of the boundary conditions.

**Table 3:** Conductivities of the solutions before (first corresponding number) and after (second corresponding number) galvanic couple experiments

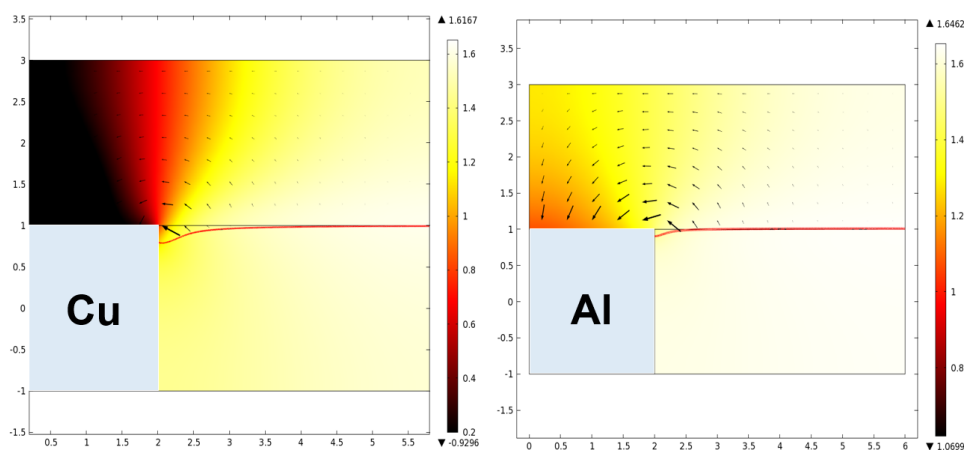
	$\sigma$ conductivity at 3.5% NaCl [mS/cm]		$\sigma$ conductivity at 0.1% NaCl [mS/cm]	
Mg-Al	51.7	52.6	1.83	2.01
Mg-Cu	55.6	56.4	1.86	2.00

Mg-Cu (left) and Mg-Al6060 (right) in 0.1% sodium chloride after 19 hours immersion are presented in the figure. The current flow to the cathode indicates the Mg dissolution.

For the higher concentrated electrolyte, massive hydrogen evolution has been observed. The magnesium dissolves very fast according to the well known reactions. The high overpotential and the arising current density of  $\sim 60 \text{ A/cm}^2$  for Al and of  $\sim 270 \text{ A/cm}^2$  for Cu lead to a rapid attack (see figure 6). On the figure the electrolyte potential (color), current flow lines (arrows) and surface deformation (red line -  $\phi = 0.5$ ) on both systems in 3.5% sodium chloride after 19 hours immersion being compared to experimental results (etched by chromic acid) are presented.

Contrary to low electrolyte conductivities the potential reached an equilibrium value as predicted by the Mixed potential theory. For copper a value of  $-1.41 \text{ V}$  and for aluminum of  $-1.63 \text{ V}$  has been determined. The comparison with the polarization curves presented in [20] provides an adequate agreement. The surface tracking by applying Faraday's law works fine. Hence, there are still certain variations. They arise from chemical conversion during immersion and massive changing conditions. The hydrogen evolution and the  $\text{Mg}(\text{OH})_2$  formation strongly influence the process. Modeling of electrode surface kinetics still needs some improvements. Especially the Butler Volmer approach is just a very rough approximation, failing for high overpotentials. The equations of Murer show this possibilities, but also the limitations.





**Figure 5:** Modelling results for the systems Mg-Cu (left) and Mg-Al6060 (right) in **0.1%** sodium chloride after 19 hours immersion [20].

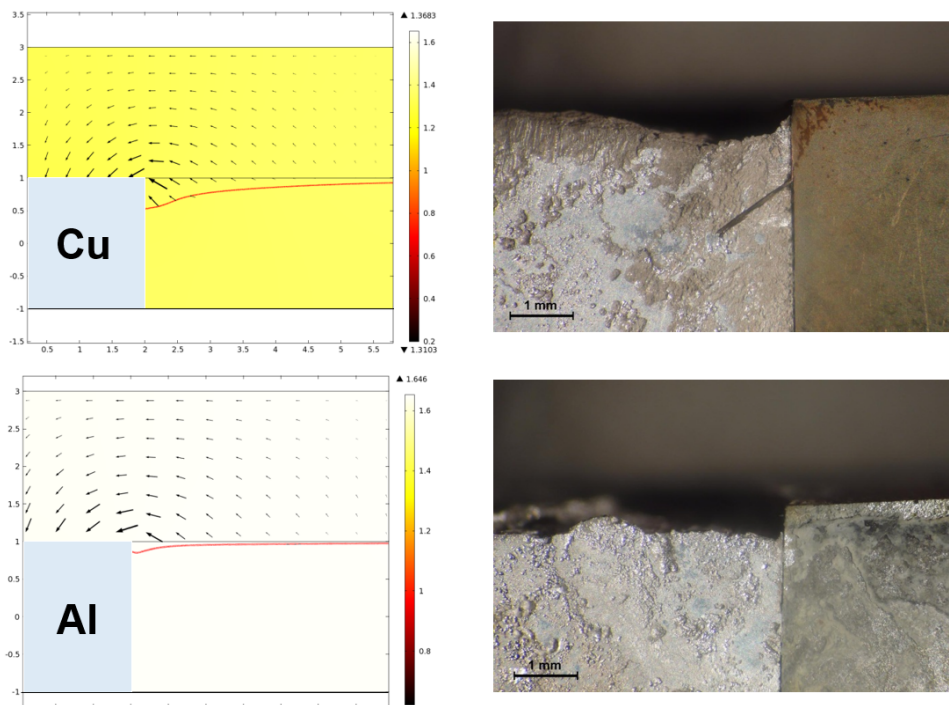
### 3 Discussion

Corrosion is a very challenging problem that involves many different fields of chemistry and physics. Particularly in this contribution authors have considered the galvanic corrosion of multi materials structures as reasonable example case. It is relevant for a number of applications for advanced constructions, lightweight structures, in automobile and airspace industry, medical devises and sensors. Thus, an ontological representation is even more encouraged.

Of course the shown numerical approach has a lot of limitations and its validity is limited. But for visualizing the transformation into an ontology based description of degradation with a more or less adequate accuracy, it is a very promising showcase. The influence of corrosion current densities and other material parameters on the process can be studied and the results computed can be utilized for laying out galvanic couples on a macroscopic scale. For more detailed computations the model has to be extended. Chemical conversion, passivation and transport processes in the electrolyte have to be considered, which requires further modelling activities.

From a mathematical point of view, this leads to a set of non linear equations and a huge amount of degrees of freedom in FEM computations. As a secondary effect of the used Level-set method the tuning of the reinitialization parameter  $\gamma_L$  is very difficult and should be considered and discussed separately. Thus, it is excluded from the ontological viewpoint. The next issue would be the step by step extension of the model by including migration, and chemical reaction kinetic, which is another story as well and out of scope of this work.

EMMO combined with a semantic framework will greatly simplify the integration of the needed range of models and data sources for correctly model corrosion phenomena. The domain specific ontologies presented in section 2.2 and illustrated in Figures 2 and 3 provide the input for the digitisation of galvanic corrosion phenomena. The introduced framework is semantically founded on the first outline of an EMMO-based galvanic corrosion ontology, describing the interrelations between real world objects, quantities and equations for the simple corrosion process.



**Figure 6:** Modelling results for the systems Mg-Cu and Mg-Al6060 in 3.5% sodium chloride after 19 hours immersion [20].

The approach indicated in this paper outlines a basic strategy for semantically describing any type of corrosion processes by development of new domain specific ontologies based on EMMO. This is especially important for the future development of accurate predictive corrosion models that include many different data from different scales and sources. Such models would require a sound semantic basis together with a generic data model to achieve robust through scale and through domain interoperability.

#### 4 Conclusions

The work shows the development of an ontological representation of a bimetallic galvanic corrosion problem at continuum level. The physio-chemical modelling of the problem has been transferred and ontologized excluding the interface tracking method. The final achievement's are:

- the EMMO has been used for ontological representation of bimetallic galvanic corrosion
- the general description of simulation of corrosion of multi materials structures is ready for application on similar computational problems
- and progress in the machine understandable way for the data exchange respective interoperability with respect to the high level of complexity and scaling has been achieved.

The approach might build the nuclei for future developments in computational corrosion engineering.

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